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AD 44174 (5cys) _____
 Errata for the Report WIS-ONR-12 "The Optimum Free
 Theory of Liquids", 15 October 1954.

- (1) In equation (7) and (8) replace all ρ_{ij} by ρ_{ij}' .
- (2) The function $\left[\frac{4z^4}{15} \Gamma(9; y, y_j) - z^2 \Gamma(3; y, y_j) \right]$ occurring in equation (13), (15), (16), (21), and (22) should be replaced by $\left[\frac{z^4}{15} \Gamma(9; y, y_j) - z^2 \Gamma(3; y, y_j) \right]$.
- (3) The factor multiplying the bracket in equation (13) should be $1 / (12 y^{1/2})$.
- (4) The factor multiplying the integrals in equations (15) and (21) should be $\pi / y^{1/2}$.
- (5) The factor multiplying the integrals in equation (16) and (22) should be $2 \pi^2$. _____

THE OPTIMUM FREE VOLUME THEORY OF LIQUIDS *

by

John S. Dahler[§] and Joseph O. Hirschfelder

ABSTRACT

An iterative method for solving Kirkwood's integral equation for the free-volume of a liquid is described. This procedure uses the results of the well-known Lennard-Jones and Devonshire theory of liquids as its starting point. The results of the calculations suggested would provide a good test of the validity of the cell theory of liquids. It is believed that this "optimum free-volume theory" will remove several of the undesirable characteristics of the Lennard-Jones and Devonshire liquid.

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[§] General Motors Corporation Fellow, 1954-55.

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THE OPTIMUM FREE VOLUME THEORY OF LIQUIDS

by

John S. Dahler and Joseph O. Hirschfelder

The following report summarizes a method of successive approximations to the "optimum" free volume theory of liquids. This approach is reminiscent of the self-consistent field approximation in molecular quantum mechanics. The initial approximation is the usual Lennard-Jones and Devonshire equation of state.^{1,2} Detailed equations and procedures are set up for determining the equation of state and the related thermodynamic properties. It is hoped that such calculations can be carried out in the normal liquid, as well as the critical regions.

Kirkwood³ has suggested a method for improving the Lennard-Jones and Devonshire "cell theory" of liquids. However, no calculations based upon this extension of the earlier theory have ever been carried out. Qualitatively, this theory is not too different from that of Lennard-Jones and Devonshire, in that each molecule is restricted to move within a cell bounded by other molecules lying near the centers of equivalent cells composing some simple crystalline lattice. Although the shape and orientation of the cells are arbitrary, we choose dodecahedral cells centered on the sites of a face-centered-cubic lattice, whose "nearest neighbor separation" is given by d_0 .

Let us adopt the notation (see Figure 1):

- (a) $\underline{R}_{1j}$ is the vector from lattice site 1 to lattice site j .
- (b) $\underline{r}_1, \underline{r}_j$ are the displacements of the first and j th molecules, respectively, from their sites.

- - - - -
1. J. E. Lennard-Jones, A. F. Devonshire, Proc. Roy. Soc. (London) A163, 53 (1937)
 2. MTGL, p. 293
 3. J. G. Kirkwood, J. Chem. Phys. 18, 380 (1950)

where all distances are measured in units of the "nearest neighbor separation", d_0 .

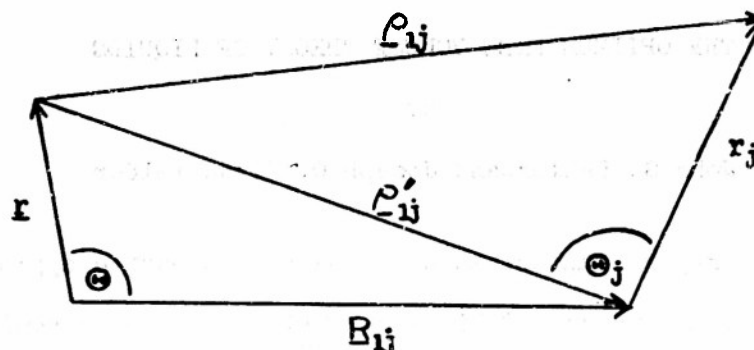


Figure 1

For present purposes, V_{1j} , the pair interaction energy of molecules 1 and j, in units of ϵ , is represented by a Lennard-Jones 12-6 potential, e.g.,

