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Equation of state of the hard-sphere crystal

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An approach to the averaged two-particle distribution function of a crystalline phase is presented. It includes an indirect check of the merit of the Gaussian approximation for the local density and a way to infer values of the thermodynamic variables from simulation data. The equation of state and the compressibility of the hard-sphere fcc crystal is computed from the Tarazona free energy density functional [Phys. Rev. A **31**, 2672 (1985)]. They are in excellent agreement with simulation results over the physical range of densities up to almost close packing. We also include the comparison with the results obtained by two other functional approaches, which are also excellent. [S1063-651X(96)08706-5]

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I. INTRODUCTION

The density-functional theory applied to nonuniform classical fluids has been able to depict a wide range of physical properties of simple solid systems. Initially, it was intended for developing a theoretical approach that was able to describe both fluid and crystalline phases consistently. Then, it was extended to other crystal properties like elasticity and phonon dispersion and to more complex systems such as surfaces and liquid crystals (see Ref. [1] for a recent review on this matter).

It is well known the crucial role of hard spheres (HS) as the usual reference system of more realistic systems that include attractive interactions. Accordingly, a considerable effort has been done to develop free energy functionals describing nonuniform HS systems and, at present, there exist quite good functional approaches for these systems [1]. Recently, Denton *et al.* [2] and Tejero *et al.* [3] have analyzed the equation of state of a HS crystal obtained from the modified weighted-density approximation (MWDA) [4] and from the generalized effective liquid approximation (GELA) [5], respectively. Their analysis include densities well inside the stable solid phase. This is especially relevant in connection with the solid-solid transition recently reported by Bolhius and Frenkel [6] in systems of HS with a short-ranged attractive interaction (we have learned that Stell and collaborators had already predicted these kind of transitions in the 1970s [7]). It happens that, for sufficiently short-ranged attractions, this transition occurs at very high densities, near close packing. To describe this kind of phenomena, the theoretical approach for the reference HS system should give a reasonable equation of state over all the physical density range, even proximal to close packing. Thus, in this paper, we evaluate the equation of state of a face-centered-cubic (fcc) HS crystal at densities up to almost close packing using the Tarazona functional approach [8,9]. What it is more important is to compare the functional predictions with simulations results. To do it, we develop a method to obtain thermodynamic information from simulation data of $\tilde{g}(r)$, the average of the two-particle distribution function. This method also allows us to infer the ideal and the excess contributions (as defined in Sec. II) to the equation of state. It also corroborates the merit of the Gaussian description of the one-particle distribution function. Finally, but no less important, the method suggest an interesting discussion on the averaged correlation between the particles beyond nearest neighbors. The agreement of the Tarazona functional predictions with simulation data is excellent up to almost close packing. The same can be said for the predictions from MWDA and GELA at least up to the densities reported. To test this accordance, we compute also the compressibility of the HS crystal and compare it with that of simulation, finding an excellent agreement. Moreover, we also work out the ideal and excess contributions to the pressure and the compressibility and compare each one with those corresponding to simulation data. Again, the accordance is quite good. Note that the evaluation of the equation of state by MWDA have some problems at high densities as pointed out by Tejero *et al.* [3].

In the next section, we briefly resume the Tarazona functional (TF) together with MWDA and GELA. In Sec. III we present the mentioned discussion of $\tilde{g}(r)$. The results and a discussion of them are presented in Sec. IV. The conclusions are exposed in Sec. V.

II. FUNCTIONAL APPROXIMATIONS

Density-functional theories [1] are based on a variational principle [10] which allows us to propose approximations to the Helmholtz free energy. The variational principle establishes that for a given interacting potential, fixed external potential, and mean density, the Helmholtz free energy $F[\rho(\mathbf{r})]$, as a functional of the one-particle density $\rho(\mathbf{r})$, has a minimum value at the equilibrium density. The free energy functional is usually written as

$$F[\rho(\mathbf{r})]=F_{id}[\rho(\mathbf{r})]+F_{ex}[\rho(\mathbf{r})], \tag{1}$$

where F_{id} is the ideal contribution to the Helmholtz free energy of a nonuniform system. Its functional form is exactly given by

