Towards a unification of the hierarchical reference theory and the self-consistent Ornstein-Zernike approximation

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The hierarchical reference theory and the self-consistent Ornstein-Zernike approximation are two liquid state theories that both furnish a largely satisfactory description of the critical region as well as phase coexistence and the equation of state in general. Furthermore, there are a number of similarities that suggest the possibility of a unification of both theories. As a first step towards this goal, we consider the problem of combining the lowest order γ expansion result for the incorporation of a Fourier component of the interaction with the requirement of consistency between internal and free energies, leaving aside the compressibility relation. For simplicity, we restrict ourselves to a simplified lattice gas that is expected to display the same qualitative behavior as more elaborate models. It turns out that the analytically tractable mean spherical approximation is a solution to this problem, as are several of its generalizations. Analysis of the characteristic equations shows the potential for a practical scheme and yields necessary conditions that any closure to the Ornstein-Zernike relation must fulfill for the consistency problem to be well posed and to have a unique differentiable solution. These criteria are expected to remain valid for more general discrete and continuous systems, even if consistency with the compressibility route is also enforced where possible explicit solutions will require numerical evaluations.

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I. INTRODUCTION

Both the self-consistent Ornstein-Zernike approximation (SCOZA) [1–3] and the hierarchical reference theory (HRT) [4] have been found to give very accurate results for fluids in thermal equilibrium. In particular, the respective nonlinear parabolic partial differential equations (PDEs) remain practical in the critical region, and their solution gives nonclassical, and partly Ising-like, critical indices. The PDEs themselves are derived by obtaining the equation of state in two independent ways and using thermodynamic consistency to fix a free parameter in the direct correlation function.

Although both approaches thus appear similar in a number of aspects, there are also marked differences: While both make use of the compressibility route to thermodynamics, SCOZA combines it with the energy route expression for the *internal* energy, whereas HRT, inspired by momentum-space renormalization group theory, relies on what might be called the fluctuation route to the *free* energy instead. In addition to the density ρ, the independent variables in the PDEs are, therefore, the inverse temperature $\beta=1/k_BT$ and the momentum-space cutoff Q, respectively. Starting from a reference system of known properties at vanishing β or at high Q, the attractive interaction is then turned on by gradually increasing its strength β (SCOZA) or by including its Fourier component of wave number Q at constant temperature until the full system is recovered in the limit $Q\rightarrow 0$.

In the present contribution, we want to investigate the possibility of combining both approaches by imposing thermodynamic consistency of the internal and free energies to obtain a differential formulation where both the strength of the interaction and its Fourier components are added successively. This introduces an additional constraint so that a further free parameter in the closure relation can be determined. In combination with the compressibility route, there are then two free parameters that may be used, e.g., to describe both the range and the amplitude of a contribution to the direct correlation function $c(r)$; it might then be possible to describe the long-ranged tail that $c(r)$ is known to develop in the critical region. However, our preliminary investigations point to several difficulties with this approach near phase coexistence. The crucial problem is the vastly different behavior of HRT and SCOZA in this part of the phase diagram: HRT has a solution at all densities, but spinodal and binodal coincide so that the two phases coexist at infinite compressibility; SCOZA, on the other hand, gives distinct binodal and spinodal curves, but does not have a solution inside the spinodal. At this point, it is unclear how these differences might be resolved by a simple modification of the direct correlation function. Even at temperatures above the critical one, $\beta<\beta_c$, where full self-consistency should be attainable, we will be confronted with differential equations that are of the first order in temperature and cutoff and of the second order in the density. The numerical solution of such a PDE can be demanding.

To simplify our task and gain some insight, we here consider a simplified lattice gas model, and we limit ourselves to combining the energy and fluctuation routes without using the compressibility at all. Then there is only one free parameter left that can be determined as a function of β and Q. After introducing our model and establishing the PDE (Sec. II), we show that consistency is achieved with the mean

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spherical approximation (MSA) and several generalizations of it (Sec. III). The solution can, therefore, be given in closed analytic form, but the price to pay for omission of the compressibility is the presence of the MSA features, viz., "irregular" isotherms in the critical region and lack of a well-defined critical behavior for the lattice gas case [5].

In general, an analytical solution is also unlikely to be known beforehand if other closures are used. In that case, one should seek the solution numerically by integrating the equations for the characteristic curves of the PDE. Especially when the compressibility relation not considered here is incorporated, a numerical solution will be required, and we expect the properties of the characteristics analyzed here to be useful more generally. After establishing relations and implications for more general situations (Sec. IV A), we once more turn to the MSA equations. It is found that the PDE admits a more general solution. Usually, the reference system also does not determine the solution at $Q=0$ for finite temperature due to the behavior of the characteristics (Sec. IV B). A generalized version of the PDE that does not make any assumptions on the form of the direct correlation function of the system at cutoff Q and inverse temperature β other than that it depends on some unknown parameter function $\lambda(\beta,Q)$ is considered in Sec. IV C. It is then found that these deficiencies can be peculiar to the specific closure employed. Based on these findings, we finally state and consider two complementary necessary conditions that a parametrization of the solution must fulfill for the consistency problem to be well posed and physically reasonable (Sec. V). These conditions have the advantage that they can be checked in the high temperature limit where significant simplifications are possible. They are also sufficiently general to apply to more realistic model systems, both discrete and continuous. In view of the relation between the properties of a nonlinear diffusion equation with two timelike parameters, on the one hand, and the first-order PDE we consider in the present contribution, on the other hand, the criteria found for the selection of suitable closure relations remain relevant even when the compressibility route to thermodynamics is also taken into account.

