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Theoretical Thermodynamic Properties of Low Temperature Fluids

Zul Azhar Zahid Jamal
Western Michigan University

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THEORETICAL THERMODYNAMIC PROPERTIES
OF LOW TEMPERATURE FLUIDS

by

Zul Azhar Zahid Jamal

A Thesis
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Faculty of The Graduate College
in partial fulfillment of the
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THEORETICAL THERMODYNAMIC PROPERTIES OF LOW TEMPERATURE FLUIDS

Zul Azhar Zahid Jamal, M.A.
Western Michigan University, 1987

The thermodynamic functions for low temperature fluids are computed using parametric integral equations and perturbation theory. Parametric integral equation \( N \) is applied to a low temperature Lennard-Jones gas. It is found that there is no significant improvement over the better known parametric integral equation \( C \). The two parameter integral equation \( T \) is applied to a low temperature square-well potential and is found to be unsatisfactory at reduced temperatures of \( T^* = 1.4 \) and 1.6, but quite accurate at \( T^* = 2.2 \). The equation \( T \) results for \( T^* = 2.2 \) are used as the reference system in perturbation theory computations. Tables of (reduced) pressure, internal energy, entropy, enthalpy, Helmholtz energy, and Gibbs energy are constructed for reduced density \( n^* \leq 0.85 \), and reduced temperature \( 0.7 \leq T^* \leq 2.15 \). The results of reduced pressure agree very well with the molecular dynamic results but results of reduced internal energy deviate from the accepted values for large perturbations.
ACKNOWLEDGEMENTS

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CHAPTER I

INTRODUCTION

A fundamental problem in statistical mechanics is to compute the thermodynamic properties of a material. In this research integral equation methods and perturbation theory are used to compute the thermodynamic functions of simple classical fluids at low temperatures. Results which are presented include the values of pressure, internal energy, entropy, enthalpy, Helmholtz energy, and Gibbs energy.

The System

In a simple classical fluid, it is assumed that the system consists of particles which interact with pairwise radial forces. These forces depend only on the separation distance of the pair of particles for which classical mechanics is good enough to describe the system. The Hamiltonian of a simple classical fluid is given by

\[ H = K + \tilde{\phi} = K + \frac{1}{2} \sum_i \sum_j \phi(r_{ij}), \]  

where \( K \) is the kinetic energy of the particles and \( \tilde{\phi} \) is the potential energy of the particles which includes the
sum of the pair energy of particles $\theta(rij)$ where $rij$ is the distance between particles $i$ and $j$.

The Lennard-Jones pair potential studied here is of the form\(^1\)

$$\theta(r) = 4\epsilon \left[ (\sigma/r)^{12} - (\sigma/r)^6 \right], \quad (2)$$

where $\epsilon$ and $\sigma$ are parameters characteristic of the molecules which make up the system.

The square-well potential studied here is of the form\(^2\)

\[
\begin{align*}
\theta(r) &= \infty, \quad r < d, \\
\theta(r) &= -\epsilon, \quad d \leq r \leq 1.5d, \\
\theta(r) &= 0, \quad r > 1.5d,
\end{align*}
\]

where $\epsilon$ is the well depth and $d$ is the diameter of the hard-sphere core.

**Radial Distribution Function**

The radial distribution function is defined by

$$g(r) = \frac{n(r)}{n}, \quad (4)$$

where $n(r)$ is the average number of particles per unit volume at a distance $r$ from a central particle and $n$ is the average number density which is the total number of particles divided by the volume of the system ($n = N/V$). In terms of potential energy, $g(r)$ can be written as\(^3\)
\[ g(r) = v^2 z^{-1} \int \cdots \int \exp \left( -\frac{\phi}{kT} \right) \, dr_3 \cdots dr_N, \quad (5) \]
\[ Z = \int \cdots \int \exp \left( -\frac{\phi}{kT} \right) \, dr_1 \cdots dr_N, \]

where \( k \) is Boltzmann's constant and \( T \) is the absolute temperature. The integration is over the position coordinates of the particles. Figure 1 shows a typical shape of a radial distribution function.