$$\beta F_{id}[\rho(\mathbf{r})]=\int d\mathbf{r}\rho(\mathbf{r})\{\ln[\Lambda^3\rho(\mathbf{r})]-1\}, \tag{2}$$

where Λ is the thermal de Broglie wavelength and $\beta=1/k_BT$. The second term of (1), F_{ex}, called free energy excess, arises from the interacting potential between particles. Several accurate approximations have been proposed for the free energy excess. These are based on a mapping of the thermodynamic properties of the nonuniform systems onto those of a uniform fluid at some effective density (weighted density). Tarazona [8,9] proposes the following expression for the free energy excess of the HS solid:

$$F_{ex}[\rho(\mathbf{r})]=\int d\mathbf{r}\rho(\mathbf{r})\Delta\Psi_{ex}(\overline{\rho(\mathbf{r})}), \tag{3}$$

where $\Delta\Psi_{ex}(\rho)$ is the free energy excess per particle of the uniform system at density ρ and the weighted density, $\overline{\rho}$, is given by

$$\overline{\rho(\mathbf{r})}=\int d\mathbf{r}'\rho(\mathbf{r}')w(|\mathbf{r}-\mathbf{r}'|;\overline{\rho(\mathbf{r})}). \tag{4}$$

The function $w(r)$, in the integral equation that defines $\overline{\rho}$, is specified by requiring that the direct correlation function $c(r)$ obtained from the free energy functional matches that of the HS liquid in the uniform limit. In the MWDA, Denton and Ashcroft use the same mapping idea. However, they propose a global map of the free energy excess onto the free energy of a unique uniform fluid [4,2]:

$$F_{ex}[\rho(\mathbf{r})]=N\Delta\Psi_{ex}(\overline{\rho}), \tag{5}$$

where the weighted density $\overline{\rho}$ is given by

$$\overline{\rho}=\frac{1}{N}\int d\mathbf{r}\rho(\mathbf{r})\int d\mathbf{r}'\rho(\mathbf{r}')w(|\mathbf{r}-\mathbf{r}'|;\overline{\rho}). \tag{6}$$

In the GELA, Lustko and Baus use again a global map but the weighted density (or effective density), $\overline{\rho}$, is given by the structural mapping [5]:

$$\begin{aligned}&\int d\mathbf{r}\rho(\mathbf{r})\int d\mathbf{r}'\rho(\mathbf{r}')c(|\mathbf{r}-\mathbf{r}'|;\overline{\rho})\\&\quad=\int d\mathbf{r}\rho(\mathbf{r})\int d\mathbf{r}'\rho(\mathbf{r}')c(\mathbf{r},\mathbf{r}';\rho(\mathbf{r})),\end{aligned} \tag{7}$$

where c is the direct correlation function.

An important advantage of the two latter approaches is that they require less computational effort. However, only the TF is a true functional approach in the sense that it is not restricted to macroscopically homogeneous systems, as the present case of the HS crystal. For more technical details, the reader is referred to Refs. [9], [4], [5] in relation to TF, MWDA, and GELA, respectively, and to Ref. [1] for an extensive discussion of these and other functional approximations.

The one-particle density, $\rho(\mathbf{r})$, is usually assumed to be described by a sum of normalized Gaussians:

$$\rho(\mathbf{r})=\left(\frac{\alpha}{\pi}\right)^{3/2}\sum_{\mathbf{R}}e^{-\alpha(\mathbf{r}-\mathbf{R})^2}, \tag{8}$$

where $\mathbf{R}$ is the vector position of the crystal lattice and α is the Gaussian width parameter. With this density parametrization, the variational principle reduces to finding the value of α that minimizes the free energy functional at each mean density. Several attempts trying to improve the parametrization of the one-particle density have shown the goodness of the Gaussian one [11]. On the other hand, simulations have shown that deviations from Gaussian form are only significant at low densities but only at the tails of the distribution [12]. For those reasons, the Gaussian parametrization seems to be an excellent description of the one-particle density of the HS crystal over all ranges of physical densities. We shall give another evidence based on Monte Carlo (MC) simulations [13–17].

