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Structure and thermodynamics of a classical hard-sphere fluid: A self-consistent theory

G. Giunta and C. Caccamo
Istituto di Fisica Teorica, Università degli Studi di Messina, I-98100 Messina, Italy

P. V. Giaquinta
Istituto di Fisica Teorica, Università degli Studi di Messina, I-98100 Messina, Italy
and International School for Advanced Studies, I-34104 Trieste, Italy

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A self-contained Ornstein-Zernike approximation scheme for calculating the structural and thermodynamic properties of a hard-sphere fluid is proposed within a simple functional *ansatz* for the tail function $d(r)$ and without resorting to any *a priori* availability of numerical simulation data. This is done by adopting a complete thermodynamic closure on the solution of the model by means of internal-consistency constraints which arise from demanding, through the knowledge of the structural correlations in the fluid, a unique estimate for the equation of state whatever route is followed for its evaluation.

I. INTRODUCTION

The comprehension of the role played by hard spheres as a primitive "reference system" for describing the structural properties of dense classical liquids may well be considered as one of the most important achievements in this field over the past two decades. A unique feature of this model system, which arises from the postulated highly singular law of interaction between the particles, is that its excess free energy is entirely entropic.[1] Moreover, as clearly appears from numerical simulation experiments, the excluded volume effects which are produced by the hard-sphere potential already yield a close-fitting picture of the short-range structure in a monatomic liquid at moderately high densities. Such peculiarities, together with the recognition of the different roles played by the repulsive and attractive parts of the interaction potential, are at the very root of the successful use of hard spheres as the proper reference system on which to engraft a perturbative theory of real dense fluids.

The absence of an exact solution of the model for space dimensionality higher than one makes it necessary and rewarding to set up semianalytical approximation schemes for describing at a quantitative level both the structure and the thermodynamics of this basic system. Most liquid-state theories usually start from a functional ansatz (which can be variously justified) on some relevant structural property of the fluid.

Recently, a very effective methodology for constructing at least a partially self-contained theory has become that of resorting to the requirement of thermodynamic consistency in order to evaluate the free parameters which enter the approximation scheme.[1–6] Identity between the pressures as calculated from the virial theorem and from the fluctuation-theory expression for the isothermal compressibility is demanded as the proper thermodynamic constraint on the solution of integral equations for the pair distribution function $g(r)$. Other distinct *ad hoc* conditions may also be introduced to fix the residual independent parameters of the theory.

Within the context of this general philosophy, it is the purpose of this paper to point out that the existence of further explicit statistical-mechanical routes to the thermodynamics of a hard-sphere system can be conveniently used to ensure a more complete internal consistency between all the possible structural estimates of the thermodynamic properties of the fluid. This is done after a well-defined ansatz for a typical structural function of the system is postulated. A totally self-contained scheme is thus set up, requiring no external information such as that provided by computer simulation experiments.

The starting point is obviously the calculation of the radial distribution function which fully describes the static structure of a classical fluid on a "pair level." In this respect, it is well known that one of the most fertile approaches to the explicit evaluation of this function for a variety of model systems is provided by the Ornstein-Zernike (OZ) equation which relates $g(r)$ to an auxiliary function $c(r)$, known as the direct correlation function:[7,8]

$$h(r)=c(r)+\rho\int d\mathbf{r}' h(|\mathbf{r}-\mathbf{r}'|)c(r') , \qquad (1)$$

where $h(r)=g(r)-1$ is the total correlation function, and $\rho=N/V$ is the particle number density. The OZ route to a model calculation of pair correlations in the fluid requires the construction of suitable approximations for $c(r)$. A deeper insight into the mathematical structure of this function can be achieved if one resorts to its formal expression in terms of the "tail" function $d(r)$:

$$c(r)=f(r)y(r)+d(r) , \qquad (2)$$

where $f(r)=\exp[-\beta u(r)]-1$ is the Mayer function, $\beta=1/k_BT$ is the inverse temperature, $y(r)=\exp[\beta u(r)]g(r)$ is the cavity distribution function, and $u(r)$ is the interatomic pair potential. The function $d(r)$ is independently defined in diagrammatic language as the set of those connected graphs which are free from bridge points and lack direct bonds between the root points.[8] For hard spheres, one has exactly

