

Radial distribution function for superfluid ^4He , and the effect of interaction potentials thereon

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Abstract The radial distribution function (rdf) for a system of interacting Bosons with the particles interacting via standard two body potentials, has been studied using expression for structure factor derived for such potentials. The dependence of the rdf on the form of potential has been studied. Calculations indicate that potential with soft core does not appear to be realistic, so far as the low temperature properties of liquid ^4He are considered. However, potentials with short-ranged hard core followed by long-ranged repulsive and attractive components yield results comparable to predictions obtainable from experimental studies on liquid ^4He .

Keywords Liquid ^4He , structure factor, radial distribution function

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1. Introduction

The radial distribution function $g(r)$ is the Fourier transform of structure factor $S(k)$. It is a qualitative measure of probability that two particles of a medium are at a distance r apart from each other. Through $S(k)$, it is related to the pair distribution function $P(r_1, r_2)$, that gives the probability of finding a particle at r_1 , when another particle is at r_2 .

Theoretical studies on the superfluid behavior of ^4He , from the standpoint of an interacting Boson System, have mainly concentrated on two forms of interaction potentials as approximation to the realistic ^4He - ^4He interaction potential. Some [1-3] considers the interaction potential to be composed of a short-ranged hard core followed by some kind of attractive or combination of attractive and repulsive components at intermediate and long distances. Yet, others have, for the sake of theoretical convenience, adapted potentials with soft core [4-9]. Aziz *et al* [10] in 1979 determined the ^4He - ^4He interparticle potential using spectroscopic methods. This potential had a short-ranged repulsion of the soft-core type, and using this potential, they were able to predict the differential cross section reasonably well. In spite of this, the potential established by

Aziz *et al* could not be established conclusively as the actual ^4He - ^4He interaction potential. We, in our present work, have studied four potentials. In three of them, the hard core is followed by (i) Two Gaussians (ii) Lennard - Jones and (iii) Square Well potentials. The fourth one is the soft repulsive core potential used by us in one of our earlier calculations [4].

It is well understood that the nature of the interaction potential will determine the form of $g(r)$. Also, since $g(r)$ is related to $S(k)$ through Fourier transformation, once we know the form of $S(k)$, $g(r)$ can be calculated.

Considerable amounts of theoretical effort have been devoted towards developing formalism for $S(k)$ for interacting Boson systems. In this paper, we have concentrated on four different approaches to $S(k)$. Three of them are for hard-core potentials, namely : (i) Feynman's formalism for $S(k)$ [11-13]; (ii) power series expression for $S(k)$, as approximation to Reatto and Chester's [14] expression for $S(k)$, with expansion coefficients obtained from experimental curve-fitting; and (iii) $S(k)$ as obtained by Khanna *et al* [1] by applying reaction matrix formalism to dense Boson system. Finally, a study of the potential with soft core was taken up, following the formalism suggested by Phukan and Bhuyan [4]. The integrations were

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solved by numerical methods, using composite Trapezoid rule [15].

2. Theoretical formulation

For a spherically symmetric Bose fluid, the radial distribution function is given by

$$g(r) = 1 + \frac{1}{(2\pi)^3 \rho} \int_0^\infty [S(k) - 1] \frac{\sin kr}{kr} 4\pi k^2 dk. \quad (1)$$

Here, ρ is the density of the fluid. From eq. (1), it is evident that if we know the structure factor $S(k)$, we can calculate $g(r)$.

2.1. $g(r)$ for hard core potential :

In our present work, three potentials, where the hard core is followed by (i) Gaussian, (ii) Lennard-Jones and (iii) Square-Well potentials have been studied. These potentials can be expressed as follows [1] :

(i) The Gaussian potential :

$$V(r)^G = \begin{cases} +\infty & r \leq a \\ 4\epsilon \exp\left(-\frac{r-a}{\mu_R}\right) - \exp\left(-\frac{r-a}{\mu_A}\right) & r > a \end{cases} \quad (2a)$$

μ_A and μ_R are the ranges of attractive and repulsive forces beyond the hard core, and a is the atomic diameter of ^4He .