$$V_{1j}(\rho_{1j}) = 4 \left[\left(\frac{\sigma}{\rho_{1j}} \right)^{12} - \left(\frac{\sigma}{\rho_{1j}} \right)^6 \right] \quad (1)$$

where σ and ϵ are adjustable parameters. We define the "reduced temperature" T^* , by $T^* = kT/\epsilon$.

In Kirkwood's theory the "free-volume", v_f , is given by,

$$v_f = \int_{\Delta} \exp(-\psi(\underline{r})/T^*) d\underline{r} \quad (2)$$

where $\psi(\underline{r})$ is the solution of the integral equation,

$$\psi(\underline{r}) = \phi(\underline{r}) - E_0 \quad (3)$$

and

$$\phi(\underline{r}) = \sum_{j>1} \int_{\Delta_j} V_{1j}(\rho_{1j}) s(\underline{r}_j) d\underline{r}_j \quad (4)$$

$$\begin{aligned} s(\underline{r}) &= v_f^{-1} \exp(-\psi(\underline{r})/T^*) \\ &= \exp(-\phi(\underline{r})/T^*) / \int_{\Delta} \exp(-\phi(\underline{r})/T^*) d\underline{r} \end{aligned} \quad (5)$$

$$E_0 = \int_{\Delta} \phi(\underline{r}) s(\underline{r}) d\underline{r} \quad (6)$$

The integrations are carried out over cells of volume Δ . In practice, it is necessary to replace the functions, $s(\underline{r})$, by "smoothed" functions obtained by averaging over all orientations of the vector $\underline{r}$; and the cell, Δ , by an "equivalent sphere". If, as a first approximation, we replace $s(\underline{r})$ by a Dirac delta function, $\delta(\underline{r})$, then we obtain the results of the Lennard-Jones and Devonshire theory. We propose using this as the first step in an iterative process, to generate the "optimum" free volume. Fortunately, this procedure is eminently suited to existing high speed computing machines such as the OARAC at Wright-Patterson Air Force Base.

When the $s(\underline{r})$'s have spherical symmetry about the lattice sites, the integrations over angles can be performed exactly. Thus we have,

$$\phi(r) = 4\pi \sum_{j>1} \int_{\Delta_j} s(\underline{r}_j) F(\rho_{1j}, r_j) r_j d\mathbf{r}_j \quad (7)$$

where

$$F(\rho_{1j}, r_j) = \frac{1}{\rho_{1j}^3} \left[\frac{\sigma^{12}}{5} \{ (\rho_{1j} - r_j)^{-10} - (\rho_{1j} + r_j)^{-10} \} - \frac{\sigma^6}{2} \{ (\rho_{1j} - r_j)^{-4} - (\rho_{1j} + r_j)^{-4} \} \right] \quad (8)$$

To calculate E_0 and also to average $\phi(\underline{r})$ over angles, we need the result:

$$\begin{aligned} H(R_{1j}; r, r_j) &= \int_0^\pi \sin \Theta F(\rho'_{1j}, r_j) d\Theta \\ &= \frac{1}{3rR_{1j}} \left[\frac{\sigma^{12}}{15} \{ (r_j + R_{1j} + r)^{-9} - (r_j + R_{1j} - r)^{-9} - (r_j - R_{1j} + r)^{-9} + (r_j - R_{1j} - r)^{-9} \} \right. \\ &\quad \left. - \frac{\sigma^6}{2} \{ (r_j + R_{1j} + r)^{-3} - (r_j + R_{1j} - r)^{-3} - (r_j - R_{1j} + r)^{-3} + (r_j - R_{1j} - r)^{-3} \} \right] \end{aligned} \quad (9)$$

for $r < R_{1j}$ and $r_j < R_{1j}$.