![Graph of Radial Distribution Function](image)

**Figure 1.** Graph of Radial Distribution Function as a Function of Particle Separation \( x \) for \( n^* = 0.80 \) at \( T^* = 1.6 \) (For Parametric Integral Equation \( T \) with Parameters \( b_2 = -1.80 \) and \( b_3 = 0.55 \))
Once g(r) is known, the pressure (P) and internal energy (U) can be computed by the following relationships:

\[ p^* = \frac{PV}{NkT} = 1 - \frac{2\pi N}{3VkT} \int_0^\infty d\theta \frac{gr^3}{\theta} \, dr, \quad (6) \]

and

\[ U^* = \frac{2U}{3NkT} = 1 + \frac{4\pi N}{3VkT} \int_0^\infty 0 \, gr^2 \, dr. \quad (7) \]

When the pressure and internal energy are computed over a range of temperatures and densities, it is possible to compute other thermodynamic functions. Therefore, g(r) contains a complete thermodynamic description of the system.
CHAPTER II

METHODS FOR COMPUTING THERMODYNAMIC FUNCTIONS

Exact Methods

The most direct way of computing \( g(r) \) (thus getting the thermodynamic functions) is by a method called molecular dynamics\(^5\) (MD). Here Newton's equations of motion are solved on a computer to obtain the motions of the particles. Then relevant averages that are related to the thermodynamic properties are computed. Another method is called Monte Carlo\(^6\) (MC). In this method the motions of the particles are governed by probability rules and averages related to the thermodynamic properties can be computed. Both methods are very direct, therefore these results are often called "exact" (however there are approximations involved) and are used as the accepted values. The main disadvantage of these methods are large computer and long computer time requirements. In this research, the molecular dynamics results of Alder, Young, and Mark\(^7\) are used for comparison for the square-well system.

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Integral Equations

Integral equations are approximate equations for computing radial distribution function. Most give good results at low densities and high temperatures but produce large errors at high densities and low temperatures. The advantage of integral equations is the shorter computer time requirements compared to the "exact" methods.

Integral equations of interest here are the hypernetted chain\(^8\) (HNC), Percus-Yevick\(^9\) (PY), and parametric integral equations. Three parametric integral equations that have been studied are equation C,\(^10\) equation N,\(^11\) and equation T.\(^12\) These integral equations are given by the Ornstein-Zernike equation,\(^13\)

\[
h(12) = c(12) + \int c(13)h(23)\,d3, \tag{8}
\]

where

\[
h = g - 1. \tag{9}
\]

The function \(h\) is the total correlation function and the function \(c\) is the direct correlation function. (12) indicates the correlation of particle 1 on particle 2 and the same apply to (13) and (23). In terms of the direct correlation function, the integral equations are given by

\[
\text{(HNC)} \quad c = g - 1 - \ln (ge^{g\phi}), \tag{10}
\]

\[
\text{(PY)} \quad c = g (1 - e^{g\phi}), \tag{11}
\]

\[
\text{(Equation C)} \quad c = g - 1 - \frac{1}{a} \ln (age^{a\phi} - a + 1), \tag{12}
\]

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\[(\text{Equation N}) \quad c = g - 1 - \frac{\log e g}{g e - 1} \quad , \quad (13)\]

where \( \beta = (kT)^{-1} \) and \( a \) is an adjustable parameter. There is no simple relationship between \( c \) and \( g \) for equation T.

A function \( S \) is introduced,

\[ S = h - c. \quad (14) \]

Using this and equation (8), the integral equations can be written as

(PY) \[ g = e^{-\beta \phi} (1 + S), \quad (15) \]

(HNC) \[ g = e^{-\beta \phi} \int e^{S} \]
\[ = e^{-\beta \phi} [1 + S + (1/2)S^2 + (1/6)S^3 + ...], \quad (16) \]

(Equation C) \[ g = e^{-\beta \phi} [1 + 1/a (e^aS - 1)] \]
\[ = e^{-\beta \phi} [1 + S + (1/2)aS^2 + (1/6)a^2S^3 + ...], \quad (17) \]

(Equation N) \[ g = e^{-\beta \phi} \frac{(1 + aS)}{1 + aS - S} \]
\[ = e^{-\beta \phi} [1 + S + (1 - a)S^2 + (1 - a)^2S^3 + ...], \quad (18) \]

(Equation T) \[ g = e^{-\beta \phi} (1 + S + b_2S^2 + b_3S^3). \quad (19) \]

where \( a, b_2, \) and \( b_3 \) are adjustable parameters.

In this research parametric equation N is studied for a low temperature Lennard-Jones gas and equation T is solved for \( g \) for three temperatures and for several densities and sets of parameters.