$$g(r)=\begin{cases}0, & r<\sigma & \text{(3a)}\\ y(r), & r>\sigma & \text{(3b)}\end{cases}$$

where σ is the hard-core diameter, and

$$c(r)=\begin{cases}d(r)-y(r), & r<\sigma & \text{(4a)}\\ d(r), & r>\sigma & \text{(4b)}\end{cases}$$

which imply a finite discontinuity of both $g(r)$ and $c(r)$ across $r=\sigma$, while the functions $y(r)$ and $d(r)$ are everywhere continuous for $r\geq 0$.[9] Furthermore, a remarkable feature of the OZ equation for systems with a hard-sphere interaction is that $g(r)$ is known to be zero inside the hard core, so that only $c(r)$, or equivalently—by Eq. (4b)—$d(r)$, outside is required for the evaluation of the pair distribution function over the whole range of r.[10]

However, this is not tantamount to a complete solution of the model. In fact, if one is also interested in the behavior of $y(r)$ in the interior of the core, then the tail function $d(r)$ needs to be independently specified also for $r<\sigma$.[10]

There are several reasons why one would like to obtain reliable theoretical information on $y(r)$ over the whole range of r. This function is intrinsically relevant since its logarithm yields the potential of mean force between two particles in the fluid.[8] In the limit of $r\to 0$, the cavity distribution function is also asymptotically related to the thermodynamics of the system, and in the case of hard spheres one explicitly finds[11,12]

$$\ln y(r)=\beta\mu^{\mathrm{ex}}-\tfrac{3}{2}(\beta P^{\mathrm{ex}}/\rho)r/\sigma+\cdots\ , \tag{5}$$

where μ^{ex} and P^{ex} are the excess parts of the chemical potential and pressure, respectively. In addition, some widely used perturbation theories for the structure of fluids with a soft-core potential require the knowledge of the hard-sphere cavity distribution function inside the core.[13] Furthermore, $y(r)$ is directly available via the free energy of the fluid excluded from two overlapping spheres.[14]

The layout of the paper is as follows: In Sec. II we introduce the basic assumption on the functional behavior of the tail function $d(r)$. The proposed thermodynamic closure of the theory is discussed in Sec. III. A short description of the numerical procedure adopted for the solution of the model is given in Sec. IV. The results are presented in Sec. V and compared with those obtained by suitable accurate parametrizations of the numerical simulation experiments. Section VI is finally devoted to concluding remarks with an emphasis on the possible improvements of the present scheme.

II. THE BASIC ASSUMPTION ON THE STRUCTURE OF THE MODEL

In view of setting up a self-contained Ornstein-Zernike approximation scheme with a fully consistent thermodynamic closure, we now introduce our key approximation for the hard-sphere tail function:

$$d(r)=\begin{cases}\exp\left[\sum_{n=0}^{3}\alpha_n(r/\sigma-1)^n\right], & r<\sigma\\ \dfrac{K}{r/\sigma}\exp[-z(r/\sigma-1)], & r>\sigma\ .\end{cases} \tag{6}$$

The function $d(r)$ is positive definite in real space. It is also positive definite in the Fourier sense for the relevant set of parameters α_n, K, and z. Inside the core our functional ansatz for $d(r)$ is equivalent to the one Henderson and Grundke (HG) proposed for the whole range of r in order to construct a simple but reliable semiempirical parametrization of the structural properties of a hard-sphere fluid.[15] This goal was actually achieved in a way which is conceptually analogous to what Verlet and Weis (VW) had previously done for the radial distribution function,[16] viz., by fitting the guessed $d(r)$ to external data for the thermodynamics and structure of the fluid essentially provided by computer simulation experiments. However, this procedure, even if very effective and successful, is intrinsically unable to yield independent theoretical predictions.

The reason for assuming a Yukawa decay for $d(r)$ outside the core is closely related to the solution of the OZ integral equation. In fact, our approximation is clearly less flexible and thus in principle less accurate than the original HG ansatz, but has the distinct advantage of allowing an analytical solution of the model, leading to closed-form results for both the thermodynamic properties and the direct correlation function inside the core.[17] It is evident that such an assumption on the functional behavior of $d(r)$ does not contain what presumably appears to be a relevant feature of the direct correlation function for hard spheres at high densities, namely the occurrence of damped oscillations outside the core.[15,18] Nevertheless, a good overall description of the structure and thermodynamics of the fluid is already possible on a quantitative basis even in this simplified picture. These conclusions are anticipated by previous results obtained by Waisman[17] and Grundke and Henderson[19] who showed, with two different procedures, that, after fitting the parameters K and z to the "exact" values of the pressure and compressibility, the Yukawa form for $c(r)$ is able to reproduce the experimental pair distribution function for hard spheres with a remarkable accuracy.