II. ADAPTATION OF THEORY AND MODEL

A. Model

In the interest of simplicity, we here consider only a lattice gas (or Ising model). The main advantage of this choice is that the core condition of vanishing pair distribution function $g(\vec{r})=h(\vec{r})+1$ affects only the single point at $r\equiv|\vec{r}|=0$. In addition, we assume a sufficiently long-ranged interaction so that the anisotropy imposed by the lattice structure can be neglected. Besides, an even more long-ranged tail appears in the interaction for intermediate Q, cf. Ref. [6] and Appendix D.1 of Ref. [7]. When following the HRT recipe of successively including Fourier components of the interaction, we will therefore do so without regard to the geometry of the Brillouin zone. In the same spirit, we will often speak of finite or infinite cutoffs and wave numbers instead of specifying the corresponding surfaces within the Brillouin zone explicitly [8,9], and we will also write Q_∞ for the maximum cutoff in the calculation. It is not expected that these simplifications should affect the results qualitatively. Furthermore, the main conclusions of Sec. V are manifestly independent of these assumptions.

The system is assumed to differ by a perturbing attractive interaction $-\psi(r)$, $\psi(r)>0$, from a reference system of known properties. In the lattice case, the simple hard core lattice gas can serve as the reference system. We can then use the inverse range γ of the attractive interaction as a perturbing parameter [10,11]. The zeroth-order contribution past the reference system is the mean-field attractive van der Waals term, corresponding to a pressure contribution of $-\frac{1}{2}\tilde{\psi}(0)\rho^2$, where a tilde indicates Fourier transformed quantities. In HRT, this corresponds to the zero-loop diagram [4]. To next order in γ, Hemmer found [10]

$$I=-\mathcal{C}\int \ln[1-\tilde{\mu}(k)\tilde{v}(k)]d^3k \propto \gamma^3 \tag{1}$$

with

$$\mathcal{C}=\frac{1}{2}\left(\frac{1}{2\pi}\right)^3,$$

$$\tilde{v}(k)=\beta\tilde{\psi}(k),$$

$$\beta=\frac{1}{k_BT}.$$

Here, I is $-\beta$ times the first-order contribution to the Helmholtz free energy per unit volume, k_B is Boltzmann's constant, and T is the temperature. The integral is to be extended over the Brillouin zone of the lattice that we here assume to be the simple cubic one for which $\mathcal{C}\int d^3k=\frac{1}{2}$.

The function $\mu(r)$ is related to the direct and total correlation functions, $c_0(r)$ and $h_0(r)$, of the reference system (indicated by the subscript 0) by

$$\tilde{\mu}(k)=\rho+\rho^2\tilde{h}_0(k)=\frac{\rho}{1-\rho\tilde{c}_0(k)}. \tag{2}$$

Here we have used the Ornstein-Zernike equation [12]

$$h(\vec{r})=c(\vec{r})+\rho\int h(\vec{r}-\vec{r}')c(\vec{r}')d^3r', \tag{3}$$

$$[1+\rho\tilde{h}(\vec{k})][1-\rho\tilde{c}(\vec{k})]=1.$$

In the lattice case, the integral is to be replaced with a sum.

In the limit $\gamma\to 0$, the function $\tilde{v}(k)$ usually reduces to a narrow peak of width γ at $k=0$. In this limit, the expression (1) is the exact correction to the mean-field result. In HRT, this corresponds to the infinite sum of one-loop diagrams [4].

The interaction $\tilde{v}(k)$ may also have a narrow peak of width γ at some other position, say, around $k=Q$. In r space, this corresponds to an infinitely weak but oscillating interaction with wave vector Q, and Eq. (1) is still exact in the limit $\gamma\to 0$. On the other hand, the renormalization procedure of HRT consists of adding narrow pieces of $\tilde{\psi}$ of width $-dQ$

>0 around shorter and shorter wave vectors Q at constant temperature. With $d^3k=4\pi k^2 dk$ and after division by dQ, Eq. (1) then becomes

$$\frac{\partial I}{\partial Q} = 4\pi \mathcal{C} Q^2 \ln[1-\tilde{\mu}(Q)\tilde{v}(Q)]. \tag{4}$$

This is also the first equation of the HRT hierarchy, provided μ is derived from the structure of the system at slightly higher cutoff $Q+|dQ|$ rather than from the reference system at Q_∞. Equation (4) is then formally exact, but it does not specify how $\tilde{\mu}$ changes as Fourier components of $\tilde{\psi}$ are added and the cutoff Q approaches zero. The formal answer to this problem is provided by the higher equations of the HRT hierarchy or some other expansion for μ. In practical applications of HRT, only Eq. (4) is used, and the evolution of $\tilde{\mu}$ is determined by introducing a free parameter into the correlation functions and fixing it by the requirement of consistency with the compressibility route.

One can also make a small change $d\beta>0$ in the inverse temperature and so effectively increase the interaction by an amount $-d\tilde{v}(k)=\tilde{\psi}(k)d\beta$. Taking the limit $d\beta\to 0$, we obtain

$$-\rho u_1 \equiv \frac{\partial I}{\partial \beta} = \mathcal{C}\int \frac{\tilde{\mu}(k)\tilde{\psi}(k)}{1-\tilde{\mu}(k)\tilde{v}(k)} d^3k. \tag{5}$$

The quantity u_1 is nothing but the configurational internal energy per particle beyond the zeroth-order mean-field term [besides a self-energy term $-\frac{1}{2}\psi(0)$ included here from the ρ term in μ] due to the structure or correlation function given by

$$\rho + \rho^2 \tilde{h}(k) = \frac{\tilde{\mu}(k)}{1-\tilde{\mu}(k)\tilde{v}(k)}. \tag{6}$$

With μ given by Eq. (2), this is exact only in the limit $\beta\to 0$, where the reference system is recovered. At higher β, it is only the first-order correction in γ for the long-range part of the pair correlation function [10,11]. To higher order, or at finite β, it is again not obvious how $\tilde{\mu}(k)$ changes with temperature. Here the SCOZA recipe is to introduce some parameter into the closure that is also fixed by the requirement of consistency with the compressibility route.

