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Integral equations for fluids of linear molecules
II. Hard dumbell solutions

by F. LADO

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The reference-hypernetted chain integral equation, incorporating a spherically symmetric approximation for the bridge function \( B_0(12) \), has been numerically solved for a fluid of hard dumbells over an extended set of densities and elongations. Comparison with Monte Carlo data shows that the spherically symmetric \( B_0(12) \) is inadequate only when a large density is combined with a large elongation.

1. INTRODUCTION

The preceding paper [1] outlined a general procedure whereby any integral equation can be solved for the structure and thermodynamics of any classical fluid of linear molecules. Here we examine the implementation of this procedure for a specific potential, the hard dumbell model, and a specific integral equation, the reference-hypernetted chain (RHNC) equation [2].

Previous work on the hard dumbell model includes the original use of an integral equation for molecular fluids by Chen and Steele [3], who evaluated two coupled spherical harmonic coefficients of \( g(12) \exp[\beta \phi(12)] \) using the Percus-Yevick closure. With the exception of the reference interaction site model [4], which constitutes a more limited approach distinct from the class of integral equations considered in this series, no other integral equation solutions have been reported for this model. Interest has focused instead on devising perturbation theories that might shortcut some of the computation entailed by the use of integral equations [5]. Since the hard dumbell model is itself the logical candidate for a non-spherical reference system in applying perturbation theory, these approaches necessarily proceed from a spherically averaged reference potential. The method of averaging chosen by Smith and by Perram and White [5] agrees with the general approach described in [1] in zeroth order.

Some aspects of the procedure followed in using the RHNC equation for hard dumbells that are specific to these choices are presented in §2; equations for the calculation of the free energy and pressure are developed here and some details of the numerical calculation are given. Up to 14 spherical harmonic coefficients were evaluated in the solutions reported, though it was found that essentially identical results could be obtained with fewer than half this number. These results, including a representative set of graphs of the various functions that appear in the analysis, are presented in §3 and compared with the Monte Carlo solutions of Streett and Tildesley [6, 7].
2. FORMULATION OF THE PROBLEM

The RHNC closure for the direct correlation function \( C(12) \) in terms of the pair distribution function \( g(12) \) is

\[
C(12) = h(12) - \ln g(12) \exp \left[ \beta \phi(12) \right] + B_0(12), \tag{1}
\]

where \( h(12) = g(12) - 1 \), \( \phi(12) \) is the intermolecular potential, and \( B_0(12) \) is the bridge function for a 'reference' potential \( \phi_0(12) \). With \( h(12) \) eliminated in favour of the series function \( S(12) = h(12) - C(12) \), equation (1) becomes

\[
C(12) = \exp \left[ - \beta \phi(12) + S(12) + B_0(12) \right] - 1 - S(12), \tag{2}
\]

and the calculation is for the spherical harmonic coefficients \( S_{l_1 l_2 m}(r_{12}) \) of \( S(12) \); for an expansion of order \( M \), we write [1]

\[
S(12) = S(r_{12}, x_1, x_2, y) = \sum_{m=0}^{M} \sum_{l_1, l_2 = -m}^{M} S_{l_1 l_2 m}(r_{12}) \mathcal{P}_{l_1 m}(x_1) \mathcal{P}_{l_2 m}(x_2) \alpha_m (-1)^m T_m(y), \tag{3}
\]

where \( x = \cos \theta \), \( y = \cos (\phi_1 - \phi_2) \), \( \mathcal{P}_{l m}(x) \) is the associated Legendre function normalized to \( 2 \), \( T_m(y) \) a Chebyshev polynomial, and

\[
\alpha_m = \begin{cases} 1, & m = 0, \\ 2, & m > 0. \end{cases} \tag{4}
\]

All other quantities can be obtained from the \( S_{l_1 l_2 m}(r) \). The largest \( M \) used in this work is 4; since only even values of \( l \) give rise to non-vanishing coefficients for homonuclear diatomic molecules, this results in a maximum of 14 coefficients:

<table>
<thead>
<tr>
<th>( l = 0 )</th>
<th>( l = 2 )</th>
<th>( l = 4 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( m = 0 )</td>
<td>( S_{000} )</td>
<td>( S_{200} )</td>
</tr>
<tr>
<td>( S_{020} )</td>
<td>( S_{400} )</td>
<td>( S_{220} )</td>
</tr>
<tr>
<td>( S_{040} )</td>
<td>( S_{420} )</td>
<td>( S_{240} )</td>
</tr>
<tr>
<td>( S_{060} )</td>
<td>( S_{440} )</td>
<td>( S_{260} )</td>
</tr>
<tr>
<td>( S_{080} )</td>
<td>( S_{460} )</td>
<td>( S_{280} )</td>
</tr>
<tr>
<td>( S_{042} )</td>
<td>( S_{442} )</td>
<td>( S_{242} )</td>
</tr>
<tr>
<td>( S_{064} )</td>
<td>( S_{464} )</td>
<td>( S_{264} )</td>
</tr>
<tr>
<td>( S_{086} )</td>
<td>( S_{486} )</td>
<td>( S_{286} )</td>
</tr>
<tr>
<td>( S_{044} )</td>
<td>( S_{444} )</td>
<td>( S_{244} )</td>
</tr>
<tr>
<td>( S_{066} )</td>
<td>( S_{466} )</td>
<td>( S_{266} )</td>
</tr>
<tr>
<td>( S_{088} )</td>
<td>( S_{488} )</td>
<td>( S_{288} )</td>
</tr>
</tbody>
</table>

With \( M = 2 \), the rightmost column above is dropped and only five \( S_{l_1 l_2 m} \) appear in the expansion; zeroth order contains just \( S_{000} \).

The thermodynamic values that can be determined directly from a solution are the free energy \( A \), pressure \( p \), and compressibility \( \kappa \).

In the RHNC approximation, the expression for the free energy is obtained by a generalization of the derivation in [1]. Writing the potential in the form [2]

\[
\phi(12; \xi) = \phi_0(12) + \xi \phi_1(12), \tag{5}
\]

with charging parameter \( \xi \) attached only to the increment \( \phi_1 \), the steps followed in [1] produce the difference in free energy between the system with potential
The hard dumbell fluid

\( \phi(12) (\xi = 1) \) and the reference system with potential \( \phi_0(12) (\xi = 0) \) as

\[
\beta \Delta A/N = \beta A_1/N + \beta A_2/N + \beta A_3/N,
\]

where

\[
\begin{align*}
\beta A_1/N &= \beta A_1/N - \beta A_1^{(0)}/N, \\
\beta A_2/N &= \beta A_2/N - \beta A_2^{(0)}/N,
\end{align*}
\]

\[
\beta A_3/N = -\frac{1}{\beta} \int d\xi \int_0^1 d\xi B(12; \xi) g(12; \xi)/\partial \xi |000>,
\]

with [1]

\[
\beta A_1/N = -\frac{1}{2\rho} \int dr \left< \frac{1}{2} h^2(12) + h(12) \right> - g(12) \ln g(12) \exp [\beta \phi(12)] |000>,
\]

\[
\beta A_2/N = -\frac{1}{2\rho} \int \frac{d\mathbf{k}}{(2\pi)^d} \sum_m \{ \ln \text{Det}[I + (-1)^m \rho \mathbf{h}_m(k)] \} (-1)^m \rho \text{Tr}[\mathbf{h}_m(k)]
\]

and similar equations for the reference fluid. The brackets in these expressions denote an average over molecular orientations; they are defined more generally for the inversion of, say, equation (3) as

\[
S_{11,2m}(r) = \int \frac{1}{4\pi} \int \frac{1}{(1-y^2)^{1/2}} dx_1 \int_{-1}^1 dx_2 S(r, x_1, x_2, y)
\]

\[\times \mathcal{P}_{1,m}(x_1) \mathcal{P}_{1,m}(x_2) (-1)^m T_m(y)\]

\[
= \left< S(12) | l_1 l_2 m \right>.
\]

The elements of the matrix \( \mathbf{h}_m(k) \) are the coefficients \( h_{11,2m}(k) \), \( l_1, l_2 \geq m \), of the Fourier transform of \( h(12) \).