The analytical solution of the OZ equation supplemented by the "core condition" on $g(r)$, as given by Eq. (3a), and by the Yukawa assumption on the spatial decay of $c(r)$ outside the core leads to[17]

$$\begin{aligned}-c(x)&=a+bx+\eta a x^3/2+v[1-\exp(-zx)]/(zx)\\&\quad+v^2[\cosh(zx)-1][2Kz^2\exp(z)]^{-1}\ ,\\&\qquad x\equiv r/\sigma<1\end{aligned} \tag{7}$$

where $\eta=(\pi/6)\rho\sigma^3$ is the packing fraction. The Wiener-Hopf factorization method in the form introduced by Baxter[20] further yields analytical expressions for the three coefficients appearing in Eq. (7), viz., a, b, and v, in terms of K, z, and an auxiliary parameter β which, at a given density, is the only physically meaningful solution of an associated quartic equation.[21,22] The above relations for a, b, and v are obtained by requiring the continuity of the function

$$\begin{aligned}\Theta(r) &\equiv y(r)-d(r)\\ &=g(r)-c(r)\end{aligned} \tag{8}$$

together with its first two derivatives across $r=\sigma$. No explicit constraint is imposed on the third derivative of $\Theta(r)$ which for hard spheres in three dimensions should also be continuous at contact.[9] In order to be consistent with such continuity requirements we demand $d(r)$ to be continuous at $r=\sigma$ up to its second derivative. This readily leads to the following relations between the coefficients which parametrize the tail function:

$$\alpha_0=\ln K\ , \tag{9a}$$

$$\alpha_1=-(z+1)\ , \tag{9b}$$

$$\alpha_2=\tfrac{1}{2}\ . \tag{9c}$$

III. THE THERMODYNAMIC CLOSURE

The thermodynamics of the hard-sphere system can be entirely described in terms of a single equation of state. From the virial theorem one has

$$\beta P^{\text{ex}}/\rho=4\eta y(\sigma)\ , \tag{10}$$

where the contact value of the pair distribution function does only depend on the packing fraction η.

On the other hand, statistical mechanics provides a number of additional exact relations which can be used to force the structure of the fluid, as modeled for instance by an approximate $d(r)$, to self-consistently adhere to the thermodynamics of the system.

Two such internal constraints for the asymptotic behavior of the function $y(r)$ near the origin clearly emerge from Eq. (5). In fact, this twofold condition may also be read as supplying an independent "structural estimate" for the excess parts of the chemical potential and pressure through the value of $\ln y(r)$, together with its first space derivative, at $r=0$. We thus require

$$\frac{d}{dr}\ln y(r)\,|_{r=0}=-6\eta y(\sigma)\ . \tag{11}$$

Furthermore, from the knowledge of the pressure it is possible to calculate the corresponding "thermodynamic" value for $\beta\mu^{\text{ex}}$ as a function of η:

$$\beta\mu^{\text{ex}}=\beta a^{\text{ex}}+\beta P^{\text{ex}}/\rho\ , \tag{12}$$

where the excess free energy per particle a^{ex} is given by

$$\beta a^{\text{ex}}(\eta)=\int_0^{\eta}(\beta P^{\text{ex}}/\rho)\frac{d\eta'}{\eta'}\ . \tag{13}$$

Hence, we consistently demand that the two estimates for the excess chemical potential coincide:

$$\ln y(0)=(\beta P^{\text{ex}}/\rho)+\int_0^{\eta}(\beta P^{\text{ex}}/\rho)\frac{d\eta'}{\eta'}\ . \tag{14}$$

Upon differentiating Eq. (14) with respect to η, we find

$$\eta\frac{\partial}{\partial\eta}[\ln y(0)]=\frac{\partial}{\partial\eta}[4\eta^2 y(\sigma)]\ , \tag{15}$$

where use has been made of Eq. (10) for $\beta P^{\text{ex}}/\rho$.