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Perturbation Method

The first order perturbation theory of Zwanzig\textsuperscript{14} will be considered. Here the pair potential energy is separated into sum of a reference potential $\varphi_r$ and perturbing potential $\varphi_p$,

$$\varphi = \varphi_r + \varphi_p.$$  \hspace{1cm} (20)

The system Helmholtz free energy ($F$) and the reference Helmholtz free energy ($F_r$) are related by\textsuperscript{15}

$$F = F_r + F_p = F_r + \frac{N^2}{2V} \int_0^\infty \varphi_r g_r \ 4\pi r^2 dr.$$  \hspace{1cm} (21)

The thermodynamic functions can then be calculated from the $g_r$ and the correction terms involving the perturbation potential. Results of equation T are used in this research to calculate the thermodynamic functions of the reference system.
CHAPTER III

COMPUTATIONAL METHOD

Solutions of Equation N

The method of solution of equation N is essentially that of Broyles. This involves an iterative procedure where equation N is solved numerically on a computer to obtain the radial distribution function.

The following dimensionless quantities are introduced for the Lennard-Jones pair potential,

\[ x = r/\sigma , \]
\[ T^* = kT/\varepsilon = 1/8\varepsilon , \]
\[ n^* = \bar{n}\sigma^3 . \]

Equations (6) and (7) can then be written as

\[ P^* = 1 - \frac{16\pi n^*}{T^*} \int_0^\infty (x^{-10} - 2 x^{-10}) g(x) \, dx , \] \hfill (25)
and

\[ U^* = 1 + \frac{16\pi n^*}{3T^*} \int_0^\infty (x^{-10} - x^{-4}) g(x) \, dx . \] \hfill (26)

The computer gives solutions for 75 points with an x interval of 0.075 and for 297 points with an x interval of 0.01875. Solutions of 297 points will only be considered. The truncation point is at x = 5.55.

The computational steps are as follows,
1) Equation N is solved for the isotherm \( T^* = 1.6 \) at reduced densities of \( n^* = 0.70 \) and \( n^* = 0.90 \). Several different values of the parameter \( a \) are used.

2) Graphs of \( P^* \) versus \( a \) for both the reduced densities are then drawn. Here, results by D. D. Carley\textsuperscript{18} using integral equations combined with perturbation theory are used for comparison.

**Solutions of Equation T**

The method of solution here is the same as the method of solving equation N. The same dimensionless quantities are introduced except for the square-well,

\[
x = r/d ,
\]

\[
n^* = N d^3 / V .
\]

\( P^* \) and \( U^* \) can then be written as\textsuperscript{19}

\[
P^* = 1 + (2/3) \pi n^* \{ g(1.0) - 3.375 g(1.5) (1-e^{-\beta \epsilon}) \},
\]

and

\[
U^* = 1 - 4 \pi n^* \frac{\int_{1.0}^{1.5} g(x) x^2 \, dx}{3 T^*},
\]

where \( g(1.0) \) is the radial distribution function as \( x \) approaches 1.0 from above and \( g(1.5) \) is the radial distribution function as \( x \) approaches 1.5 from below.

The computer gives solutions for 75 points with an \( x \) interval of 0.07142857 and for 297 points with an \( x \)
interval of 0.01785714. As before, solutions of 297
points will only be considered. The truncation point is
at \( x = 5.2857 \).

The computational steps of equation T are as
follows:

1) Equation T is solved for the isotherms \( T^* = 1.4, \)
1.6, and 2.2 at a high and an intermediate density.
Different sets of parameters are used so that a grid
can be set up for the parameters \((b_2, b_3)\).

2) With the above results, a least square-fit computer
program is used to determine the c's that give the
best least square fit according to the equation
\[ P^* = P_0 + c_1 b_2 + c_2 b_3 + c_3 b_2^2 + c_4 b_3^2 + c_5 b_2 b_3. \]  

3) A computer program is then used to find values of \(b_2\)
and \(b_3\) that give results equal to the accepted
results. Molecular dynamics results of Alder are
used for comparison. A graph of \(b_3\) versus \(b_2\) is
then plotted, and the point of interception is taken
to be the best choice of parameters for the iso­
therm. Since equation (31) is approximate, equation
T is solved with these "best" values to see if
indeed they give agreement with accepted values of
\(P^*\).

4) Equation T is solved again at other densities, using
the best choice of parameters. With the results,
another computer program is then used to obtain the
interpolation coefficients (Pade coefficients) which can then be used to find the values for other densities without going through the long iteration procedure again. The two interpolation formulas for \( p^* \) and \( U^* \) are

\[
p^* = \frac{1 + a_1n^* + a_2n^*2 + a_3n^*3}{1 + a_4n^* + a_5n^*2 + a_6n^*3}, \tag{32}
\]

and

\[
U^* = \frac{1 + a_1n^* + a_2n^*2 + a_3n^*3}{1 + a_4n^* + a_5n^*2 + a_6n^*3}. \tag{33}
\]

The results for pressure only are used in determining the best choice of parameters because results in the internal energy are not as sensitive to changes in the parameters.