The equation of state for the HS solid is obtained as follows. After the minimization process we obtain the free energy per particle $f(\rho)=f(\rho,\alpha(\rho))$ at each mean density ρ. Notice that α is, after the minimization, a function of the mean density ρ. Then, at fixed temperature, the pressure is given by

$$\frac{\beta P}{\rho}=\beta\rho\,\frac{\partial f(\rho)}{\partial\rho}. \tag{9}$$

Because the standard division of the Helmholtz free energy (1) into the ideal contribution and the excess one, it follows the equivalent for the free energy per particle, namely, $f=f_{id}+f_{ex}$. Therefore, the equation of state (9) can be formally split into two terms:

$$\frac{\beta P_{id}}{\rho}=\beta\rho\,\frac{\partial f_{id}(\rho)}{\partial\rho}, \tag{10}$$

$$\frac{\beta P_{ex}}{\rho}=\beta\rho\,\frac{\partial f_{ex}(\rho)}{\partial\rho}, \tag{11}$$

where $P=P_{id}+P_{ex}$. Following Denton *et al.*, we call P_{id} and P_{ex} ''ideal-gas pressure'' and ''excess pressure,'' respectively. Notice that P_{id} is not the usual ideal gas pressure, i.e., that which gives the ideal compressibility factor. In addition to the different functional approach used by Denton *et al.* for the Helmholtz free energy excess, these authors approximate the ideal free energy per particle by

$$\beta f_{id}(\alpha)=\frac{3}{2}\ln\left(\frac{\alpha}{\pi}\right)+3\ln(\Lambda)-\frac{5}{2}, \tag{12}$$

which is exact in the limit of nonoverlapping Gaussians. It is a very good approximation over all density ranges of the HS solid. However, we use the exact functional expression (2) to obtain the TF ideal contribution.

An important and direct test for the equation of state is its capacity to predict the isothermal compressibility of the HS crystal. This is given by

$$\rho k_BT\chi_T=\left[\frac{\partial\beta P(\rho)}{\partial\rho}\right]^{-1}, \tag{13}$$

which, extending the above formal division into an ideal part and an excess part, can be written as

$$\rho k_B T \chi_{id} = \left[\frac{\partial \beta P_{id}(\rho)}{\partial \rho} \right]^{-1}, \tag{14}$$

$$\rho k_B T \chi_{ex} = \left[\frac{\partial \beta P_{ex}(\rho)}{\partial \rho} \right]^{-1}, \tag{15}$$

with

$$\frac{1}{\chi} = \frac{1}{\chi_{id}} + \frac{1}{\chi_{ex}}. \tag{16}$$

III. AVERAGE OF THE TWO-PARTICLE DISTRIBUTION FUNCTION

The averaged two-particle distribution, $\tilde{g}(r)$, is defined as

$$\tilde{g}(r_{12}) = \frac{1}{4\pi V \rho^2} \int d\Omega_{12} \int d\mathbf{r}_2 \rho^{(2)}(\mathbf{r}_1,\mathbf{r}_2), \tag{17}$$

where V is the volume, $\rho^{(2)}(\mathbf{r}_1,\mathbf{r}_2)$ the two-particle density function, and $d\Omega_{12}$ the differential solid angle aperture around $\mathbf{r}_{12}$. In the uniform limit Eq. (17) reduces to the well known radial distribution function. MC results for this function were parametrized originally by Weis [13] with the following analytical expression:

$$\tilde{g}(r) = 0, \quad 0 \leqslant r \leqslant 1 \tag{18}$$

and

$$\tilde{g}(r) = \sum_{i \geqslant 1} \tilde{g}^{(i)}(r), \quad r \geqslant 1, \tag{19}$$

with

$$\tilde{g}^{(1)}(r) = \frac{A}{r} e^{-[W_1(r-r_1)]^2 - [W_2(r-r_1)]^4} \tag{20}$$

and

$$\tilde{g}^{(i)}(r) = \frac{1}{4\pi\rho} \left(\frac{W^2}{\pi} \right)^{1/2} n_i \frac{e^{-[W(r-R_i)]^2}}{R_i r}, \quad i \geqslant 2, \tag{21}$$

where n_i is the number of lattice sites at distance R_i and A is determined by the virial theorem. The parameters r_1, W_1, W_2, and W are elaborated analytic functions of the density that give a good fitting to results of MC simulation (distances in all above equations are given in HS diameter units). These functions were refined successively [14–16]. The parameters provided by Choi *et al.* [17] overcome those of previous authors giving an accurate description of the MC results. The maximum root mean square deviation of $\tilde{g}(r)$ from MC data computed over the distance range (r up to 3.3) is 0.17 at the highest density. It quickly decreases to less than 0.06 at packing fractions lower than $\eta = 0.65$.