The "sums over all molecules in the system except the first" can be re-grouped into "sums over shells of neighbors in the lattice". We denote the separation of the kth shell from the lattice site of "the first molecule" by

a_k , and the number of such neighbors as n_k . If we denote $\overline{\phi(r)}$ as the result of averaging $\phi(r)$ over all orientations of r , then

$$\phi(r) = 2\pi \sum_k n_k \int_{\Delta_k} s(r_k) H(a_k; r, r_k) r_k dr_k \quad (10)$$

$$\begin{aligned} E_0 &= 4\pi \int_{\Delta} s(r) \overline{\phi(r)} r^2 dr \\ &= 8\pi^2 \sum_k n_k \int_{\Delta} \int_{\Delta_k} s(r_k) s(r) H(a_k; r, r_k) r^2 r_k dr dr_k \end{aligned} \quad (11)$$

where $s(r_k)$ and r_k refer to any molecule in the k th shell.

In the case of the Lennard-Jones and Devonshire equation of state it has proved useful to introduce the new variables $y = r^2$ and $y_j = r_j^2$. The transformed relations become:

$$s(r) = s(y) = \exp(-\overline{\phi(y)}/T^*) / 2\pi \int_{\Delta} \exp(-\overline{\phi(y)}/T^*) y^{1/2} dy, \quad (12)$$

$$H(1; y, y_j) = \frac{1}{3y^{1/2}} \left[\frac{4z^4}{15} \Gamma(9; y, y_j) - z^2 \Gamma(3; y, y_j) \right], \quad (13)$$

where the "reduced density", $z = \sigma^3 \sqrt{2}$, is the number of molecules in a volume $(\sigma d_0)^3$, and

$$\Gamma(n; y, y_j) = \left[(y_j^{1/2} + 1 + y^{1/2-n}) - (y_j^{1/2} + 1 - y^{1/2-n}) - (y_j^{1/2} - 1 + y^{1/2-n}) + (y_j^{1/2} - 1 - y^{1/2-n}) \right] \quad (14)$$

The contributions of the first shell of neighbors for the face-centered-cubic lattice can then be written as,

$$\overline{\phi(y)}^{(1)} = \frac{1}{y^{1/2}} \int_{\Delta_j} s(y_j) \left[\frac{4z^4}{15} \Gamma(9; y, y_j) - z^2 \Gamma(3; y, y_j) \right] dy_j \quad (15)$$

$$E_0^{(1)} = \int_{\Delta} \int_{\Delta_j} s(y) s(y_j) \left[\frac{4z^4}{15} \Gamma(9; y, y_j) - z^2 \Gamma(3; y, y_j) \right] dy dy_j \quad (16)$$

Previous calculations have shown that the contributions to $\overline{\phi}(y)$ and E_0 by the nearest neighbors is far more important than for the other shells. Thus, little error will result from treating only the nearest neighbors by the more detailed theory and using the Lennard-Jones and Devonshire for the remaining shells.

For a face-centered-cubic lattice, the Lennard-Jones and Devonshire theory gives,

$$[\overline{\phi}(y) - E_0]_{LJD} = 4 \sum_k n_k \left[\frac{4z^4}{k^6} l(y/k) - \frac{2z^2}{k^3} m(y/k) \right] \quad (17)$$

$$[E_0]_{LJD} = 4 \sum_k n_k \left[\frac{4z^4}{k^6} - \frac{2z^2}{k^3} \right] \quad (18)$$