(ii) The Lennard-Jones potential

$$V(r)^{L-J} = \begin{cases} +\infty & ; r \leq a, \\ 4\epsilon \left(\frac{a}{r}\right)^{12} - \left(\frac{a}{r}\right)^6 & ; r > a. \end{cases} \quad (2b)$$

and, iii) The Square-Well potential

$$V(r)^{SW} = \begin{cases} +\infty; & r \leq a \\ -V_0 & a \leq r \leq b \\ 0; & r > b \end{cases} \quad (2c)$$

Here, V_0 is the depth of attractive potential.

2.1.a. $g(r)$ from Feynman's relation for $S(k)$:

Feynman's theory of liquid ^4He [11-13] leads to the following form for structure factor :

$$S(k) = \frac{\hbar^2 k^2}{2mE_k} \quad (3)$$

Here, m is the mass, and E_k is the excitation energy. This relation shows good concordance with experimental findings in

the low k (*i.e.* long wavelength) region. In this (*i.e.* $k \rightarrow 0$) region, Landau's relation between E_k and momentum $\hbar k$ is given by

$$E_k = \hbar C k. \quad (4)$$

Here, C is the velocity of sound in liquid ^4He at absolute zero temperature. Using eq. (4) in eq. (3), we get an approximate relation for $S(k)$ in the long wavelength region as

$$S(k) \approx \hbar k / 2mC, \quad (5)$$

where $m = 6.646 \times 10^{-27}$ kg, $\hbar = 1.054 \times 10^{-34}$ joules. sec. C , in the above expression depends on the kind of potential involved. The values of C , in different potentials are [1] : $C_G = 254.93$ m.s. $^{-1}$; $C_{L-J} = 250.44$ m.s. $^{-1}$; $C_{SW} = 238.87$ m.s. $^{-1}$.

Using eq. (5) in eq. (1), we get the expression for $g(r)$ as

$$g(r) = 1 + \frac{1}{r} \left\{ \left(\frac{184.284}{C} \right) \int_0^\infty k^2 \sin kr dk - 2.324 \int_0^\infty k \sin kr dk \right\} \quad (6)$$

Solving eq. (6), we get with integration limit extending from $k = a$ to $k = b$,

$$g(r) = 1 + \frac{1}{r} \left\{ \left(\frac{184.284}{C} \right) \left(\frac{2}{r^2} - k^2 \right) \Big|_a^b + 2.324 k \cos kr \Big|_a^b \right. \\ \left. + \left[2 \frac{184.284}{C} \frac{k}{r} - 2.324 \frac{1}{r} \right] \sin kr \Big|_a^b \right\}$$

C , the sound velocity appearing in eqs. (6) and (7) has different values, as mentioned earlier, depending on the nature of the interaction potential involved. Thus, from eqs. (6) and (7) we get different expression for $g(r)$, for different interaction potentials.

2.1.b. $g(r)$ from power series expression for $S(k)$:

Reatto and Chester [14] have suggested the following form for structure factor

$$S(k) = S_{S,R}(k) \left[1 + \frac{2mC}{\hbar k} G(k, k_c) S_{S,R}(k) \right] \quad (8)$$

where $S_{S,R}(k)$ is the part of the structure factor describing the short-range correlations. The long-wavelength ($k \rightarrow 0$) limit of $S_{S,R}(k)$ is non-zero. Not much is known about the shape of the function G , except that it is flat, is nearly unity for small k , and drops to a small value at the characteristic wave number k_c .

The above expression for $S(k)$ can be approximated [16] to form a power series, in line with the power series expansion of $S(k)$. We have, however, used another form of the power series

expansion for $S(k)$ that may give better agreement with experimental results. The power series is [1]

$$S(k) = \frac{\hbar k}{2m^*c} + \sum_{i=2}^8 C_i k^i, \quad (9)$$

where C_i 's are the constants that are empirically determined to fit experimental curves. The C_i values that we have used in our present calculation, are [1] ; $C_2 = -0.3581166\text{\AA}^2$; $C_3 = 1.5355811\text{\AA}^3$; $C_4 = -2.4346377\text{\AA}^4$; $C_5 = 1.4526855\text{\AA}^5$; $C_6 = -0.3516916\text{\AA}^6$; $C_7 = 0.0244387\text{\AA}^7$; $C_8 = 0.0013406\text{\AA}^8$.

Using eq. (9) in eq.(1), we get an integral equation for $g(r)$ as

$$g(r) = 1 + \frac{2.324}{C_p r} \frac{64.468}{r} \int_0^{\infty} k^2 \sin kr dk + \sum_{i=2}^8 (C_i k^{i+1}) \int_0^{\infty} k \sin kr dk \quad (10)$$

2.1.c $g(r)$ from $S(k)$ obtained by applying reaction matrix formalism to a dense Boson gas .