The RHNC approximation consists in assuming that the bridge function \( B(12; \xi) \) in (9) does not change significantly as the potential is turned on from \( \phi_0(12) \) at \( \xi = 0 \) to \( \phi(12) \) at \( \xi = 1 \). In this case, the \( \xi \) integration in (9) may be performed approximately to give

\[
\beta \Delta A_3/N \approx -\frac{1}{\beta} \int dr \left< \beta \phi(12) \right> |000>,
\]

so that the excess free energy is finally determined as

\[
\beta A_{\text{ex}}/N = \beta A_{\text{ex}}^{(0)}/N + \beta \Delta A/N,
\]

where \( \beta A_{\text{ex}}^{(0)}/N \) is the excess free energy of the reference system and all the terms of \( \Delta A \) are computable.

As with other hard core models, the integration over intermolecular separation in the pressure formula,

\[
\beta p/\rho = 1 - \frac{1}{\beta} \int dr \left< \frac{1}{2} g(12) r \partial \phi(12) / \partial r \right> |000>,
\]

must be performed analytically. Introducing the Mayer f-function

\[
f(12) = \exp [-\beta \phi(12)] - 1,
\]

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we write (15) as
\[
\beta p / \rho = 1 + \frac{2}{3} \pi \rho \left( \int_{0}^{\infty} dr r^3 g(12) \exp \left[ \beta \phi(12) \right] \frac{\partial \phi(12)}{\partial r} |_{000} \right)
\]
\[
= 1 - \frac{2}{3} \pi \rho \left( \int_{0}^{\infty} dr f(12) \frac{\partial [r^3 g(12) \exp \left[ \beta \phi(12) \right]]}{\partial r} |_{000} \right)
\]
\[
= 1 + \frac{2}{3} \pi \rho \left( \int_{0}^{\infty} dr \frac{\partial [r^3 g(12) \exp \left[ \beta \phi(12) \right]]}{\partial r} |_{000} \right)
\]
\[
= 1 + \frac{2}{3} \pi \rho \langle r^3 g(\hat{r}, x_1, x_2, y) |000\rangle,
\]
(17)
where \( \hat{r} = \hat{r}(x_1, x_2, y) \) is the largest value of the centre-to-centre separation \( r \) for which \( f(12) = -1 \) with the specified orientations of the two molecules. Equation (17) was first obtained by Chen and Steele [8].

The calculation of the compressibility is straightforward:
\[
\rho \chi / \beta = 1 + 4\pi \rho \int_{0}^{\infty} dr r^2 [g_{000}(r) - 1],
\]
(18)
or alternatively [3]
\[
\rho \chi / \beta = \left[ 1 - 4\pi \rho \int_{0}^{\infty} dr r^2 C_{000}(r) \right]^{-1}.
\]
(19)
The second form is more stable in numerical evaluations and was used to obtain the values reported in the next section.

Figure 1. Specification of molecular orientations. The z-axis passes through the molecular centres. (Only half the molecular axis is shown.)

The hard dumbbell potential itself is expressed in terms of the separations between the constituent hard spheres. Labelling the atoms of molecules 1 and 2

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as \( a, b \) and \( a', b' \) respectively, we have (see figure 1)

\[
\begin{align*}
    r_{aa'} &= (A - B_1 + B_2 - C)^{1/2}, \\
    r_{ab'} &= (A - B_1 - B_2 + C)^{1/2}, \\
    r_{ba'} &= (A + B_1 + B_2 + C)^{1/2}, \\
    r_{bb'} &= (A + B_1 - B_2 - C)^{1/2},
\end{align*}
\]

where

\[
\begin{align*}
    A &= r^2 + \frac{1}{2}l^2, \\
    B_j &= rl \cos \theta_j, \\
    C &= \frac{1}{2}l^2(\cos \theta_1 \cos \theta_2 + \sin \theta_1 \sin \theta_2 \cos \phi_{12}),
\end{align*}
\]

with \( r \) the separation between molecular centres and \( l \) the interatomic distance within a molecule. For spheres of diameter \( \sigma \), the potential is then

\[
\phi(12) = \infty, \quad \text{if any } r_{aa'} < \sigma
\]

\[
= 0, \quad \text{otherwise.}
\]

For \( r \) less than

\[
r_0 = (\sigma^2 - \frac{1}{2}l^2)^{1/2},
\]

there is overlap for all orientations, while for all orientations with \( r \) greater than

\[
r_1 = \sigma + l
\]

the potential is zero. Equations (20) can be solved for \( r \) at contact of the four atom-atom pairs, from which the largest \( r \) can be picked for use in (17).