The last structural constraint follows by requiring the second correlation moment for density fluctuations in the fluid to be given by the isothermal compressibility of the system, evaluated as the density derivative of the virial pressure. Use of the OZ equation readily yields

$$\begin{aligned}a &\equiv 1-24\eta\int_0^{\infty}c(x)x^2dx\\ &=1+\frac{\partial}{\partial\eta}[4\eta^2 y(\sigma)]\ .\end{aligned} \tag{16}$$

Obviously, Eqs. (11), (15), and (16) would be identically satisfied with the exact input for the structural properties of the system. However, in the context of an approximate theory, these conditions play the role of well-defined constraints which ensure an intrinsic consistency in the quantitative description of both the structure and the thermodynamics of the model fluid.

After expressing the function $y(r)$ inside the core in terms of $d(r)$ and $c(r)$ through Eq. (4a), we make explicit use of our basic ansatz for the tail function which is formulated in Eq. (6), together with the derived expression for the direct correlation function inside the core as given by Eq. (7). Equations (11), (15), and (16) constitute the thermodynamic closure of the model, and are used to determine the three independent parameters of the theory, namely K, z, and α_3, as a function of density.

IV. NUMERICAL PROCEDURE

The analytical solution of the OZ equation for hard spheres with a Yukawa closure is formulated, at a given density, in terms of K, z, and an additional parameter β. These quantities are further related by the equation[22]

$$K^2+\beta(12\beta\eta-Y)K+\beta^3(36\eta^2\beta-X)=0\ , \tag{17}$$

where

$$\begin{aligned}X=6\eta\Bigg\{ & z\exp(-z)-\frac{6\eta}{z^2(1-\eta)}[2-2z-\exp(-z)(2-z^2)]\\ & -\frac{18\eta^2}{z^2(1-\eta)^2}[2-z-\exp(-z)(2+z)]\Bigg\}\ ,\end{aligned} \tag{18}$$

and

$$\begin{aligned}Y=z & -\frac{6\eta}{z^2(1-\eta)}[2-z^2-2\exp(-z)(1+z)]\\ & -\frac{18\eta^2}{z^2(1-\eta)^2}[2-z-\exp(-z)(2+z)]\ .\end{aligned} \tag{19}$$

Equation (17), complemented by Eqs. (18) and (19) for the coefficients X and Y, leads to a simple expression for K as a function of β, z, and η, the physical root being that one which preserves the ideal-gas behavior in the limit of vanishing density. We are thus lead to choose β, z, and α_3 as the most convenient set of independent parameters for the model at a given density. Our system of nonlinear differential equations is then numerically integrated for a series of densities increasing stepwise from zero, the low-density boundary condition being provided by the Percus-Yevick solution for the hard-sphere system.[23,24] Our numerical procedure is strictly equivalent to selecting out of the ∞^2 solutions supplied, at a given density, by the algebraic equation (11) that one which evolves as a function of η in accordance with the two differential constraints (15) and (16), the free parameters being thus fixed so as to ensure the self-consistency of the model up to that density. The isothermal compressibility K_T is then systematically used for a numerical check on the achieved level of internal consistency via the three independent routes provided by the model for its calculation:

$$\begin{aligned}[\rho(k_B T)K_T]^{-1} &\equiv \left[\frac{\partial(\beta P)}{\partial\rho}\right]_\beta \\ &= \rho\left[\frac{\partial(\beta\mu)}{\partial\rho}\right]_\beta \\ &= a\ . \end{aligned} \tag{20}$$

The three estimates for the inverse compressibility are then required to be congruent within 1%.

V. RESULTS AND DISCUSSION

The system of differential equations (11), (15), and (16) has been integrated for a number of densities up to the freezing region. The calculated values for the three independent parameters of the theory are given in Table I. As expected, β correctly tends to zero at low densities.

In order to assess the quality of the basic ansatz adopted in this theory, we first present our results for $d(r)$ in Fig. 1 in a range of densities. The comparison with the correspondent HG parametrization[15] clearly shows that the composite thermodynamic closure succeeds in modeling the structure of the fluid with remarkable accuracy. The small quantitative discrepancies which still emerge with respect to the external fit can be easily explained in terms of our simplified approximation for the tail function outside the core.