Khanna *et al* [1] have calculated excitation energy E_k and structure factor $S(k)$, by applying reaction matrix formalism to a dense Boson gas, for each of the above mentioned potentials. In general, all the three $S(k)$ s have similar expressions, differing only in constants. We, here, express the structure factor by a composite expression, representing all three potentials, with the index p standing for any one of G, L-J or SW.

$$S(k)^p = 1 + \frac{1}{\rho E_k^p} \frac{[4A_1 k^3 + B_p (\sin ka + ka \cos ka)] ak^2}{(ka \cos ka - \sin ka) t_{00,00}^p} \quad (11)$$

A_1 and B_p are constants, and

$$E_k^p = [A_1 k^4 + B_p \cdot k \sin ka]^{1/2} \text{ is the Excitation energy} \quad (12)$$

The matrix element $t_{00,00}^p$ has the value :

$$t_{00,00}^p = \frac{4\pi a \hbar^2}{m^*} \left\{ 1 + (8\pi \rho a^3)^{1/2} + \frac{16}{3} \pi \rho a^3 \right\} + T^p. \quad (13)$$

One can write T^p explicitly for the three potentials as :

$$T^p \text{ (Gaussian)} = T^G = 4\pi^{3/2} \epsilon (\mu_R^3 - \mu_A^3); T^p \text{ (Lennard - Jones)} = T^{L-J} = -0.5008 \pi \epsilon a^3; \text{ and } T^p \text{ (Square - Well)} = T^{SW} = -\frac{4}{3} \pi V_0 (b-a)^3.$$

Similarly, one can write A_1 and B_p explicitly as

$$A_1 = \frac{\hbar^4}{4m^{*2}}$$

$$\text{and, } B_p = \frac{\chi^2 \hbar^4}{2a^3 m^{*2}} + C^p;$$

where the constant C^p has the following expressions :

$$C^p \text{ (Gaussian)} = C^G = \frac{4\hbar^2 \pi^{3/2} \rho \epsilon}{m^* a} (\mu_R^3 - \mu_A^3),$$

$$C^p \text{ (Lennard - Jones)} = C^{L-J} = \frac{0.5008 \hbar^2 \pi \rho a^2 \epsilon}{m^* a}$$

$$C^p \text{ (Square - Well)} = C^{SW} = -\frac{4\pi \rho V_0 \hbar}{3m^* a} (b-a)^3.$$

Using eq. (11) in eq. (1), we get the expression for the rdf, $g(r)$ as :

$$g(r)^p = 1 + A_p \int_0^{\infty} \frac{[2k^2 + D_p \frac{\sin X + X \cos X}{2X}]}{[k^2 + D_p \frac{\sin X}{X}]^{3/2}} \frac{X^4}{(X \cos X - \sin X)} \frac{\sin kr}{dr} dX; \text{ With } X = ka \quad (14)$$

As before, eq. (14) is a composite expression, with p having the same meaning as explained before. A_p and D_p are constants, whose values depend on the kind of interaction potential influencing the system.

Under ground state approximation, eq. (14) modifies to :

$$g(r)^p = 1 + A_p \int_0^{\infty} \frac{[2k^2 + D_p]}{[k^2 + D_p]^{1/2}} \frac{X^4}{(X \cos X - \sin X)} \frac{\sin kr}{kr} dX. \quad (15)$$

2.2 $g(r)$ for soft core :

The fourth and last potential studied was the soft core repulsive potential, where the potential is as follows :

$$V(r) = \begin{cases} V_a & ; r < a \\ -V_b & ; a < r < b \\ 0 & ; r > b \end{cases} \quad (16)$$

$$V_a > 0; V_b > 0.$$

Here, V_a and V_b are the depth of attractive and repulsive potentials respectively. Phukan and Bhuyan [4] have derived an expression

for structure factor, $S(k)$, for Boson system interacting via the above mentioned potential, in terms of the Bose distribution function, $f(E_k)$. Following Ref. (4), we express $S(k)$ as :

$$S(k) = \frac{\hbar^2 k^2}{2m^* E_k} [1 + 2f\{E_k\}]; \quad (17)$$

$$\text{With } f(E_k) = \frac{1}{e^{\beta E_k} - 1}; \quad \beta = k_B T. \quad (18)$$