The numerical evaluation of the coefficients \( g_{1_11_2m}(r) \), performed in the calculation as [1]

\[
g_{1_11_2m}(r) = \frac{1}{n_{k_1, k_2, j_1}} \sum_{n_{k_1, k_2, j_1}} w_{k_1} w_{k_2} g(r, x_{k_1}, x_{k_2}, y_j) \times \mathcal{P}_{1_1m}(x_{k_1}) \mathcal{P}_{1_2m}(x_{k_2})(-1)^m T_m(y_j),
\]

where

\[
w_k = (1 - x_k^2)[P_n'(x_k)]^2)^{-1}
\]

and \( x_{k_1}, y_j \) are respectively the roots of the Legendre polynomial \( P_n(x) \) and Chebyshev polynomial \( T_n(y) \), will be most sensitive to the degree \( n \) in the region \( r_0 < r < r_1 \), where the anisotropy of \( \phi(12) \) plays a role. The results of the next section were obtained with \( n = 28 \) in this interval. (There is some indication that this is only marginally adequate for the largest densities and elongations considered here.) Figure 2 illustrates the calculation for the first few coefficients of the Boltzmann factor for a pair of molecules,

\[
e_{1_11_2m}(r) = \langle \exp[ -\beta \phi(12)] \rangle_{1_11_2m}.
\]

For \( r \) larger than \( r_1 \), the integrand \( g(12) \) is much smoother; here the coefficients \( g_{1_11_2m}(r) \) were determined using \( n = 14 \), until the anisotropic part of \( g(12) \) became so small that this quadrature could be avoided altogether by linearization of the anisotropic part of the exponential in (2).
Spherical harmonic coefficients of $\exp \left( -\beta \phi(12) \right)$ for hard dumbells of elongation $l = 0.4\sigma$.

For the specific choice of reference potential determining the bridge function $B_0(12)$ in the RHNC closure we have used the hard sphere model, as parametrized by Verlet and Weis [9] and Henderson and Grundke [10]. Better approximations, incorporating some orientational dependence into $B_0(12)$, are doubtless possible; this choice is a compromise between the desire to improve on the HNC closure [$B_0(12) = 0$] and the need to avoid materially increasing the computing time for an extended set of solutions. In the spirit of Rosenfeld and Ashcroft's view of $B_0(12)$ as an adjustable function [11], the hard sphere diameter $d$ for the spherically symmetric $B_0(r_{12})$ is that of a sphere with the same volume as the hard dumbell,

$$d = (\sigma^3 + \frac{8}{3} \sigma^2 l - \frac{1}{6} l^3)^{1/3},$$

rather than any actual spherical core of $\phi(12)$; no attempt was made to find a 'best' value of the sphere diameter. (For numerical convenience, the diameter actually used to compute $B_0(r)$ was rounded to the nearest grid point.) The excess free energy of the hard sphere reference system, needed in (14), is obtained from the Carnahan–Starling equation of state [12].

3. Results

The RHNC equation has been solved at four densities, $\rho d^3 = 0.2$, 0.4, 0.6, and 0.8, and four elongations at each density, $l/\sigma = 0.2$, 0.4, 0.6 and 0.8. To permit comparisons of the $g_{l/t2m}(r)$ with the Monte Carlo results of Streett and Tildesley [6], additional solutions were obtained for elongations $l/\sigma = 0.2$, 0.4, and 0.6 at the respective densities $\rho d^3 = 0.648$, 0.784, and 0.896 (all three equivalent to $\rho d^3 = 0.5$, as used in [6]).