**Computation of Thermodynamic Functions (Perturbation Method)**

The reference potential is taken to be

\[
\varphi_r(r) = \begin{cases} 
\infty, & r < d, \\
-\alpha \epsilon, & d \leq r \leq 1.5d, \\
0, & r > 1.5d,
\end{cases} \tag{34}
\]

where \( \alpha \) is a constant.

The following dimensionless quantities are introduced,

\[
T^+ = T^*/\alpha, \tag{35}
\]

\[
F^* = 2F/3NkT. \tag{36}
\]
The system and the reference system are then both square-well where in the well

\[
\frac{\theta}{kT} = \frac{1}{T^*}, \quad (37)
\]

\[
\frac{\theta_r}{kT} = -\frac{1}{T^+}. \quad (38)
\]

\( P^* \) and \( U^* \) are related by the following equation

\[
dF^* = \left( \frac{2P^*}{3n^*} \right) dn^* - \left( \frac{U^*}{T^*} \right) dT^*. \quad (39)
\]

Equation 21 can be written as

\[
F^* = F_r^* + F_p^* = F_r^* + \frac{4\pi n^*(\kappa - 1)}{3T^*} \int_{1.0}^{1.5} g(x)x^2dx. \quad (40)
\]

Comparison of the above equation with equation 30 will show that

\[
F_p^* = \left( 1 - \frac{T^+}{T^*} \right) \left( 1 - U_r^* \right). \quad (41)
\]

Equations 39, 40, and 41 are the basic equations that are used to compute the thermodynamic functions.

A computer program by D. D. Carley and modified by Z. Z. Jamal is used to do the computations. The program does computations at reference temperature \( T^* = 2.2 \) and the thermodynamic functions are constructed for \( 0.7 \leq T^* \leq 2.15 \), and \( 0.05 \leq n^* \leq 0.85 \) for an interval of 0.05. The computational steps in the program are as follows:

1) \( T^* = 20.0, n^* = 0.001 \) is taken to be the standard state and \( F^* \) in that state is denoted by \( F_0^* \).
Equation 39 is then integrated at $n^* = 0.001$, using the approximation $g = e^{-3\phi}$ at this low density. The result will be

$$F^* - F_0^* = -\int_{0.001}^{T^*} \left(1 - \frac{9.5 \times 10^{-3} T}{9T^*}\right) \frac{dT^*}{T^*}. \quad (42)$$

In the proceeding steps $F^* - F_0^*$ will be denoted as $F^* (n^* = 0.001, T^*)$.

2) $F_T^* (n^* = 0.001, T^*)$ can now be computed using equations 40 and 41, resulting in

$$F_T^*(n^*=0.001,T^*) = F^*(n^*=0.001,T^*) - \left(1 - \frac{T^*}{T^*}\right) \left(1 - U_T^*\right). \quad (43)$$

3) Equation 39 is integrated again at a constant $T^*$. $F_T^* (n^*, T^*)$ can then be computed by the following equation,

$$F_T^* (n^*, T^*) = F_T^*(n^* = 0.001, T^*) + \int_{0.001}^{n^*} \left(\frac{2P^*}{3n^*}\right) dn^*. \quad (44)$$

For low densities ($n^* \leq 0.10$), the values of $P^*$ are interpolated values in the form

$$P^* = 1 + b_1n^* + b_2n^*^2 + b_3n^*^3. \quad (45)$$

The integral can then be done analytically. At higher densities, interpolated values of $P^*$ are in the form of equation 32. Here numerical integration must be used.

4) $F_p^* (n^*, T^*)$ can be computed directly from equation 41.
5) Finally, with $F_r^*$ and $F_p^*$ known, $F^*(n^*, T^*)$ is obtained from the relationship

$$F^* = F_r^* + F_p^*. \quad (46)$$

6) Numerical differentiation is then used to compute $P^*$ and $U^*$ as

$$P^* = \frac{3}{2} n^* \left( \frac{\partial F^*}{\partial n^*} \right), \quad (47)$$

and

$$U = - T^* \left( \frac{\partial F^*}{\partial T^*} \right). \quad (48)$$

7) Other thermodynamic functions can easily be obtained by the following thermodynamic relationships$^{22}$

$$S^* = 2 S/3Nk = U^* - F^*, \quad (49)$$

$$H^* = 2 H/3NkT = U^* + (2/3) P^*, \quad (50)$$

and

$$G^* = 2 G/3NkT = F^* + (2/3) P^*, \quad (51)$$

where $S$ is the entropy, $H$ is the enthalpy, and $G$ is the Gibbs energy.
CHAPTER IV

RESULTS AND COMPARISONS

Parametric Integral Equation N and The Lennard-Jones Potential

For the Lennard-Jones potential, equation N was solved for the isotherm $T^* = 1.6$ at reduced densities of $n^* = 0.70$ and $n^* = 0.90$ for several different values of the parameter $a$. Results of $P^*$ and $U^*$ obtained can be found in the appendix. Graphs of $P^*$ versus $a$ were then drawn. The accepted values for the isotherm $T^* = 1.6$ are $P^* = 1.77$ at $n^* = 0.70$, and $P^* = 4.61$ at $n^* = 0.90$ (Results by Carley using integral equations combined with perturbation theory). The accepted values were also shown on the graphs (see figure 2).