The excitation energy, in this formalism is given as :

$$E_i = \hbar k \left[\frac{\rho}{m^*} \right]^{1/2} v(k) + \frac{\hbar^2 k^2}{4m^* \rho} \quad (19)$$

$$\text{and } v(k) = \frac{204.3}{\rho} \left[\frac{j_1(2.8k)}{2.8k} - 0.6 \frac{j_1(4.4k)}{4.4k} \right]. \quad (20)$$

(in C.G.S. Units)

In the above equation, j_1 is the spherical Bessel function of first order. Using eqs. (17)–(20) in eq. (1), and using the numerical values of the various constants (given in the next section), we get the expression for $g(r)$ as :

$$g(r)^{SC} = 1 + 1.406 \int_0^\infty \left\{ \frac{k^2}{F(k)} \left(1 + \frac{2}{e^{1356F(k)} - 1} - 1.653 \right) \frac{\sin kr}{kr} k^2 dk \right\} \quad (21)$$

$$\text{with } F(k) = [\sin 2.8k - 2.8k \cos 2.8k - 0.155 \sin 4.4k + 0.68k \cos 4.4k + 0.365k^5]^{1/2}. \quad (22)$$

3. Numerical calculations

The values of various parameters used for calculating $g(r)$ are : $a = 2.1 \text{ \AA}$ ($= 2.8 \text{ \AA}$ in soft repulsive core), $b = 4.5 \text{ \AA}$ ($= 4.4 \text{ \AA}$ in the soft repulsive core), $T = 0.6 \text{ K}$, $m^* = 1.23m$, $m = 6.646 \times 10^{-27} \text{ kg}$, $\rho = 2.18 \times 10^{28} \text{ particles/m}^3$, $\chi^2 = 33$, $V_0 = 5 \times 10^{-23} \text{ jules}$, $\mu_R^2 = 0.1103 \text{ \AA}^2$, $\mu_A^2 = 0.2206 \text{ \AA}^2$, $V_a = 40 \times 10^{-23} \text{ jules}$, $V_b = -7.2 \times 10^{-27} \text{ jules}$, $k_B = 1.38 \times 10^{-23} \text{ jules/kelvin}$.

The reason for our choosing these values lies in the fact that earlier calculations [1-4], using these values in interaction potentials as given by eqs. (2a-c) and (16) could bring out good qualitative as well as quantitative agreement between the theoretical values of excitation energy and the experimental energy excitation spectrum for ^4He .

Using these values, the constants A_p and D_p appearing in the eqs. (14) and (15) come out as :

Potential Used	A_p	D_p
Gaussian	0.0314	14.9
Lennard-Jones	0.0327	14.3
Square-Well	0.0357	13.1

Using these constants, eqs. (6), (10), (14), (15) and (21) were numerically integrated using composite Trapezoid rule [15]. As we are concerned with the Superfluid State of ^4He only, we are considering only the phonon interactions, which is the dominant form of interaction in superfluid ^4He . As the phonon region dominates upto $k \sim 2.0 \text{ \AA}^{-1}$, we are integrating the above eqs. From $k = 0.1 \text{ \AA}^{-1}$ to 2.0 \AA^{-1} only (except for one set of calculation involving Gaussian potential, where the upper limit of integration extends upto 2.1 \AA^{-1}). Eq. (7) can also be used, with $a = 0.1$ and $b = 2.0$, which will give the same result as given by eq. (6). The results are summarized in Figures 1-4.

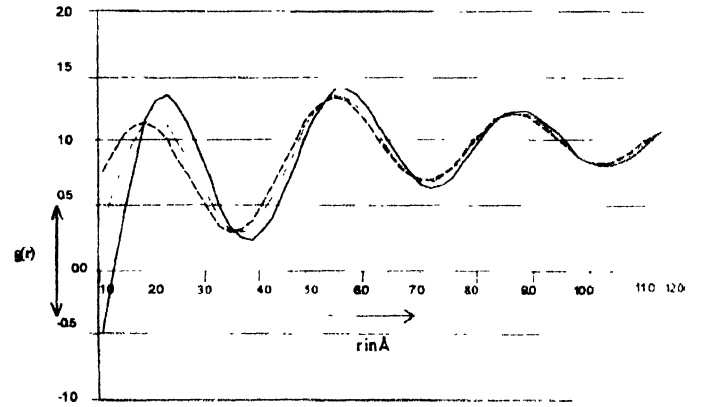


Figure 1. Variations of $g(r)$, with r , as calculated using $S(k)$ derived by reaction matrix formalism (eq 14) for Hard core Potentials Gaussian (.....), Lennard Jones (.....) and Square Well (.....)