B. Specialization to lattice gas

In the case of a lattice gas or the Ising model, the above expressions are simpler. The direct correlation function for the reference system vanishes except at $r=0$ so that $\tilde{\mu}(k)$ is constant, $\tilde{\mu}(k)\equiv\mu$; and from the pressure $p=-\ln(1-\rho)/\beta$ of the hard core lattice gas serving as reference system, we obtain its value as

$$\mu = \tilde{\mu}(0) \equiv \left(\frac{1}{\rho}\frac{\partial \beta p}{\partial \rho}\right)^{-1} = \rho(1-\rho). \tag{7}$$

As a simple approximation, we can now replace the μ of Eq. (7) by some effective value μ_e that may be determined by imposing thermodynamic consistency between Eqs. (4) and (5); in general, μ_e will be a function of β and Q. Such a change may cause the core condition $h(0)=-1$ to be violated, which might be corrected by the introduction of an additional parameter. For the qualitative features we are interested in here, however, the core condition is not expected to be important, and we do not impose it below [13].

As Fourier components are added to the interaction at successively smaller cutoff Q, the interaction vanishes for $k<Q$ (except for the mean-field term at $k=0$), and the integral in Eq. (5) must be restricted to that part of the Brillouin zone where $k\geqslant Q$. Furthermore, a small change dQ in the cutoff corresponds to the addition of a weak long-ranged oscillating tail in r space, which also contributes at $r=0$. By setting the potential ψ at $r=0$ equal to zero, the corresponding unphysical contributions to the internal energy from within the hard core can be avoided approximately, and exactly in the limits $Q\to Q_\infty$ and $Q\to 0$, even when the core condition is not fulfilled. With these considerations and using spherical symmetry, Eqs. (4) and (5) become

$$\frac{\partial I}{\partial Q} = 4\pi \mathcal{C} Q^2 \ln[1-z\tilde{\psi}(Q)], \tag{8}$$

$$\frac{\partial I}{\partial \beta} = 4\pi \frac{\mathcal{C}}{\beta}\int_{k>Q} \frac{z\tilde{\psi}(k)}{1-z\tilde{\psi}(k)} k^2 dk$$

with

$$z = \mu_e \beta. \tag{9}$$

The above equations define a PDE for $\mu_e(\beta,Q)$ that is to be solved for all $Q\geqslant 0$, $\beta\geqslant 0$. In particular, the solution should give the structural and thermodynamic properties of the target system at $Q=0$ and all $\beta\geqslant 0$. In order to turn this into a well-defined problem, we also have to impose some boundary conditions. The natural choice is to demand that the reference system ($\mu_e=\mu$) should be recovered at $\beta=0$ and at $Q=Q_\infty$. Whether this is sufficient to determine the solution throughout the domain of the PDE, however, is not obvious and depends not only on the general consistency problem but also on the particular closure (the replacement of μ by μ_e) and the model chosen. We will return to this point in greater detail in Sec. IV.

At first sight, an analytic solution of the given equations is not obvious. But as we will show in Sec. III, the MSA solution and generalizations thereof indeed fulfill Eq. (8). On the other hand, the multiplicity of solutions found already indicates that the reference system boundary conditions (that are fulfilled in all cases) do not determine a unique solution, cf. Secs. III B and IV B.

III. MSA AND MSA-LIKE SOLUTIONS

A. Specific solution

The internal energy corresponding to the MSA correlation function can be integrated explicitly to yield the free energy by utilizing general MSA expressions [3]. The result is

$$I = -4\pi \mathcal{C} \int_{k>Q} \ln[1 - z\tilde{\psi}(k)]k^2 dk + J - \frac{1}{2}\ln(1+2J). \tag{10}$$

Here J is the integral appearing in Eq. (8),

$$J = \beta \frac{\partial I}{\partial \beta} = 4\pi \mathcal{C} \int_{k>Q} \frac{z\tilde{\psi}(k)}{1 - z\tilde{\psi}(k)} k^2 dk, \tag{11}$$

and the core condition determines z as

$$z = \frac{\beta\mu}{1+2J},$$

$$\mu = \rho(1-\rho). \tag{12}$$

By differentiation of Eq. (10) with respect to β and Q and using Eqs. (11) and (12), one finds that Eq. (8) is fulfilled.

The pair correlation function consistent with solution (10)–(12) is obtained from Eq. (6) by replacing $\tilde{\mu}(k)$ by $\mu_e = z/\beta$ and restricting the interaction to $k > Q$,

$$\rho + \rho^2 \tilde{h}(k) = \frac{z/\beta}{1 - z\tilde{\psi}(k)\Theta(k-Q)},$$

where Θ is Heaviside's function. Integrating the above over the Brillouin zone and using Eq. (11), one easily obtains

$$\rho + \rho^2 h(0) = \frac{z}{\beta}(1+2J) = \mu = \rho - \rho^2,$$

which shows that the core condition is fulfilled.

B. More general solution

The MSA solution just discussed is one specific solution of Eq. (8). A more general solution is suggested by previous work on the resummation of γ ordering for fluids [5,14], a procedure that transforms the serious divergence at the mean-field critical point into MSA-type criticality while maintaining self-consistency between the free and internal energies.