Here, the interesting point is the functional form used to fit the MC data. The function $\tilde{g}(r)$ is written as a sum of peaks corresponding to successive shells of neighbors. The first peak, Eq. (20), has a characteristic form but all the rest, Eq. (21), have exactly the same functional form. These latter peaks only differ from each other on the distances where they are located, R_i, and on their normalizations which correspond to the number of neighbors at distance R_i (according to a lattice without vacancies).

The crucial point is to realize that if the spherical average of $\rho^{(2)}(\mathbf{r},\mathbf{r}')$, which defines $\tilde{g}(r)$, is done on the product of two sums of Gaussians, $\rho(\mathbf{r})\rho(\mathbf{r}')$, we obtain precisely all the peaks of $\tilde{g}(r)$ except the first one. In effect, from the definition of this average:

$$\tilde{g}_0(r_{12}) = \frac{1}{4\pi V \rho^2} \int d\Omega_{12} \int d\mathbf{r}_2 \rho(\mathbf{r}_1)\rho(\mathbf{r}_2), \tag{22}$$

it is straightforward to obtain

$$\tilde{g}_0(r) = \sum_{i \geqslant 0} \tilde{g}_0^{(i)}(r), \quad r \geqslant 0, \tag{23}$$

with

$$\tilde{g}_0^{(0)}(r) = \frac{1}{4\pi\rho} \left(\frac{\alpha}{2\pi} \right)^{1/2} 2\alpha e^{-(\alpha/2r)^2}, \tag{24}$$

and

$$\tilde{g}_0^{(i)}(r) = \frac{1}{4\pi\rho} \left(\frac{\alpha}{2\pi} \right)^{1/2} n_i \frac{e^{-(\alpha/2)(r-R_i)^2} + e^{-(\alpha/2)(r+R_i)^2}}{R_i r} \tag{25}$$

$$= \frac{1}{4\pi\rho} \left(\frac{\alpha}{2\pi} \right)^{1/2} n_i \frac{e^{-(\alpha/2)(r-R_i)^2}}{R_i r}, \quad i \geqslant 1. \tag{26}$$

We have dropped the second exponential in the expression (25) as its contribution to each peak is, at most, 20 orders of magnitude smaller than the first exponential (for the current values of the Gaussian parameter α). The first peak, Eq. (24), of $\tilde{g}_0(r)$ does not appear in $\tilde{g}(r)$ because the exclusion of the self-interaction. The second one, Eq. (26) for $i=1$, differs from the first one, Eq. (20), of $\tilde{g}(r)$. However, identifying $\alpha/2$ with W^2, all the remaining peaks are exactly the same both in $\tilde{g}(r)$ and in $\tilde{g}_0(r)$.

The identification of peaks in $\tilde{g}(r)$ with those in $\tilde{g}_0(r)$ has several interesting consequences. The immediately obvious one is that the whole two-body correlation between particles beyond nearest neighbors is already included in the product $\rho(\mathbf{r})\rho(\mathbf{r}')$. This point has been suggested and extensively discussed by two of us in relation with the perturbation weighted density approximation (PWDA) for simple systems with attractive interaction potentials [18,19] and it is confirmed in the present discussion. We have proposed that most of the correlation of the two-particle distribution function,

$$\rho^{(2)}(\mathbf{r},\mathbf{r}') \equiv \rho(\mathbf{r})\rho(\mathbf{r}')g(\mathbf{r},\mathbf{r}'), \tag{27}$$

is already described by the mentioned product. Thus, $g(\mathbf{r},\mathbf{r}')$ could be basically approximated by a step function to exclude the self-interaction. Instead, we went further and mapped $g(\mathbf{r},\mathbf{r}')$ into a homogeneous fluid at a very low effective density determined by the compressibility equation. This proved to be an excellent criterium to determine the perturbation contribution to the free energy [19].