Figure 1 plots the variation of $g(r)$ as obtained from eq. (14), for Gaussian (G), Lennard-Jones (L-J) and Square-Well (SW) potential. Figure 2 is drawn using the same eq. (14), with the upper limit of integration extending upto 2.1 \AA^{-1} .

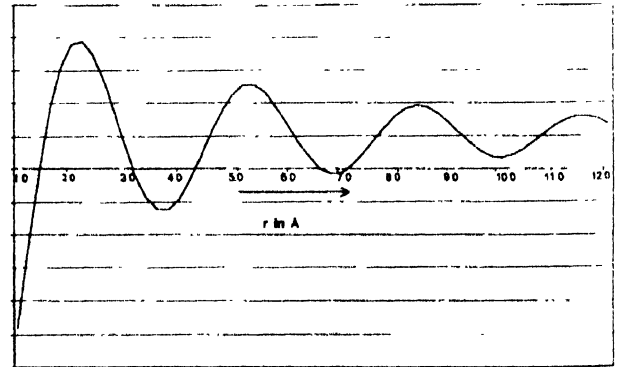


Figure 2. Variation of $g(r)$ with r for Gaussian potential, as calculated from eq. (14) with upper limit of integration extending upto 2.1 \AA^{-1} .

Figure 3 is the $g(r)$ obtained from ground state approximation of eq. (14), as given by eq. (15). As before, it contains three

curves, one each for Gaussian, Lennard Jones and Square- Well potential.

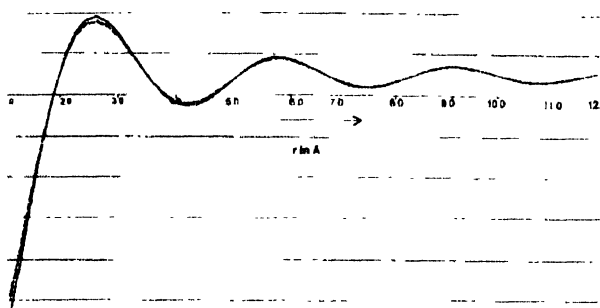


Figure 3. Variations of $g(r)$ with r , as calculated taking ground- state approximation to eq (14), (i.e. eq (15)) Gaussian (-----), Lennard Jones () and Square Well ()

Figure 4 is a composite figure of eight curves. Three of these are drawn for $g(r)$ calculated using the power series expression for $S(k)$ i.e. eq. (10), with one curve each for Gaussian, Lennard Jones and Square-Well Potentials. The next three were drawn for the same potentials using eq. (6). The seventh curve is drawn using $g(r)$ for soft core, as given in eq. (21). The last curve is the experimental curve, as obtained by Svensson *et al* [17] in their neutron diffraction experiment

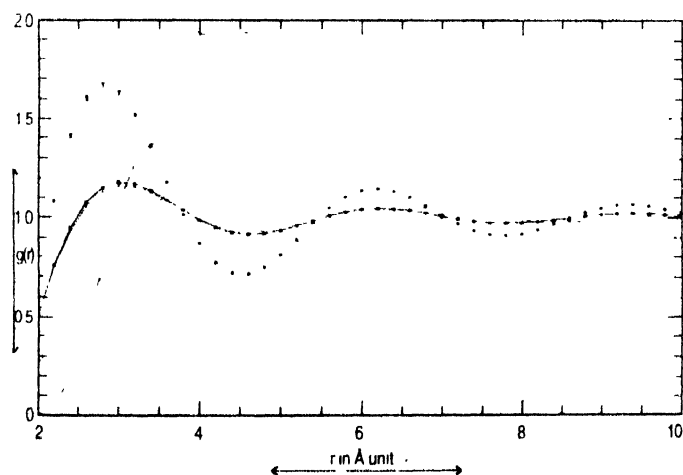


Figure 4. (1) Variation of $g(r)$ with r , as calculated from (a) power series expression for $S(k)$ (eq 10), ● - Gaussian; ▼ - Lennard-Jones; × - Square Well, (b) from Feynman's relation for $S(k)$ (eq 6), ■ - Gaussian, ● - Lennard Jones, ▲ - Square- Well; (c) - soft repulsive core expression for $S(k)$, and (2) - experimental curve (Ref. 17)

4. Discussion

While the curves differ in magnitude of $g(r)$, as well as in the position of maximas and minimas, all of them show periodic variations resembling, to some extent, damped harmonic variation, with $g(r) \rightarrow 1$, as $r \rightarrow \infty$.