It can be seen clearly that there is no single parameter $a$ that give $P^*$ values equal to the accepted $P^*$ values at both the reduced densities. Thus equation N does not produce good results.
Figure 2. $P^*$ Versus $a$ for $T^* = 1.6$ at $n^* = 0.70$ and $0.90$ (Equation N)
For the square-well, equation T was solved at T* = 1.4, 1.6, and 2.2 which were above the critical temperature. These choices of T* were to keep the perturbation as small as possible while still providing good thermodynamic values for the reference system (for the perturbation theory computations).

The first thing that was done was to solve equation T for T* = 1.4 at n* = 0.70, and n* = 0.88 with different sets of the parameters b_2 and b_3. At n* = 0.88 the results of P* obtained were unsatisfactory. Computations were done again at n* = 0.85 and the results of P* obtained were still unsatisfactory. The same problem occurred when equation T was solved at T* = 1.6. Therefore at T* = 1.4 and 1.6, equation T was found to be unsatisfactory. The results of P* and U* can be found in the appendix.

Equation T was then solved for the isotherm T* = 2.2 at n* = 0.65, and n* = 0.85 with different sets of the parameters b_2 and b_3. The P* and U* results obtained were satisfactory and can be found in the appendix. The results were then used with a least squares program to find the coefficients in equation 31. Table 1 gives the values of these coefficients.
Table 1
Coefficients for Least Square Equation at $T^* = 2.2$

<table>
<thead>
<tr>
<th>$n^*$</th>
<th>$c_1$</th>
<th>$c_2$</th>
<th>$c_3$</th>
<th>$c_4$</th>
<th>$c_5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.65</td>
<td>0.819257</td>
<td>0.825071</td>
<td>-0.573483</td>
<td>-0.39785</td>
<td>-1.14248</td>
</tr>
<tr>
<td>0.85</td>
<td>5.70092</td>
<td>11.345</td>
<td>0.193985</td>
<td>-23.4007</td>
<td>-14.7053</td>
</tr>
</tbody>
</table>

The coefficients were used to compute various values of $b_2$ and $b_3$ that give $P^*$ results equal to the accepted $P^*$ results which were taken from the extrapolated and interpolated results of Alder. Graphs of $b_3$ versus $b_2$ were then drawn for both the reduced densities. The point of interception ($b_2 = -0.517$, $b_3 = 0.208$) gives the best choice of parameters (see figure 3).

The best choice of parameters was then used to solve equation $T$ again at ten other densities along the isotherm. The results are given in Table 3. Here, values of $P^*$ and $U^*$ are rounded off and only the 297 points are taken. A more complete result can be found in the appendix.

Results in Table 3 were applied to the interpolation formulas for $P^*$ and $U^*$ (equations 32 and 33) to obtain the Pade coefficients. Other interpolated results of $P^*$ and $U^*$ were then easily obtained without going through the long procedure again. The Pade coefficients are given in Table 4 and the other interpolated results of $P^*$.
and $U^*$ are given in Table 5.

The results in Table 5 were used to plot graphs of $P^*$ versus $n^*$, and $U^*$ versus $n^*$. Interpolated and extrapolated results of Alder were also plotted on the graphs for comparison (see figure 4 and figure 5). The agreement is quite good, thus equation T produces good results.
Table 2
Equation T Results at $T^* = 2.2$
($b_2 = 0.208, b_3 = -0.517$)

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Table 3
Pade Coefficients for $P^*$ and $U^*$ for $T^* = 2.2$

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Figure 4. $P^*$ versus $n^*$ at $T^* = 2.2$ The Points are the Accepted Results of Alder
Figure 5. $U^*$ versus $n^*$ at $T^* = 2.2$. The Points are the Accepted Results of Alder.
Perturbation Theory Applied to the Square-Well Potential

A computer program was used to compute the thermodynamic functions. The program was modified to do computations at a reference temperature of $T^* = 2.2$. Results of the reduced internal energy (Table 5) and the Pade coefficients for the pressure (Table 4) were used in the program for the reference part computations.