A more detailed insight into the behavior of $d(r)$ at contact as a function of density is gained from Fig. 2. This quantity is directly given by one of the parameters which appear in our theory, namely K, and is also equal to the value at $r=\sigma^+$ of the direct correlation function $c(r)$, information which can thus be independently extracted from the numerical simulation data for $g(r)$ after use of the OZ equation. The agreement between the theoretical prediction and the corresponding experimental values is good over the whole range of explored densities.

We now turn to a critical discussion of our results for

TABLE I. Values of the three independent parameters of the theory as a function of the packing fraction η.

η	β	z	α_3
0.10	0.001	4.16	1.30
0.20	0.006	4.87	1.26
0.30	0.021	5.84	1.04
0.35	0.031	6.84	0.86
0.40	0.041	8.34	0.59
0.45	0.057	10.46	0.16
0.49	0.069	12.87	−0.30

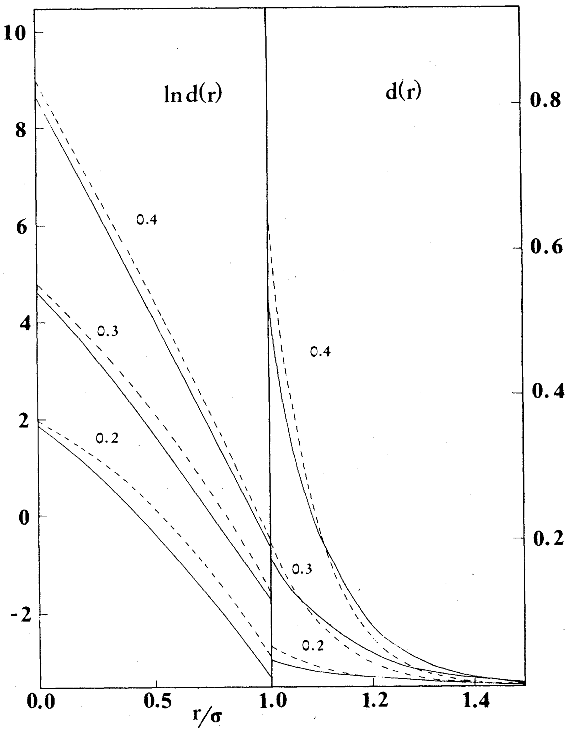

FIG. 1. The function $d(r)$ for hard spheres. Solid curves: present theory. Dashed curves: Henderson and Grundke parameterization. The curves are labeled with the appropriate values of the packing fraction η.

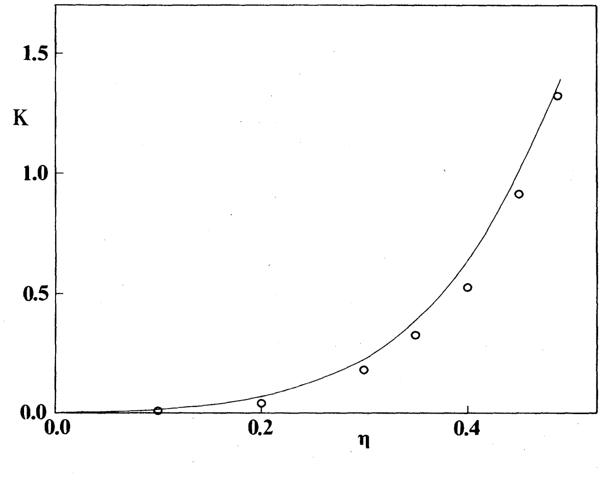

FIG. 2. Contact value of $d(r)$ as a function of η. The continuous line is obtained via the Verlet and Weis parametrization whereas the open circles indicate the predictions of the present theory.

TABLE II. Thermodynamic properties of the hard-sphere fluid as given by the present theory and by the Carnahan and Starling equation of state.