The generalized solution is then

$$I = -\mathcal{C} \int_{k>Q} \ln[1 - \mu_e \tilde{v}(k)] d^3k - \sum_{n=1}^{\infty} \frac{n}{n+1} A_n K^{n+1} \tag{13}$$

with $(z = \beta\mu_e)$

$$K = \frac{J}{\mu_e} = \mathcal{C} \int_{k>Q} \frac{\tilde{v}(k)}{1 - \mu_e \tilde{v}(k)} d^3k,$$

$$\mu_e = \mu + \sum_{n=1}^{\infty} A_n K^n. \tag{14}$$

Here the coefficients A_n are independent of β and Q and implicitly define μ_e. The A_n must be fixed by some suitable boundary conditions. These must be imposed at finite temperature and cutoff as K vanishes at $\beta = 0$ and at $Q = Q_\infty$. This shows that the reference system alone does not define a unique solution in the present case, cf. end of Sec. II B. The MSA solution (10) corresponds to the more general Eq. (13) with one specific choice of the A_n. By differentiation, it is easily verified that the above expression for I, like expression (10), solves both equations (8).

C. Wave number dependent direct correlation function

Solution (13) can easily be extended to some very specific k dependent μ_e and z. Replacing μ_e by $\tilde{\mu}_e(k)$, the Fourier transform of $\mu_e(r)$ that now extends to outside the core, we can write

$$\mu_e(r) = \mu\delta(\vec{r}) + \sum_{n=1}^{\infty} A_n(r) K(r)^n. \tag{15}$$

This corresponds to graphs with n parallel $K(r)$ bonds. $K(r)$ is the chain bond with renormalized hypervertex $\mu_e(r)$, and its Fourier transform is

$$\tilde{K}(k) = \frac{1}{2} \frac{\tilde{v}(k)}{1 - \tilde{\mu}_e(k)\tilde{v}(k)} \Theta(k-Q), \tag{16}$$

the generalization of the integrand in Eq. (14). Equation (13) now changes into

$$I = -\mathcal{C} \int_{k>Q} \ln[1 - \tilde{\mu}_e(k)\tilde{v}(k)] d^3k - \sum_{n=1}^{\infty} \frac{n}{n+1} \int A_n(r) K(r)^{n+1} d^3r, \tag{17}$$

and the PDE (8) becomes

$$\frac{\partial I}{\partial Q} = 4\pi \mathcal{C} Q^2 \ln[1 - \tilde{\mu}_e(Q)\tilde{v}(Q)],$$

$$\frac{\partial I}{\partial \beta} = 4\pi \frac{\mathcal{C}}{\beta} \int_{k>Q} \frac{\tilde{\mu}_e(k)\tilde{v}(k)}{1 - \tilde{\mu}_e(k)\tilde{v}(k)} k^2 dk. \tag{18}$$

As mentioned at the beginning of Sec. II, we neglect the lattice structure for our simplified model. Of course, for a true lattice system $\delta(\vec{r})$ in Eq. (15) should be a Kronecker δ at the origin, and the spatial integral in Eq. (17) should be a lattice sum.

The above solution is suggested by the work of Høye and Olaussen on the two-dimensional Coulomb gas where the well-known Kosterlitz-Thouless phase transition was evaluated on the basis of a graph expansion [15,16].

Again it can be verified by differentiation that expression (17) solves Eq. (18) in the same way expression (13) solves Eq. (8). To do so, one also needs the identity

$$\int f(r) g(r) d^3r = 2\mathcal{C} \int \tilde{f}(k) \tilde{g}(k) d^3k.$$

IV. CHARACTERISTICS OF THE DIFFERENTIAL EQUATIONS

A. First-order PDEs and characteristics

So far, we have only considered specific cases where exact solutions are known. In more general situations, the characteristics can be used. These are curves along which the PDE of interest is equivalent to a set of coupled ordinary differential equations. In the case of a first-order PDE considered here, the solution can always be obtained by integrating the characteristic equations, starting from some point where the solution is known from some boundary condition. This is exact and does not incur any loss of information or generality, and it generates a solution that is necessarily differentiable along the characteristic.[1]

Clearly, the solution along a full characteristic depends on only a single point at the boundary. This reflects the well-known capacity of first-order PDEs for discontinuous solutions. Another way in which discontinuities may arise is from crossings of characteristic curves: the solution then becomes multivalued, and a resolution restoring uniqueness naturally leads to a *shock* where the solution is no longer differentiable. Other complementary difficulties that one may encounter are *rarefactions*, i.e., regions not entered by any characteristics. In this case, the solution is not defined there, although there is often a natural weak formulation of the PDE that can be used instead.

In the present work, we consider consistency of the free and internal energies at inverse temperature β when gradually turning on Fourier components of the interaction at ever smaller wavelength Q. Neither shocks nor rarefactions are expected on physical grounds, i.e., the solution of the PDE should both exist and be differentiable for $\beta \geqslant 0$ and $Q \geqslant 0$, at least away from the critical point and outside the spinodal. Using subscripts to denote partial derivatives, a PDE such as Eq. (8) is naturally written in the form

$$\Psi_x(x,y) = X(x,y;\lambda(x,y)),$$

$$\Psi_y(x,y) = Y(x,y;\lambda(x,y)).$$

Here λ is an unknown function of the independent variables x and y. As we will see, these equations imply a first-order PDE for λ, the solution of which also gives the quantity of interest, Ψ.

The above relations are not in the standard form $F(x,y,\Psi,\Psi_x,\Psi_y)=0$ for a first-order PDE. The usual expressions for the characteristics are thus not directly applicable. One option is to invert, say, $X(x,y;\lambda)$ with respect to λ. The resulting expression for λ can then be inserted into the equation for Ψ_y, allowing the usual equations for the characteristics of nonlinear PDEs to be used. The disadvantage of this approach is that it leads to highly involved expressions.