On the other hand, the identification of peaks also confirms the goodness of the Gaussian parametrization for the

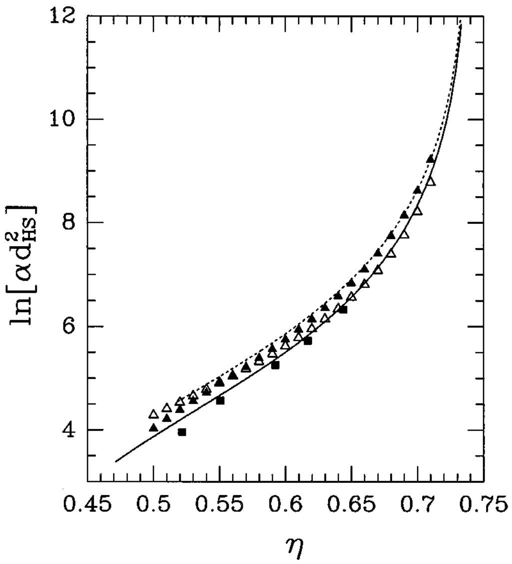

FIG. 1. Logarithm of the Gaussian width parameter α vs packing fraction η. Solid curve inferred, in this paper, from simulation data of Choi *et al.* Solid squares inferred by Denton *et al.* from simulation data of Young and Alder. The dashed curve is the TF prediction. Solid triangles and open triangles are the results from GELA and MWDA, respectively, obtained by Tejero *et al.*

one-particle density. Moreover, it suggests that from the theoretical minimization process of the free energy one can obtain information on the crystal average distribution function $\tilde{g}(r)$. (A work along this line is in progress.) Here, we explore the other way: from MC data [the parameter W in (21)] we obtain the corresponding Gaussian width parameter α of the ''MC Gaussian one-particle density.'' Then, we are able to compute the MC ideal contribution to the free energy, F_{id}, throughout the exact functional form Eq. (2). The MC free energy excess is now immediately obtained and so are the ideal and the excess contributions of the pressure and the compressibility. Notice that the excellent fitting of Choi *et al.* allows us to evaluate analytically all these thermodynamics properties over the density range up to almost close packing. In Fig. 1, for α, we show some of these results in comparison with those inferred by Denton *et al.* [2] from Young and Alder [20] simulation data. The fair agreement is a sign of the consistency of the different MC data. In any case, the treatment of MC data we have followed is much more powerful as it gives a continuous expression of the thermodynamic variables, as a function of density, up to almost close packing.

IV. RESULTS AND DISCUSSION

Figure 1 shows the logarithm of the Gaussian parameter α inferred by us from Choi *et al.* MC simulations [17] and by Denton *et al.* [2] from Young and Alder MC simulations [20]. The predictions of TF, MWDA, and GELA are also displayed. (MWDA data in all figures are those obtained by Tejero *et al.* which we assume free of possible convergence problems.) TF predicts a systematic overestimation of the Gaussian width parameter α but always in fair agreement with simulation results. GELA predictions are better at low densities as it was already known. At high densities both TF and GELA predict close values for α. The MWDA gives the best predictions except at the lowest densities. Figure 2 shows the equation of state predicted by TF, MWDA, and GELA together with simulation results. At low densities (see also Fig. 3), the GELA pressure is slightly above the simulation pressure while TF and MWDA pressures have lower values. All of them improve their agreement with simulation as the pressure increases. At the highest density where we have performed calculations, the TF pressure differs from the simulation one in less than 0.3%. In Fig. 4, we show the two contributions to the pressure, ideal and excess, obtained from TF, MWDA, and GELA in comparison with those inferred