	$\beta P/\rho$		$\frac{\partial(\beta P)}{\partial\rho}$		$\beta\mu^{\text{ex}}$	
η	Theory	CS	Theory	CS	Theory	CS
0.10	1.52	1.52	2.19	2.19	0.98	0.98
0.20	2.39	2.41	4.70	4.71	2.40	2.47
0.30	3.91	3.97	10.02	10.25	4.68	4.87
0.35	5.09	5.21	14.76	15.31	6.38	6.65
0.40	6.69	6.93	22.22	23.22	8.60	9.04
0.45	8.98	9.38	33.60	35.92	11.71	12.33
0.49	11.51	12.16	48.12	51.85	14.95	15.92

the three relevant thermodynamic properties of a hard-sphere fluid, i.e., pressure, inverse isothermal compressibility, and excess chemical potential, which are given in Table II at different values of η. It is apparent that this theory systematically underestimates the experimental values of such quantities as calculated from the Carnahan and Starling (CS) phenomenological equation of state[25] with a discrepancy which steadily increases with η. At the Alder transition density such discrepancies reach 5.3%, 7.2%, and 6.1% for $\beta P/\rho$, $\partial(\beta P/\partial\rho)_T$, and $\beta\mu^{\text{ex}}$, respectively.

The compressibility factor is also graphically displayed as a function of density in Fig. 3. In order to illustrate in a pictorial way the most relevant feature of the present scheme we also show in the same figure the four different estimates which can be obtained for $\beta P/\rho$ versus η when following an inconsistent approach as that given, for instance, by the Percus-Yevick integral theory which is equivalent to assuming $d(r)=0$.[7] Besides the two well-known virial and compressibility equations of state, we plot the value of the compressibility factor calculated from the independent knowledge of the excess chemical potential when this quantity is obtained from the Percus-Yevick value of $\ln y(r)$ at $r=0$. Furthermore, as appears from Eq. (5), the space derivative of the cavity distribution function at the origin is in principle directly related to the virial pressure via the Yvon-Born-Green integral equation.[26] Of course, this relation is not necessarily verified in an approximate scheme, but it can be used as a further measure of the level of structural inconsistency present in the theory. The assumption of a vanishing tail function in the whole range of r clearly leads to a most dramatic discrepancy from the correspondent CS value of these last two estimates for $\beta P/\rho$.

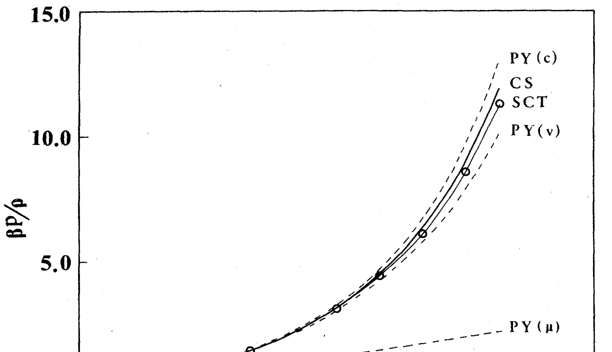

FIG. 3. Equation of state of a hard-sphere fluid. Solid curve: Carnahan and Starling (CS) reduced pressure. Open circles: present self-consistent theory (SCT). Broken curves: independent estimates for $\beta P/\rho$ obtained in the Percus-Yevick (PY) approximation. Labels refer to the different methods adopted for the evaluation of the PY equation of state, as fully discussed in the text.

The agreement between our calculated equation of state and the experimental results is comparable with that achieved by other theories which also make use of partial consistency criteria.[3-6] However, as already emphasized, the novelty of this approach is that, at variance with previously exploited approximation schemes, fully consistent information is obtained for the whole body of thermodynamic properties which can be calculated from the knowledge of the pair correlations in the fluid. This, in turn, leads to a more reliable description of the way the structural properties of the system globally evolve as a function of the thermodynamic state.

The direct correlation function $c(r)$ is shown in Fig. 4 for $\eta=0.2$, 0.3, and 0.4. The same function when calculated after Fourier inversion of the VW radial distribution function[15,16] is also plotted for the same values of the packing fraction η. For $r<\sigma$ it is difficult to resolve between the two curves on the scale of the picture. Furthermore, the naive Yukawa form for $c(r)$ outside the core, when modeled by the thermodynamic consistency requirements, succeeds to mediate between the oscillations exhibited by the hard-sphere direct correlation function at high densities. Such an oscillating space decay appears at $\eta\sim 0.3$, on the scale given by the contact value of $d(r)$. In passing, it is interesting to note that roughly in correspondence with this density the quantity $c\ (r=\sigma^{+})$ starts to grow exponentially as a function of η, as can be verified through the data shown in Fig. 2. Furthermore, the Kirkwood superposition approximation, which is fairly accurate for $\eta\lesssim 0.3$, dramatically breaks down just in