A simpler way of obtaining the characteristic equations is by cross differentiation of the PDE. Setting $\Psi_{xy}=\Psi_{yx}$ gives

$$X_y + X_\lambda \lambda_y = Y_x + Y_\lambda \lambda_x.$$

This is a quasilinear first-order PDE for λ of the form $a\lambda_x + b\lambda_y = c$, for which the characteristic equations are $dx/a = dy/b = d\lambda/c$. In combination with the PDE for Ψ itself, we immediately find the set of characteristic equations,

$$\frac{dx}{Y_\lambda} = -\frac{dy}{X_\lambda} = \frac{d\lambda}{X_y - Y_x} = \frac{d\Psi_x}{X_x Y_\lambda - X_\lambda Y_x} = \frac{d\Psi_y}{X_y Y_\lambda - X_\lambda Y_y} = \frac{d\Psi}{\Psi_x Y_\lambda - \Psi_y X_\lambda}. \tag{19}$$

At every point (x,y) along a characteristic, the ratios of these differentials determine the direction of its tangent in the (x,y) plane as well as the corresponding changes in λ, Ψ_x, Ψ_y, and Ψ along the curve. The equations can easily be solved numerically, e.g., by predictor-corrector methods. In the present contribution, however, we will not concern ourselves with numerical evaluations. Suffice it to say that a straightforward implementation of these equations may require very small step sizes, and that an accurate evaluation of the direction of the tangent may be difficult, in the vicinity of points where $X_\lambda = Y_\lambda = 0$; discretizations treating X_λ and Y_λ as perturbations relative to, e.g., $X_{\lambda\lambda}$ and $Y_{\lambda\lambda}$ can be employed there.

An important consequence of the equations given above concerns the orientation of the characteristics in the (x,y) plane at special points: Evidently, wherever $X_\lambda = 0$ and $Y_\lambda \neq 0$, the tangent is parallel to the x axis, i.e., $y=$const. By locating the zeros of the λ derivatives of the right-hand sides of the PDE, we can thus rapidly gain a qualitative overview of the geometry of the field of characteristic curves. Considering the path of integration in the (β,Q) plane, pure HRT obviously corresponds to characteristics at constant temperature, and pure SCOZA to those at constant cutoff. We will, therefore, refer to these directions as HRT-like or SCOZA-like, respectively.

For the PDE (8), this directly implies the presence of a SCOZA-like characteristic for $Q=Q_\infty$ and of an HRT-like one at $\beta=0$; together they cover that part of the domain where the solution corresponds to the reference system. In addition, characteristics can have an HRT-like tangent only in points where $\tilde{\psi}(Q)=0$, and by considering all orders in the free parameter, it is seen that all characteristics have HRT-like tangents whenever $\tilde{\psi}(Q)$ vanishes at $Q<Q_\infty$ and $\beta>0$.

It is worth stressing that the significance of the characteristic curves extends not only to the numerics: In addition to the significance of shocks and rarefactions, neither of which are expected in the present setting, the PDE is well-posed only when the characteristics establish a one-to-one mapping from the points at $Q=0$ to a subset of the boundary where the solution and its first derivatives are known. It is then possible to start out from the boundary and to integrate along those curves until the final solution is obtained at vanishing Q.

[1] See textbooks on PDEs such as, e.g., Leon Lapidus, George F. Pinder, *Numerical Solution of Partial Differential Equations in Science and Engineering* (Wiley, New York, 1982).

But even for a stable and well-posed PDE, the characteristics are of vital importance for the numerics, independently of whether the discretization explicitly makes use of them: According to the Courant-Friedrichs-Levy criterion [17], any numerical scheme that does not adhere to the flow of information represented by the characteristics cannot be stable.

B. Characteristics for the MSA-like case

Even without prior knowledge of the generalized MSA solutions presented in Sec. III, some progress can be made towards the solution of Eq. (8) by using the characteristics of the PDE alone. In the MSA case, the unknown parameter function $\lambda(x,y)$ of the preceding section corresponds to $\mu_e(\beta,Q)$.

1. Explicit results

The situation is most transparent when considering only the limit of small z where Eq. (8) becomes to order $O(z^2)$

$$\frac{\partial I}{\partial Q} = F'(Q)z,$$

$$\frac{\partial I}{\partial \beta} = \frac{1}{\beta}F(Q)z,$$

$$F(Q) = 4\pi\mathcal{C}\int_{k>Q}\tilde{\psi}(k)k^2dk.$$

After elimination of z and a change of variables from Q to F, we obtain a PDE for I as a function of F and β, viz.,

$$F\frac{\partial I}{\partial F} - \beta\frac{\partial I}{\partial \beta} = 0. \tag{20}$$

The characteristics of this linear first-order PDE are determined by

$$\frac{dF}{F} = -\frac{d\beta}{\beta} = \frac{dI}{0},$$

which is trivially integrated to

$$F\beta = \text{const},$$

$$I = \text{const}. \tag{21}$$

The general solution of the PDE is then any functional relation between the two constants of integration above, i.e., I can only depend on $F(Q)\beta$,

$$I = I[F(Q)\beta]. \tag{22}$$

This is also the case for the solutions of Eq. (8) given in Sec. III to first order in z. Equations (10) and (13) are both recovered to first order in z by imposing $I=0$ for $\beta=0$.