The thermodynamic functions obtained were for $0.7 \leq T^* \leq 2.15$ and $0.05 \leq n^* \leq 0.85$ for an interval of 0.05. The results of $T^* = 0.85, 1.05, 1.35, 1.55,$ and 2.10 will only be discussed here. A complete result of the thermodynamic functions can be found in the appendix.

Graphs of $P^*$ versus $n^*$ and $U^*$ versus $n^*$ were plotted for the selected $T^*$. As before, interpolated and extrapolated results of Alder were shown in the graphs for comparisons. Results at $n^* = 0.05$ were dropped from the graphs because of numerical error in the numerical differentiation computations of $P^*$ and $U^*$. These graphs are shown in figures 6-9. In addition, graphs of $F^*, S^*, H^*, \text{ and } G^*$ versus $n^*$ were also plotted (figures 10-13).

From these graphs, it can be seen that the $P^*$ results agree quite well with the accepted results. However, the $U^*$ results do not agree with the accepted results at lower temperatures but agree very well at higher temperatures.
For further comparisons, graphs of $P^*$ and $U^*$ versus $n^* = 0.85$, 0.65, and 0.45 were plotted together with the interpolated and extrapolated results of Alder (figures 14-19).

Again, it can be seen that good agreements with the accepted results are only at higher temperatures.

![Graph showing $P^*$ versus $n^*$ for $T^* = 0.85$, 1.05, and 1.35.](image)

Figure 6. $P^*$ versus $n^*$ for $T^* = 0.85$, 1.05, and 1.35. The Numbers Near the Curves are the Temperatures ($T^*$) and the Points are the Accepted Results of Alder.
Figure 7. $P^*$ versus $n^*$ for $T^* = 1.55$ and $2.10$. The Numbers Near the Curves are the Temperatures ($T^*$) and the Points are the Accepted Results of Alder.
Figure 8. $U^*$ versus $n^*$ for $T^*$ = 0.85, 1.05, and 1.35. The Numbers Near the Curves are the Temperatures ($T^*$) and the Points are the Accepted Results of Alder.
Figure 9. $U^*$ versus $n^*$ for $T^* = 1.55$ and 2.10. The Numbers Near the Curves are the Temperatures ($T^*$) and the Points are the Accepted Results of Alder.
Figure 10. $F^*$ versus $n^*$ for Constant $T^*$. The Numbers Near the Curves are the Temperatures ($T^*$).

Figure 11. $S^*$ versus $n^*$ for Constant $T^*$. The Numbers Near the Curves are the Temperatures ($T^*$).
Figure 12. $H^*$ versus $n^*$ for Constant $T^*$. The Numbers Near the Curves are the Temperatures ($T^*$)
Figure 13. $G^*$ versus $n^*$ for Constant $T^*$. The Numbers Near the Curves are the Temperatures ($T^*$)
Figure 14. $P^*$ versus $T^*$ for $n^* = 0.85$. The Points are the Accepted Results of Alder.

Figure 15. $P^*$ versus $T^*$ for $n^* = 0.65$. The Points are the Accepted Results of Alder.
Figure 16. $p^*$ versus $T^*$ for $n^* = 0.45$. The Points are the Accepted Results of Alder

Figure 17. $u^*$ versus $T^*$ for $n^* = 0.85$. The Points are the Accepted Results of Alder
Figure 18. $U^*$ versus $T^*$ for $n^* = 0.65$. The Points are the Accepted Results of Alder

Figure 19. $U^*$ versus $T^*$ for $n^* = 0.45$. The Points are the Accepted Results of Alder
CHAPTER V

SUMMARY AND CONCLUSIONS

In the first part of this research, equation N, a one parameter integral equation, was solved for a Lennard-Jones potential at $T^* = 1.6$. It was found out that there was no single parameter that produced $P^*$ results equal to the accepted $P^*$ results at a high and intermediate density. Thus equation N results have no better improvement over equation C.

In the second part of this research, equation T was solved for a square-well potential at $T^* = 1.4, 1.6,$ and $2.2$. The choices of $T^*$ were to keep the perturbation as small as possible but still provide good thermodynamic values. At $T^* = 1.4$ and $1.6$, the best choice of parameters were difficult to determine and thus the $P^*$ results obtained were unsatisfactory. However at $T^* = 2.2$, the $P^*$ and $U^*$ results obtained were in good agreement with the accepted results.