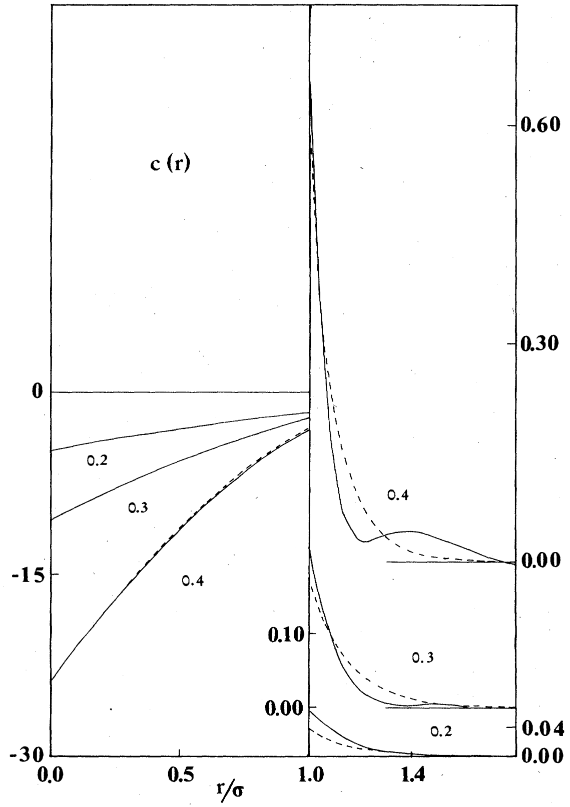

FIG. 4. Direct correlation function for hard spheres at $\eta=0.2$, 0.3, and 0.4. The solid curves are obtained via the Verlet and Weis parametrization for $g(r)$ whereas the dashed curves give the results of the present theory.

this region of densities, as is apparent, for instance, from the density derivative of $g(r)$ at contact.[27] The joint occurrence of the above peculiarities in the density evolution of the hard-sphere structure might be understood in terms of a change of fluid behavior which would take place well before the Alder transition.

We finally present our theoretical prediction for the hard-sphere bridge functions

$$b(r)=[h(r)-c(r)]-\ln y(r) \tag{21}$$

whose role for developing a "universal" picture of the short-range structure in simple fluids has been amply discussed.[1] In Figs. 5–7 the $b(r)$'s are shown at different densities against their "experimental estimate" which is obtained by Eq. (21) after use of the VW and HG parameterizations for the structural properties of the fluid. The agreement is very good at each density even in the range of medium to large values of r where the $b(r)$ also attains negative values showing very small oscillations. Still, after a magnification of the tail, the phase of these oscillations turns out to be accurately reproduced by this theory. In this respect we recall that the Percus-Yevick bridge functions are always positive definite.[1,6] We would finally like to comment that a conclusive quantitative assessment on the relative height of the second peak in the VW $b(r)$ in the region of high densities heavily rests upon a more detailed analysis of the space decay of $c(r)$.[28]

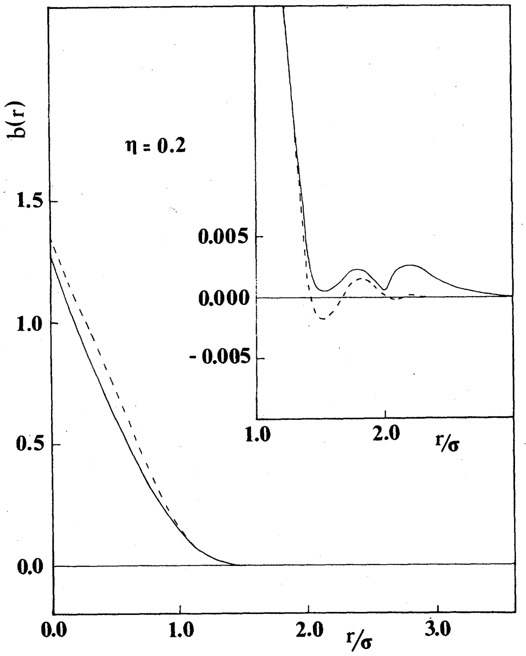

FIG. 5. The bridge function for hard spheres at $\eta=0.2$. Continuous line: results obtained via the Verlet and Weis, and Henderson and Grundke parametrizations. Dashed line: present theory.