The situation is only slightly more complicated when terms of higher order in z are not neglected in Eq. (8). We can obtain an equivalent PDE by cross-differentiating the Eqs. (8) and eliminating $\partial^2 I/\partial Q\,\partial\beta$, as was done in Sec. IV A. From the relations (8) and the definition (11), we find

$$-4\pi\mathcal{C}Q^2\frac{\tilde{\psi}(Q)}{1-z\tilde{\psi}(Q)}\left(\frac{\partial z}{\partial \beta}\right)_Q = \frac{1}{\beta}\left(\frac{\partial J}{\partial Q}\right)_\beta$$

or

$$\frac{1}{z}\left(\frac{\partial J}{\partial Q}\right)_z\left(\frac{\partial z}{\partial \beta}\right)_Q = \frac{1}{\beta}\left[\left(\frac{\partial J}{\partial Q}\right)_z + \left(\frac{\partial J}{\partial z}\right)_Q\left(\frac{\partial z}{\partial Q}\right)_\beta\right].$$

Rearranging this yields a quasilinear first-order PDE for $z(\beta,Q)$, viz.,

$$\frac{1}{z}\left(\frac{\partial J}{\partial Q}\right)_z\left(\frac{\partial z}{\partial \beta}\right)_Q - \frac{1}{\beta}\left(\frac{\partial J}{\partial z}\right)_Q\left(\frac{\partial z}{\partial Q}\right)_\beta = \frac{1}{\beta}\left(\frac{\partial J}{\partial Q}\right)_z. \tag{23}$$

As in Eq. (19), the equation for the characteristics becomes

$$\frac{d\beta}{\frac{1}{z}\left(\frac{\partial J}{\partial Q}\right)_z} = -\frac{dQ}{\frac{1}{\beta}\left(\frac{\partial J}{\partial z}\right)_Q} = \frac{dz}{\frac{1}{\beta}\left(\frac{\partial J}{\partial Q}\right)_z}.$$

This immediately gives $zd\beta=\beta dz$ so that

$$\mu_e = \frac{z}{\beta} = \text{const} \tag{24}$$

along the characteristics. The relation between dQ and dz merely reproduces the total differential of $J(Q,z)$, implying

$$J = \text{const}. \tag{25}$$

The general solution of Eq. (23) is then any functional relation between z/β and J. It is easily seen that this is consistent with the expression for μ_e given in Sec. III B; constant $\mu_e = z/\beta$ and $J=\mu_e K$ also imply constant K so that μ_e must be a function of K only, just as indicated in Eq. (14).

2. Implications of the characteristics

As pointed out in Sec. IV A, the PDE has a well-defined and unique solution only when the characteristics establish a one-to-one mapping of the target system at various temperatures $(Q=0,\beta>0)$ onto points where the solution is known from a suitable boundary condition. For the PDE (8), the most natural condition to impose is provided by the reference system, the properties of which must be recovered both at $\beta=0$ and at $Q=Q_\infty$. On the other hand, among the general features of this type of PDE in Sec. IV A, we found that these boundaries also coincide with the location of two characteristics. Consequently, there can be no characteristic connecting a point on one of those boundaries with a target system at nonzero β. At first sight, this seems to render the reference system useless as a boundary condition. Indeed, for the MSA-like solution considered here, Eq. (25) shows that the reference system only determines the solution where $J=0$, i.e., on the boundary itself.

However, this is not necessarily a severe problem as $I(\beta,Q)$ is expected to be continuous everywhere, including at the boundaries. The reference system therefore still provides a valid description for systems removed from the boundaries by a very small amount, where the characteristic may have a direction different from that of the boundary and may actually lead to the target system at nonzero β. In this

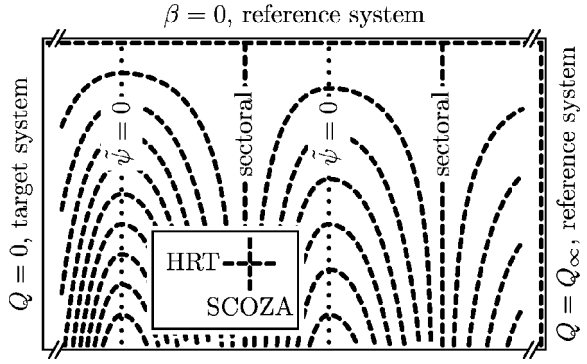

FIG. 1. Sketch of the characteristics for the MSA-like solution to first order in z: Dashed lines show the characteristics whereas the dotted lines give the locus of $\tilde{\psi}=0$ where the characteristics have an HRT-like tangent. Sectorals are marked, as are the limits where the target and reference systems are recovered and the characteristics at $\beta=0$ and $Q=Q_\infty$. The inset gives the directions corresponding to pure HRT and SCOZA, respectively. There may or may not be a sectoral at $Q=0$, depending on whether $\psi(0)$ vanishes or not. As expected, there are no rarefactions, nor are there crossings of characteristics except at $\beta=0$.

case, the reference system may still be used as a boundary condition; an example where even an infinitesimal separation is sufficient will be given in Sec. IV C. This situation echos the one found in continuum fluids in HRT: For those systems it has always been necessary to start numerical integration of the equations at some large but finite cutoff Q_∞, typically on the order of $10^2/\sigma$, where σ is a length characteristic of the repulsive hard core reference interaction [7,18]. Neglecting the contribution of very high wave number fluctuations has never been a practical problem, nor is this expected to be the case here.

In order to investigate whether a purely numerical solution is possible for the MSA-like equations considered in Sec. IV B 1, let us consider the results obtained by expanding the PDE to first order in z, valid in the limit of high temperature. For the moment assuming $\psi(0)\neq 0$, there are potentials such that $F(Q)$ vanishes only for $Q=Q_\infty$. In this case, I may be prescribed for all β at some fixed $Q<Q_\infty$, the characteristics stay at finite β at all lower Q, and the solution at $Q=0$ can be obtained at every temperature.