The last part of this research was to compute the thermodynamic functions of a square-well potential using the first order perturbation theory. Equation T results at $T^* = 2.2$ were used as the reference system. In summary, the agreement with the accepted results was...
found to be fairly good for $P^*$ results in all the
densities tried ($n^* = 0.45, 0.65, \text{ and } 0.85$) and for $U^*$
the results deviate from the accepted results for large
perturbations.
REFERENCES


4. Ibid.


9. Ibid.


11. Ibid.

12. Ibid.

13. Ibid., p. 269.

15. Ibid.


20. Carley and Dotson, loc. cit.

21. Ibid.


APPENDICES
APPENDIX A

SOLUTIONS OF EQUATION N FOR THE LENNARD-JONES POTENTIAL

The following notations are used:

- \( a \)  
  Value of the parameter.

- \( n^* \)  
  Value of the reduced density.

- \( T^* \)  
  Value of the reduced temperature.

- \( N \)  
  An A in the column indicates the computer gives solutions for 75 points with an x interval of 0.075. A B indicates solutions for 297 points with an x interval of 0.01875.

- \( SDSS \)  
  A measurement of the difference between the final guessed value for \( S \) and computed value of \( S \). A smaller value of \( SDSS \) shows a better convergence.

- \( P^* \)  
  Value of the reduced pressure.

- \( U^* \)  
  Value of the reduced internal energy.
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APPENDIX B

SOLUTIONS OF EQUATION T FOR THE SQUARE-WELL POTENTIAL

The following notations are used:

- $b_2, b_3$: Values of the parameters.
- $n^*$: Value of the reduced density.
- $T^*$: Value of the reduced temperature.
- $N$: An A in the column indicates the computer gives solutions for 75 points with an $x$ interval of 0.07142857. A B indicates solutions for 297 points with an $x$ interval of 0.01785714.
- SDSS: A measurement of the difference between the final guessed value for $S$ and computed value of $S$. A smaller value of SDSS shows a better convergence.
- $P^*$: Value of the reduced pressure.
- $U^*$: Value of the reduced internal energy.
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APPENDIX C

COMPUTER PROGRAM

This appendix gives a listing of the computer program that was used to compute the thermodynamic functions of a square-well potential using perturbation theory.
57