where

$$l(t) = (1 + 12t + 25.2t^2 + 12t^3 + t^4) (1 - t)^{-10} - 1$$

$$m(t) = (1 + t) (1 - t)^{-4} - 1$$

are functions which have been previously investigated by several workers.^{4,5}

Thus, the contributions of the 2nd and 3rd shells of neighbors are given by,

$$[\overline{\phi}(y) - E_0]_{LJD}^{(2,3)} = 96 \left[z^4 \left\{ \frac{l(y/2)}{2^6} + \frac{4l(y/3)}{3^6} \right\} - z^2 \left\{ \frac{m(y/2)}{2^4} + \frac{6m(y/3)}{3^4} \right\} \right] \quad (19)$$

$$[E_0]_{LJD}^{(2,3)} = 24 \left[z^4 \left\{ \frac{1}{16} + \frac{16}{3^6} \right\} - z^2 \left\{ \frac{1}{4} + \frac{8}{27} \right\} \right] \quad (20)$$

Because of the short range of the intermolecular forces, the effect of the more distant shells is completely negligible. Combining the results obtained above we have,

$$\begin{aligned} \overline{\phi}(y)^{(1,2,3)} &= \frac{1}{y^{1/2}} \int_{\Delta_j} s(y_j) \left[\frac{4z^4}{15} \Gamma(9; y, y_j) - z^2 \Gamma(3; y, y_j) \right] dy_j \\ &+ 96 \left[z^4 \left\{ \frac{l(y/2)+1}{2^6} + \frac{4(l(y/3)+1)}{3^6} \right\} - z^2 \left\{ \frac{m(y/2)+1}{2^4} + \frac{6(m(y/3)+1)}{3^4} \right\} \right] \end{aligned} \quad (21)$$

4. Wentorf, Buehler, Hirschfelder, Curtiss, J. Chem. Phys. 18, 1484 (1950)

5. Kirkwood, Salsburg, J. Chem. Phys. 21, 2169 (1953)

$$E_o^{(1,2,3)} = \iint_{\Delta \Delta_j} s(y)s(y_j) \left[\frac{4z^4}{15} \Gamma(9; y, y_j) - z^2 \Gamma(3; y, y_j) \right] dy dy_j$$

$$+ 24 \left[z^4 \left(\frac{1}{16} + \frac{16}{36} \right) - z^2 \left(\frac{1}{4} + \frac{8}{27} \right) \right] \quad (22)$$

where $s(y)$ is now the solution of the modified integral equation,

$$\overline{\Psi}(y)^{(1,2,3)} = \overline{\Phi}(y)^{(1,2,3)} - E_o^{(1,2,3)} \quad (23)$$

The equation of state of the liquid and its internal energy per molecule in units of ϵ are then given by,

$$\frac{pV}{RT} = \frac{z}{T^*} \left[\frac{\Delta \int \exp(-\overline{\Phi}(y)/T^*) \frac{\partial \overline{\Phi}(y)}{\partial z} y^{1/2} dy}{\Delta \int \exp(-\overline{\Phi}(y)/T^*) y^{1/2} dy} - \frac{1}{2} \frac{\partial E_o}{\partial z} \right] \quad (24)$$

$$U = 3T^*/2 - E_o/2 + T^*/2 \frac{\partial E_o}{\partial T^*} + \left[\frac{\Delta \int \exp(-\overline{\Phi}(y)/T^*) \overline{\Phi}(y) y^{1/2} dy}{\Delta \int \exp(-\overline{\Phi}(y)/T^*) y^{1/2} dy} \right] - T^* \left[\frac{\Delta \int \exp(-\overline{\Phi}(y)/T^*) \frac{\partial \overline{\Phi}(y)}{\partial T^*} y^{1/2} dy}{\Delta \int \exp(-\overline{\Phi}(y)/T^*) y^{1/2} dy} \right] \quad (25)$$

SOLUTION OF THE INTEGRAL EQUATION. (Eq. 23), BY AN ITERATIVE PROCEDURE:

For a particular choice of the density, z , and the temperature, T^* , we can obtain the "optimum function $s(y)$ " by an iterative procedure. For an initial trial function, $s(y)_0$, the first step in this iteration leads to a "first improved" trial function, $s(y)_1$. The next step, or cycle, of the iteration leads to a "second improved" trial function, $s(y)_2$; etc. ... When the thermodynamic properties calculated from the "nth improved" trial solution do not differ sensibly from those calculated from the (n+1)th, we have obtained the "optimum function, $s(y)_n$ ". In this sense, the procedure is quite similar to the "self-consistent field calculations" of atomic and molecular quantum mechanics.