Functions of $r$ and $k$ were determined on a grid of 512 points in all cases, with $\Delta r/\sigma = 0.02$ and $\Delta k = \pi/N\Delta r$. (For the state with $\rho d^3 = 0.8$ and $l/\sigma = 0.8$, the interval was increased to $\Delta r/\sigma = 0.03$.) Iterations were continued until the largest difference between successive iterates of the $S_{t1t2m}(r)$ satisfied

$$r_j | S_{t1t2m}^{\text{out}}(r_j) - S_{t1t2m}^{\text{in}}(r_j)| < 10^{-3}. \quad (31)$$

Typically, $S_{000}(r)$ was the last to reach convergence.
The hard dumbell fluid

Figure 3. The radial distribution function for hard spheres of diameter 1.16σ and hard dumbells of elongation l=0.4σ at the density $\rho \sigma^3 = 0.8$ for three orders of the spherical harmonic expansions.

Figure 4. Spherical harmonic coefficients of $g(12)$ for hard dumbells of elongation $l=0.4\sigma$ at the density $\rho \sigma^3 = 0.8$. 
Figure 5. Spherically symmetric part of $S(12)$ for hard spheres of diameter $1.16\sigma$ and hard dumbbells of elongation $l=0.4\sigma$ at the density $\rho d^3=0.8$ for three orders of the spherical harmonic expansions. For comparison of scale, $S_{200}(r)$ is also shown.

Figure 6. Spherical harmonic coefficients of $S(12)$ for hard dumbbells of elongation $l=0.4\sigma$ at the density $\rho d^3=0.8$ for two orders of the spherical harmonic expansions.

A representative sample of the functions that appear in the calculation is shown in figures 3 through 9 for the state with density $\rho d^3=0.8$ and elongation $l/\sigma=0.4$.

The successive stages of the centre-to-centre distribution function $g_{000}(r)$ are shown in figure 3, beginning with the hard sphere $g_0(r)$ for spheres of diameter $d/\sigma$ (rounded to 1.16) and passing through the spherical harmonic expansions of $S(12)$ of order 0, 2, and 4. There is no difference, on the scale shown, between the last two cases, indicating a rapid convergence of the $S(12)$ expansion. The first four coefficients of $g(12)$, in figure 4, are seen to retain the general form of their low density limit $\epsilon_{t_1t_2m}(r)$, with more pronounced structure.
Figure 7. Spherical harmonic coefficients of $C(12)$ for hard dumbells of elongation $l=0.4\sigma$ at the density $\rho d^3=0.8$.

Figure 8. Spherical harmonic coefficients of the Fourier transform of $h(12)$ for hard dumbells of elongation $l=0.4\sigma$ at the density $\rho d^3=0.8$. 
The same stages in the calculation of $S_{000}(r)$ are depicted in figure 5, along with $S_{200}(r)$ for a comparison of scale. Again, the $M=2$ and $M=4$ expansions yield the same curve. (Curiously, the hard sphere $S_0(r)$ is here a closer approximation to the final $S_{000}(r)$ than the $M=0$ curve based on an angle averaged Boltzmann factor. This is not true in all cases.) The most noteworthy feature of this figure is the great disparity in scale between $S_{000}(r)$ and the first nonspherical coefficient $S_{200}(r)$, giving graphic weight to the expected rapid convergence of the expansion of $S(12)$. Additional harmonic coefficients of $S(12)$ are shown in figure 6. On this expanded scale, a small change can be seen at very small $r$ in going from the second order to the fourth order expansion.

The same set of coefficients of the direct correlation function are shown in figure 7 as are given of $g(12)$ in figure 4. As with $S(12)$, the difference in scale of $C_{000}(r)$ and higher coefficients contrasts with that of the $g_{l_1,l_2,m}(r)$. The $C_{l_1,l_2,m}(r)$ show structure both for small $r$ (as in the series function $S$) and in the range of variation of the potential (as in the pair distribution function $g$). All coefficients quickly become very small outside the range of $\phi(12)$.