On the other hand, F will be an oscillating function of Q for many short-ranged interactions, especially when the core condition is approximately taken into account by demanding $\psi(0)=0$, cf. Sec. II B. At high temperature, the field of characteristics then qualitatively looks as sketched in Fig. 1.

An important but unfortunate feature of that sketch are the SCOZA-like characteristics starting at $\beta=0$, whenever $F(Q)=0$: These *sectorals*, as we shall call any characteristic the tangent of which at $\beta=0+$ has a nonvanishing component in the β direction, divide the domain of the PDE into a sequence of *sectors*. As there are no characteristics that cross from one sector into a neighboring one, no information is transmitted across a sectoral, and the solutions in adjacent sectors are largely independent. The special significance of these characteristics comes from the fact that the solution for the target system ($Q=0$) is determined by a boundary condition only when both lie in the same sector.[2] Furthermore, the SCOZA-like orientation of a sectoral means that the solution along it cannot depend on Fourier components of the interaction at other Q in the high temperature limit.

For lower temperatures, we have to use the full solution. According to Eq. (25), the sectorals are the curves where $J=0$. As we further follow along them, their direction will no longer be SCOZA-like, and the solution along them will depend on a range of Fourier components. Still, one of the properties that can be inferred from the temperature dependence of the J integral is that no sectoral can ever get to $Q=0$ at nonzero β, since $J>0$ there.

This feature of the sectorals also conforms to our expectation for a realistic closure relation. For suppose that some sectoral starts at $\beta=0$ and intermediate $Q>0$ and finally reaches $Q=0$ at $\beta=\beta_s>0$. Clearly it can only cover a finite Q interval so that the solution at $Q=0$, $\beta=\beta_s$ cannot depend on Fourier components of the interaction outside this Q range. Absence of crossings of characteristics immediately implies that the same is also true for the properties of the target system at all $\beta<\beta_s$. This situation is clearly unacceptable except for $\beta_s=0$.

Another consequence of the temperature dependence of the J integral is the absence of sectorals at given Q for β above some Q-dependent limit; this limit is generally higher for larger Q. There are then only a few classes of admissible configurations of the characteristics in the (β,Q) plane. Most likely, the sectorals are either driven towards infinite β at ever higher Q, or pairs of neighboring sectorals join at their shared maximum β and so form loops; in the latter case, the maximum β occurs at a cutoff where $\tilde{\psi}(Q)=0$. Unfortunately, both of these possibilities are highly problematic: If sectorals stay separate but go towards infinite β and Q, there can be no finite temperature characteristic connecting a point close to the reference boundary with one at $Q=0$, $\beta>0$. If, on the other hand, sectorals form loops, the same problem will arise at high cutoff. In addition, loops demarcate regions that are not entered by characteristics, implying rarefactions for $\beta\to 0$.

C. Connecting reference and target systems

The properties of the characteristics just inferred for the MSA case studied here are certainly unexpected and disappointing because they do not allow us to go from the reference system at $Q=Q_\infty$ to the target system at vanishing Q. We now show that this is not so much a general defect of the combination of the energy and fluctuation routes, but a consequence of the specific parametrization of the consistency problem. To see this, let us consider an arbitrary closure relation to the Ornstein-Zernike relation (3), giving the correlation functions c and $g=h+1$ for any combination of Q, β, and some unknown parameter $\lambda(\beta,Q)$. For example, the

[2]This is not the case for the analytical solution of Sec. III B as constancy of the A_n is built right into Eqs. (13) and (14). It is then possible to use a boundary condition to determine the A_n in one sector and to use their values in another one.

MSA case considered so far corresponds to the direct correlation function

$$\tilde{c} = \frac{1}{\rho} - \frac{\lambda}{\mu} + \Theta(k-Q)\tilde{v}, \quad \lambda = \frac{\mu}{\mu_e}.$$

Another simple possibility is

$$\tilde{c} = \frac{1}{\rho} - \frac{1}{\mu} + [\lambda + \Theta(k-Q)]\tilde{v}.$$

This is suggested by the usual HRT recipe for the continuum case when the core condition is not explicitly taken into account [4,13,19,20].

In this more general situation with unspecified closure, the correct PDE is given by the earlier relations (4) and (5), except that the energy integral must be restricted to $k>Q$ and that $\tilde{\mu}$ now depends not only on k, β, and Q but also on the free parameter function $\lambda(\beta,Q)$. As noted in Sec. II B, the core condition must be fulfilled at every Q if the simple expression (5) is used or else there will be an unphysical contribution to the internal energy from $r=0$.

In general, $\tilde{\mu}(k)$ is continuous at $k=Q$, whereas the direct correlation function $\tilde{c}(k)$ of the system corresponding to cutoff Q has a discontinuity of height $\tilde{v}(Q)$ there. The relation between $\tilde{\mu}$ and $\tilde{c}$ is obtained from Eq. (6) by restricting the interaction to $k<Q$ again,

$$\rho + \rho^2\tilde{h}(k) = \frac{\tilde{\mu}(k)}{1 - \tilde{\mu}(k)\tilde{v}(k)\Theta(k-Q)}$$

or

$$\tilde{c}(k) = \frac{1}{\rho} - \frac{1}{\tilde{\mu}(k)} + \tilde{v}(k)\Theta(k-Q).$$

Without further specifying the closure, a number of properties of the characteristics can easily be deduced: As the unknown parameter λ enters Eq. (8) only through $\tilde{c}$, we immediately see that the HRT-like characteristic at $\beta=0$, the SCOZA-like characteristic at $Q=Q_\infty$, and the HRT-like direction of the tangents of all characteristics whenever $\tilde{\psi}(Q)=0$ remain in this general setting.