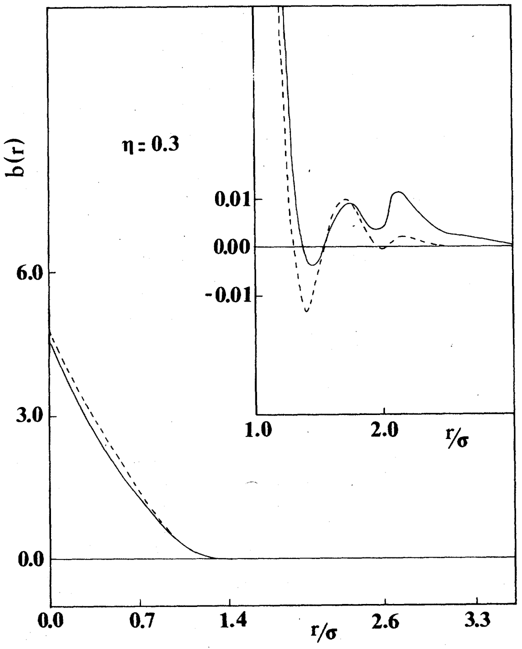

FIG. 6. Same as for Fig. 5 at $\eta=0.3$.

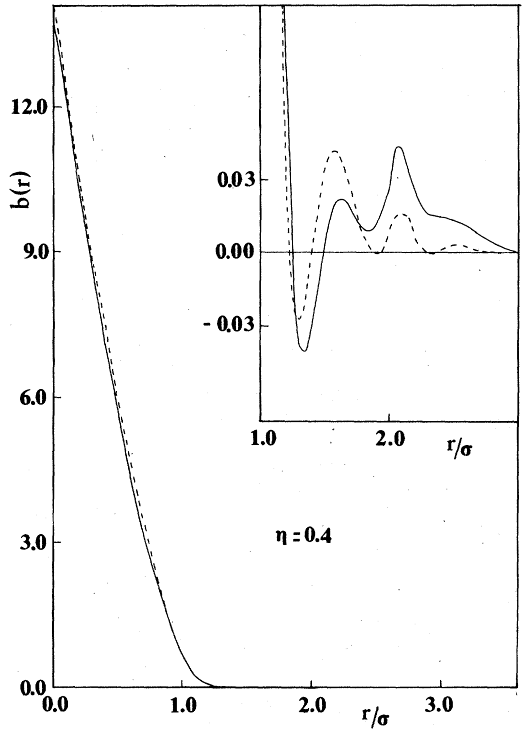

FIG. 7. Same as for Fig. 5 at $\eta=0.4$.

VI. CONCLUDING REMARKS

An analysis of structure versus thermodynamics consistency requirements, beyond the usual virial-compressibility route, for developing a self-contained picture of pair correlations in a hard-sphere fluid, is the main contribution given in the present paper on methodological grounds. In order to verify on a quantitative level the consequences of imposing such nontrivial constraints on the equilibrium structural properties of the system, we have resorted to a simple but reliable ansatz for the tail function $d(r)$ which allows an analytical solution of the OZ integral equation. The numerical results, which have been obtained without using any external data, demonstrate that this theoretical approach is quite promising, leading to a close quantitative description of the equilibrium properties of the model fluid. A refinement of the ansatz for $d(r)$, so as to include the possible occurrence of oscillations in the medium-range decay of this function, is being considered in order to produce a more realistic description of the hard-sphere fluid in the high-density region, without losing the simplicity of an analytical OZ representation of $c(r)$ inside the core. This structural feature may turn relevant for a proper theoretical description of the freezing transition. In this respect we anticipate that our numerical procedure appears to meet with serious convergence problems in the region of the Alder transition. In particular, gradually beyond $\eta \sim 0.51$, we do not succeed in obtaining a totally consistent solution within the previously invoked numerical accuracy. This might be interpreted as an intrinsic instability occurring in the system due to a progressive inability of the fluid to simultaneously saturate the set of coupled thermodynamic constraints. More-detailed investigations are in progress to clarify this point.

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