Figure 8 shows the spherical harmonic coefficients of $\hat{h}(12)$ expanded in a frame with $z$-axis along $k$. Off-diagonal coefficients ($l_1 \neq l_2$) vanish identically at the origin; for the case shown, the diagonal elements $\hat{h}_{l_1m}(k)$ are all very small at $k=0$ (except of course for $l=0$).

Concluding this sample of graphs from a single thermodynamic state, we show in figure 9 three radial slices of $g(12)$ for the crossed ($\theta_1 = \theta_2 = \phi_{12} = \pi/2$), parallel ($\theta_1 = \theta_2 = \pi/2, \phi_{12} = 0$), and $T$-shaped ($\theta_1 = \pi/2, \theta_2 = 0$) configurations. (The respective distances of closest approach are $r_0, \sigma$, and $\frac{1}{2} l + (\sigma^2 - \frac{1}{4} l^2)^{1/2}$.) The monotonic decline of these cross sections in the vicinity of contact is in qualitative accord with the perturbation theory results of Melnyk and Smith [5] and the painstaking Monte Carlo calculations of Cummings et al. [13].
Computed results at low densities are quite good. For a comparison with Monte Carlo data, we have chosen the density most thoroughly studied by Streett and Tildesley [6], $\rho\sigma^3=0.5$. At the elongations $l/\sigma=0.2$, 0.4, and 0.6, this corresponds to the densities $\rho\sigma^3=0.648$, 0.784, and 0.896, respectively. Figures 10, 11, and 12 display the comparisons for the first few harmonic coefficients of $g(12)$. Agreement is good at the smallest elongation, with only two peaks at small $r$ being missed. As the elongation increases, these discrepancies at small $r$ progressively worsen and spread to the main peak of $g_{000}(r)$. This behaviour is not unexpected: the simple spherically symmetric approximation chosen here for $B_0(12)$ cannot adapt to changing asymmetry in the molecule and becomes progressively less adequate as this asymmetry increases. Because a meaningful increase in the degree $n$ of the numerical quadrature for the $g_{n\ell m}$ leads to greatly increased computing times, it is not known what part, if any, of these disagreements stems from inadequacies of the numerical algorithm.

![Figure 10](image.png)

Figure 10. Comparison of MC and RHNC results for the spherical harmonics of $g(12)$ for hard dumbells of elongation $l=0.2\sigma$ at the density $\rho\sigma^3=0.648$.

Finally, the thermodynamic data obtained from the solutions of the RHNC equation are summarized in the table, along with the corresponding Monte Carlo values [7]. (The Monte Carlo pressure is the direct result of the simulation; the free energy and compressibility are obtained from the excellent fit to these data given by Tildesley and Streett [7].) Here we see the same trends mentioned above. At low densities, the computed values are uniformly good. With increasing density, a dependence on elongation in the agreement between the two sets of data becomes apparent; the computed pressure in particular deteriorates quickly.
Figure 11. Comparison of MC and RHNC results for the spherical harmonics of $g(12)$ for hard dumbbells of elongation $l = 0.4\sigma$ at the density $\rho d^3 = 0.784$.

Figure 12. Comparison of MC and RHNC results for the spherical harmonics of $g(12)$ for hard dumbbells of elongation $l = 0.6\sigma$ at the density $\rho d^3 = 0.896$. 
The free energy, pressure, and compressibility of hard dumbells from MC simulation and the RHNC equation using a spherically symmetric $B_0(12)$.

<table>
<thead>
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<th>$l/\sigma$</th>
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<th>0-6</th>
<th>0-8</th>
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<td>RHNC</td>
<td>MC</td>
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<td>1.56</td>
<td>1.59</td>
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<tr>
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<td>$\rho x/\beta$</td>
<td>0.0354</td>
<td>0.0348</td>
<td>0.0334</td>
<td>0.0338</td>
</tr>
</tbody>
</table>

Viewed as a feasibility study of the procedure outlined in [1], the present results are encouraging. Even the simple, spherically symmetric approximation for $B_0(12)$ produces reliable data if not overly strained by a large asymmetry of the molecule at a high density. One may hope to do better with an approximation that will reflect the changing geometry of the molecular species.

I should like to thank Professor William B. Streett for providing me with the tabulated data from his Monte Carlo studies.

**References**