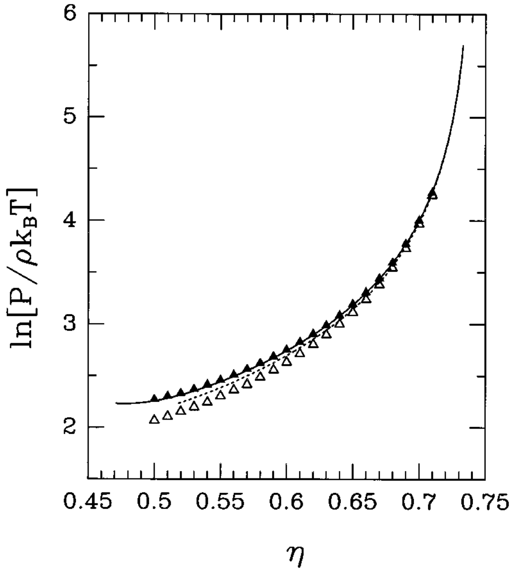

FIG. 2. Logarithm of the equation of state $P/\rho k_B T$ vs packing fraction η. Curves and symbols as in Fig. 1.

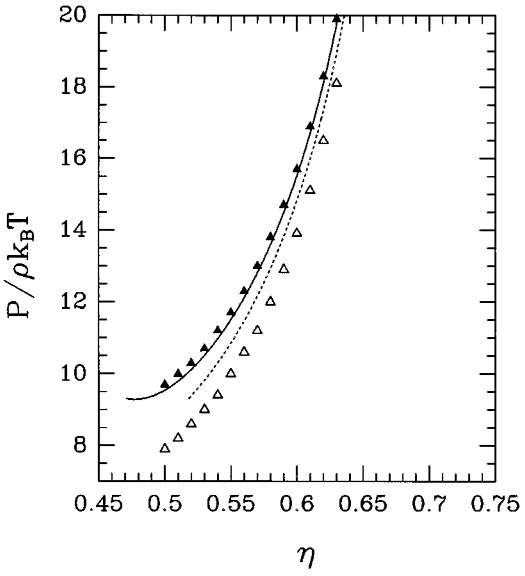

FIG. 3. Equation of state $P/\rho k_B T$ vs packing fraction η at low densities. Curves and symbols as in Fig. 1.

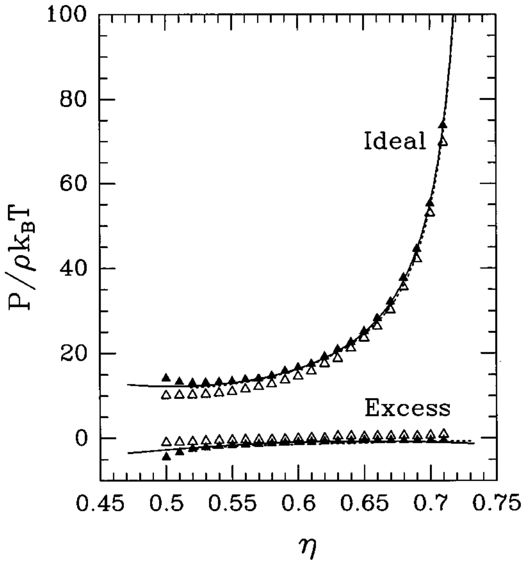

FIG. 4. Equation of state $P/\rho k_B T$ vs packing fraction η. Curves and symbols as in Fig. 1.

from simulation data. TF and GELA show an excellent agreement with simulation results. The MWDA results are also quite good. Notice that the good theoretical predictions for the α parameter (Fig. 1) will necessarily mean comparable good predictions for the ideal pressure (Fig. 4) because Eq. (2) is exact. However, due to the functional approximations proposed for the excess free energy (or for the total free energy) the theoretical excess pressure (or the total pressure) must be also compared with simulation data. This is even more important at high densities where a significant error in the excess contribution would not have appreciable effects on the total pressure. Figure 5 shows the compressibility predicted by TF together with that obtained from simulation. We have used the standard cubic spline treatment of the pressure data to obtain the compressibility via Eq. (13). Once more, the agreement is quite good. We do not have accurate enough data of GELA and MWDA to obtain the compressibility properly. It exhibits unphysical oscillations which, most probably, are due to roundoff effects of pressure data. For completeness, we present in inset of Fig. 5 the inverse of the ideal and excess compressibilities. The agreement with simulation is good.