10 PRINT "SUPT.BAS-THIS PROGRAM COMPUTES THE THERMODYNAMIC FUNCTIONS*
20 PRINT "FOR A SQUARE-UELL LIQUID USING PERTURBATION THEORY."
30 DATA 0.9976260, 0.9763320, 0.95544, 0.94493, 0.93345, 0.92395, 0.91456
40 REM ENTER UR(REF INT ENER) T1(TEMP) A1,...,A6(FADE COEF)
50 FOR I = 1 TO 19
60 READ UR(I)
70 NEXT I
80 DATA 0.019, -0.096, -0.212, -0.326, -0.434, -0.537, -0.634, -0.725, -0.812
90 DATA -2.335/7, -2.22295, -1.90424, -1.58833, -0.79671
100 DATA 0.019, -0.096, -0.212, -0.326, -0.434, -0.537, -0.634, -0.725
110 READ T1
120 DATA 2.2
130 READ A1, A2, A3, A4, A5, A6
140 DATA -2.335/7, 4.22295, -1.90424, -1.58833, -0.79671, 117877
150 REM CALCULATION OF F(0.001,TSTAR)
160 FOR J = 1 TO 30
170 T2 = .65 + J*.05
180 F(1,J) = LOG(T2) - 3.316125579*.001*(EXP(1/T2) - EXP(1/20))
190 NEXT J
200 REM CALCULATION OF FR(.001,TSTAR)
210 FOR J = 1 TO 30
220 T2 = .65 + J*.05
230 FR(1,J) = F(1,J) - (1 - (T1/T2))*(1 - UR(1))
240 NEXT J
250 REM CALCULATION OF FR(NSTAR,TSTAR)
260 PRINT 'TYPE THE NO OF INT F'TS*
270 INPUT N1
280 B1 = A1 - A4
290 B2 = A2 - A5 - A4*A1 + A4*A4
300 B3 = A3 - A6 - A5*A1 + A5*A4 - A4*A2 + A4*A5 - A4*A4*A1 + A4*A4*A4
310 DEF FNX(X) = (2/3)*(LOG(X/.001) + (X - .001) + (B2/2)*(X*X - .000001) + (B3/3)*(X*X*X - .00000001))/(1 + A4*X + A5*X*X + A6*X*X*X)
320 DEF FNX2(X) = (1 + A1*X + A2*X*X + A3*X*X*X)/(1 + A4*X + A5*X*X + A6*X*X*X)
330 D = .05/(N1 - 1)
340 G(1) = FNX(.001)
350 G(2) = FNX(.01)
360 G(3) = FNX(.05)
370 G(4) = FNX(.1)
380 FOR J = 5 TO 19
390 X0 = (J - 3)*.05
400 FOR J = 1 TO N1
410 X = X0 + (J - 1)*D
420 S = S + FNX2(X)/X
430 NEXT J
440 S = S + .5*((FNX2(X0)*X0) + FNX2(X0+.05))/X0+.05
450 S = (2*S*D)/D
460 G(1) = G(1) + S
470 S = 0
480 NEXT J
490 FOR J = 1 TO 30
500 FOR I = 1 TO 19
510 F(I,J) = G(I) + FR(I,J)
520 NEXT I
530 NEXT J
540 REM CALCULATION OF FP(NSTAR,TSTAR)
550 FOR J = 1 TO 30
560 FOR I = 1 TO 19
570 T2 = .65 + J*.05
580 FP(I,J) = (1 - T1/T2)*(1 - UR(I))
590 NEXT J
600 NEXT J
610 REM CALCULATION OF F(NSTAR,TSTAR)
620 FOR J = 1 TO 30
630 FOR I = 1 TO 19
640 F(I,J) = FR(I,J) + FP(I,J)
650 NEXT I
660 NEXT J
670 REM CALCULATION OF P(NSTAR,TSTAR)
680 DN = .05
690 DT = .05
700 CH = 1/(12*DN)
710 CT = 1/(12*DT)
720 FOR J = 1 TO 30
730 F(3,J) = 1.5*SINCH(-25*F(3,J)+49*F(4,J)-36*F(5,J)+16*F(6,J)-38*F(7,J))
740 F(4,J) = 1.5*SINCH(-35*F(3,J)-10*F(4,J)+18*F(5,J)-69*F(6,J)+9*F(7,J))
750 FOR I = 5 TO 17
760 D = (I - 2)*.05
770 F(I,J) = 1.5*CHNF(F(I-2,J)-5*F(I-1,J)+8*F(I+1,J)-F(I+2,J))
780 NEXT I
790 P(18, J) = 1.5#B#CN#:F(15, J)-6#F(16, J)-18#F(17, J)+10#F(18, J)+25#F(19, J)
800 P(19, J) = 1.5#B#CN#:F(15, J)-6#F(16, J)-16#F(17, J)+36#F(18, J)-48#F(19, J)
810 NEXT J
820 REM CALCULATION OF U(NSTAR, TSTAR)
830 FOR I = 3 TO 19
840 U(I, 1) = -.75#CT#:F(I, 1)+48#F(I, 2)-36#F(I, 3)-16#F(I, 4)+36#F(I, 5)
850 U(I, 2) = -.75#CT#:F(I, 1)-10#F(I, 2)+18#F(I, 3)-6#F(I, 4)+F(I, 5)
860 FOR J = 3 TO 28
870 T2 = .65 + J*.05
880 U(I, J) = -T2#CT#:F(I, J-2)-B#F(I, J-1)+8#F(I, J+1)-F(I, J+2)
890 NEXT J
900 U(I, 29) = -.25#CT#:F(I, 26)+6#F(I, 27)-18#F(I, 28)+10#F(I, 29)+3#F(I, 30)
910 U(I, 30) = -.25#CT#:F(I, 26)-16#F(I, 27)+36#F(I, 28)-48#F(I, 29)+25#F(I, 30)
920 NEXT I
930 REM PRINT OUT OF N*, F*, P*, U*
940 FOR J = 1 TO 30
950 T2 = .65 + J*.05
960 PRINT
970 PRINT *TSTAR = *"T2*
980 PRINT "N-STAR", "F-STAR", "P-STAR", "U-STAR"
990 FOR I = 3 TO 19
1000 D = (I - 2)*.05
1010 PRINT D:F(I, J)-P(I, J)+U(I, J)
1020 NEXT I
1030 NEXT J
1040 REM PRINT OUT OF N*, S*, H*, G*
1050 FOR J = 1 TO 30
1060 T2 = .65 + J*.05
1070 PRINT
1080 PRINT *T-STAR = *"T2*
1090 PRINT "N-STAR", "S-STAR", "H-STAR", "G-STAR"
1100 FOR I = 3 TO 19
1110 D = (I - 2)*.05
1120 PRINT D:U(I, J)-F(I, J)+2#P(I, J)/3:F(I, J)+2#F(I, J)/3
1130 NEXT I
1140 NEXT J
1150 END
APPENDIX D

RESULTS OF THERMODYNAMIC FUNCTIONS

The following notations are used:

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<td>Value of the reduced density.</td>
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<td>Value of the reduced Helmholtz free energy.</td>
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BIBLIOGRAPHY