At sufficiently close approach, (say \approx ^4He atomic diameter), the Bosons interacting with a hard core potential show quiet

understandably a repelling effect, manifested by a decrease in $g(r)$ values with decreasing r . But the $g(r)$ curve for soft-core interactions shows an increase in $g(r)$, (indicating an increase in probability) when r is lowered below 1.4\AA . This can be explained from the fact that with hard core interactions, particles are repelled on close approach; whereas in the case of interaction with a soft core potential, the particles, because of the soft core, tend to gather together upon close approach. Again, for the soft core potential, $g(r)$ assumes fractional values for $r < 3.4\text{\AA}$, whereas for potentials with hard core, $g(r)$ attains such values only below 1.4\AA . The value of $r = 3.4\text{\AA}$ is far greater than the inter atomic distance in liquid ^4He , which is about 2.5\AA . Thus $g(r)$ becoming fractional at these values of interatomic distances, appear to be unrealistic and consequently, potential with a soft core do not appear to be a realistic potential for systems like liquid ^4He in its superfluid state.

The plot of $g(r)$, as obtained by using $S(k)$ derived from energy excitation values in the reaction matrix formalism [1] shows its second peak (at around 5.4\AA) to be the highest [Figure 1]. However, the same $g(r)$, when approximated for ground state [eq (15)] shows the first peak (at around $r = 2.6\text{\AA}$) to be the highest [Figure 3]. Other curves, obtained from Feynman's relation for $S(k)$ as well as the series expression for $S(k)$ also show their first peak to be the highest, and also has their peaks around $r = 2.8\text{\AA}$. This similarity in form for all the three $g(r)$ can be explained on the basis that all of them are derived from the expression from the expression of ground state structure factor.

While no negative region is seen above $r = 1.2\text{\AA}$ in case of $g(r)$ obtained from eq. (14) with integration limit extending from $k = 0.1\text{\AA}^{-1}$ to 2.0\AA^{-1} , negative regions do appear above $r = 3\text{\AA}$, if one increases the upper limit of integration by some amount say, to 2.1\AA^{-1} . A graph depicting this behavior is the one shown in Figure 2 which is for the Gaussian potential (eq. 14), with integration limit extending from $k = 0.1\text{\AA}^{-1}$. To 2.1\AA^{-1} . This curves shows negative $g(r)$ in the region between $r = 3.2\text{\AA}$ and 4.4\AA and again between $r = 6.8\text{\AA}$ and 7\AA . $g(r)$ becoming negative beyond $r = 3\text{\AA}$ for k - values greater than 2\AA^{-1} may, at first sight indicate breakdown of the formalism for such k - values. We however, would like to consider this to correspond to the existence of the energy gap between the 'phonon' states and 'roton' states in the excitation spectrum of liquid ^4He at around $k = 2\text{\AA}^{-1}$ with a dip in the $E_k - k$ graph at $k = 2\text{\AA}^{-1}$. Moreover, as calculated by Khanna *et al* [1] using reaction matrix formalism, the $S(k)$ values of interacting Bose system becomes negative (approaches negative infinity) and differs sharply with the experimental values above $k = 2\text{\AA}^{-1}$. This disagreement between the theoretical and experimental values was attributed by Khanna *et al* [1] to the disagreement between the theoretical and experimental $E_k - k$ graphs beyond $k = 2\text{\AA}^{-1}$. The negative values of $g(r)$ obtained in our present calculations may thus also be considered to be due to the negative $S(k)$ value beyond $k = 2\text{\AA}^{-1}$ as $g(r)$ is, after all, the Fourier Transform of $S(k)$.

Ceperley [18] had used Path Integral Monte Carlo (PIMC) method to study the pair distribution function and the structure