Both for discrete and continuous systems, the question is then whether these boundary conditions at large but finite cutoff determine the solution at $Q=0$. We have just seen that this is usually not the case for the PDE (8). As pointed out before, the boundary conditions must be imposed in the same sector where the target system's properties are to be recovered. For this to be possible and the reference system to provide a valid initial condition for the integration of the characteristic equations, it is necessary but not sufficient that there be no sectorals above the initial value of Q, nor between the boundary condition and the target system.

We then have to consider the existence and distribution of sectorals once again, which crucially depends on the precise way in which the free parameter enters the equations. As an example, let $\tilde{c}_{\mathrm{ex}}(k;\beta,Q)$ and $\tilde{h}_{\mathrm{ex}}(k;\beta,Q)$ be the direct and total correlation functions for any solution of the PDE such as, e.g., any of those presented in Sec. III. Inserting either of the two sample closures

$$\tilde{c}(k;\beta,Q,\lambda) = \tilde{c}_{\mathrm{ex}}(k;\beta,Q) + \lambda[f_1(k) - f_1(Q)] \tag{26}$$

and

$$\tilde{h}(k;\beta,Q,\lambda) = \tilde{h}_{\mathrm{ex}}(k;\beta,Q) + \frac{\lambda}{\tilde{\psi}(k)}[f_2(2k-Q-Q_\infty) - f_2(Q+Q_\infty-2k)] \tag{27}$$

(with largely arbitrary functions f_i) into Eqs. (4) and (5) yields a PDE for $\lambda(\beta,Q)$. Obviously, the solution is $\lambda=0$ for all β and Q in either case so that the original solution is reproduced, $\tilde{c}=\tilde{c}_{\mathrm{ex}}$ and $\tilde{h}=\tilde{h}_{\mathrm{ex}}$. According to Eq. (19), however, for Eq. (26) the characteristics are HRT-like everywhere ($d\beta=0$, so that there are no sectorals at all), whereas they are all SCOZA-like everywhere for Eq. (27) ($dQ=0$, so that there are infinitely many sectorals). Artificial and impractical as these examples are, they demonstrate that two different parametrizations of one and the same solution may have vastly different consequences, and Eq. (26) in particular shows the potential for a closure free of sectorals.

V. NECESSARY CONDITIONS FOR WELL-POSEDNESS

We therefore conclude that the difficulties encountered in the case studied in Sec. IV B are not inherent in the consistency problem, but merely a consequence of the particular MSA-like closure used. The results of our investigation thus leave open the possibility of a practical scheme, drawing our attention to the precise parametrization of the solution.

While formulation of practical closure relations to be used together with Eq. (8) lies outside the scope of the present contribution, we can give two general nontrivial conditions that an ansatz for $\tilde{c}$ must fulfill for the consistency problem to be well-posed and for a unique and differentiable solution to exist at all $\beta\geqslant 0$, $Q\geqslant 0$: (i) There must not be any sectorals in the sense of the definition given in Sec. IV C; and (ii) any characteristic passing through a state with $0<\beta=O(\epsilon)$ at some $Q>0$ must remain at a strictly positive inverse temperature β of order $O(\epsilon)$ at lower cutoffs. While the former is quite obvious in the light of Sec. IV B 2, the second condition has not featured prominently so far: It merely expresses the absence of rarefactions and shocks at infinite temperature, and for the MSA case as treated here these already imply sectorals, cf. Sec. IV B 2.

Both of these criteria share the advantage of involving only the limit $\beta\to 0$. They can therefore be checked by low order expansions in β, which vastly simplifies analysis of the suitability of some specific closure relation in the context of Eq. (8).

Another important property is that both of them are quite general: Not only do they apply to discrete and continuous systems equally, they are also independent of the particular simplifications that we chose to make in the present contribution. In particular, a closure may give a solution where the core condition is violated, leading to a spurious contribution to the energy integral of Eq. (5). In this case, more care should be exercised when evaluating the internal energy, leading to a modification of the PDE [21]. The two conditions set out above, however, remain equally valid nevertheless.

A careful study of the characteristics, especially of their compliance with the criteria just set out, is thus of vital importance for the consistency problem involving the energy and fluctuation routes only. The results of such an analysis are also highly relevant when the compressibility route enters the picture. Suppose that $I(\beta,Q,\rho)$ is the solution of the more elaborate PDE so obtained. By evaluating $\partial^2 I/\partial\rho^2$ and inserting this into the compressibility sum rule used in both pure HRT and pure SCOZA, we arrive at a constraint involving both of the two unknown parameters in the parametrization of the solution at any (β,Q,ρ). Restriction to fixed density in the spirit of the line method then again leads to a problem of the type considered here, and the properties of the characteristics of this restricted problem must be taken into account when solving, or discretizing, the PDE. In particular, the density dependence of the directions of the characteristics in the (β,Q) plane are then of prime importance for the numerical tractability of the PDE by finite difference methods. At this point, however, it is not clear how to handle the situation where β does not increase monotonously along the characteristics, nor do we know whether that case actually occurs with physically plausible closure relations and interactions.

In summary, in the present contribution we have introduced and started to tackle the problem of unifying SCOZA and HRT, pointing out the importance of the condition of consistency between free and internal energies and for the time being setting aside consistency with the isothermal compressibility. A simplified lattice model system allowed us to derive a suitable PDE and to study its known analytical solutions of MSA type. In general, however, we cannot assume an analytical solution to be available, and we therefore then turned to the problem of solving the PDE without previous knowledge of the exact solution, especially with a view towards a numerical implementation: This highlighted the importance of the characteristic curves of the PDE, and we have studied some of their properties and consequences.

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