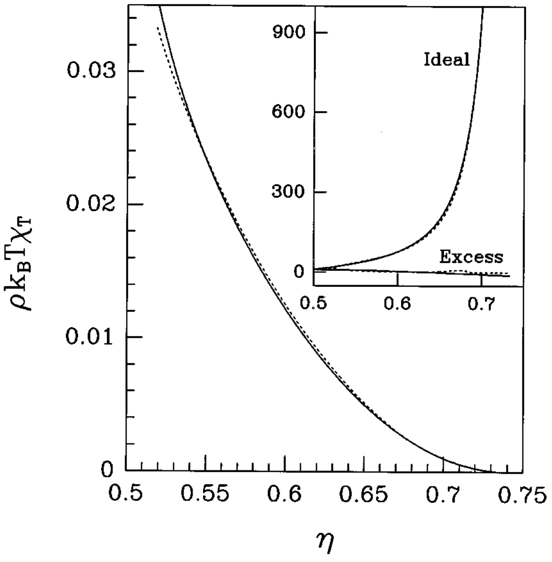

FIG. 5. $\rho k_B T \chi_T$ vs packing fraction η. Inset: $1/\rho k_B T \chi_T$ vs packing fraction η. Curves as in Fig. 1.

The splitting of the pressure into those parts is quite convenient for all the above discussion. However, we are inclined to think that there is no physical meaning in the ideal and the excess pressures, such as they are defined by Eqs. (10) and (11).

V. CONCLUSIONS

In this paper, we have presented two different contributions. One concerns the method used to handle MC data of the $\tilde{g}(r)$ and the other concerns the accuracy of the equation of state computed with functional approaches.

We have proposed a method, using MC data of $\tilde{g}(r)$, to determine the rms displacement of a particle from its equilibrium lattice site [i.e., the parameter α if $\rho(\mathbf{r})$ is approximated by a sum of Gaussians]. This should work perfectly for peaks of $\tilde{g}(r)$ at distances where the two-particle distribution function is already given by the product $\rho(\mathbf{r})\rho(\mathbf{r}')$. In practice, it is possible to deduce already the rms displacement (or parameter α) from the second peak of $\tilde{g}(r)$. This procedure should be consistent with a direct determination of the rms displacement from MC configurations. Assuming the Gaussian parametrization for $\rho(\mathbf{r})$, an accurate fitting of $\tilde{g}(r)$ allows us to determine the parameter α as a function of the density. From this, the ideal pressure is straightforwardly obtained via the exact expression (2). From the total and the ideal pressures, the excess pressure follows immediately.

As a consequence of the analytical fitting of MC data for $\tilde{g}(r)$, it can be deduced that beyond the nearest neighbors the two-particle correlation function is practically given by the product $\rho(\mathbf{r})\rho(\mathbf{r}')$, i.e., $g(\mathbf{r},\mathbf{r}')\approx 1$. We are not aware that this quite interesting and important result was previously mentioned by other authors.

The other aim of this paper has been to show the excellent behavior of the equation of state of the fcc HS solid computed with functional approaches. They agree with simulation results up to almost close packing. This remarkable behavior is extended to the contributions to the pressure, namely, the ideal and the excess pressures, and to the compressibility and their ideal and excess parts. They all are in notable agreement with simulation up to almost close packing.

Finally, we want to remark that functional approaches provide a reliable reference HS system for perturbation theories, especially at high densities. This makes them particularly suitable for describing the solid-solid isostructural transition of simple systems with extremely short-ranged attractive potentials [6]. We have applied the PWDA [18,19] mentioned above to an attractive square well [21] and to HS plus Yukawa attractive tail [22]. Precisely, the PWDA is based, among other things, on two of the points explored here: the goodness of the TF, even at high densities, for describing HS and the accuracy of the product $\rho(\mathbf{r})\rho(\mathbf{r}')$ for describing the correlation between particles beyond nearest neighbors. Note that after Tejero *et al.* discussion on the numerical problems at high densities of the MWDA, results based on this functional approach must be seem with some caution [23].

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