The cyclic procedure described above can be illustrated in the following manner:

CYCLE 1:

- (a) Choose a trial solution called $s(y)_0$.
- (b) Substituting $s(y)_0$ for $s(y)$ in Eq. 21 and performing the indicated integration, we calculate the corresponding function, $[\overline{\phi(y)}^{(1,2,3)}]_0$.
- (c) Substituting $[\overline{\phi(y)}^{(1,2,3)}]_0$ for $\overline{\phi(y)}^{(1,2,3)}$ in Eq. 12 and performing the indicated integration, we obtain the "first improved" function,

$$s(y)_1 = \frac{\exp(-[\overline{\phi(y)}^{(1,2,3)}]_0/T^*)}{2\pi \int_{\Delta} \exp(-[\overline{\phi(y)}^{(1,2,3)}]_0/T^*) y^{1/2} dy}$$

CYCLE 2:

- (a) Substituting $s(y)_1$ for $s(y)$ in Eq. 21 and performing the indicated integration, we calculate the corresponding function, $[\overline{\phi(y)}^{(1,2,3)}]_1$.
- (b) Substituting $[\overline{\phi(y)}^{(1,2,3)}]_1$ for $\overline{\phi(y)}^{(1,2,3)}$ in Eq. 12 and performing the indicated integration, we obtain the "2nd improved" function,

$$s(y)_2 = \frac{\exp(-[\overline{\phi(y)}^{(1,2,3)}]_1/T^*)}{2\pi \int_{\Delta} \exp(-[\overline{\phi(y)}^{(1,2,3)}]_1/T^*) y^{1/2} dy}$$

CYCLE 3:

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CYCLE n:

- (a)
- (b) Substituting $[\overline{\phi(y)}^{(1,2,3)}]_{n-1}$ for $\overline{\phi(y)}^{(1,2,3)}$ in Eq. 12 and performing the indicated integration, we obtain the "nth improved" function,

$$s(y)_n = \frac{\exp(-[\overline{\phi(y)}^{(1,2,3)}]_{n-1}/T^*)}{2\pi \int_{\Delta} \exp(-[\overline{\phi(y)}^{(1,2,3)}]_{n-1}/T^*) y^{1/2} dy}$$

This iterative procedure must be carried out separately for each pair of the parameters z and T^* . Now, to obtain the thermodynamic properties corresponding to the "kth improved" function, we must calculate: (i) $[E_o^{(1,2,3)}]_k$ by substitution of $s(y)_k$ for $s(y)$ in Eq. 22, and (ii) the derivatives of $[\phi(y)^{(1,2,3)}]_k$ and $[E_o^{(1,2,3)}]_k$ arising in Eqs. 24 and 25. If the above iterative process is carried out simultaneously for several choices of z and/or T^* in the neighborhood of the values in question, we can then determine the derivatives with respect to these parameters by standard procedures.

As stated earlier, we shall choose $s(y)_0 = \delta(y)$, where $\delta(y)$ is the Dirac delta function. This choice leads to the "first improved" function,

$$s(y)_1 = \frac{\exp(-12[z^4 L(y) - 2z^2 M(y)]/T^*)}{2\pi G(z, T^*)} \quad (26)$$

where,

$$L(t) = l(t) + \frac{1}{128} l(t/2) + \frac{2}{729} l(t/3)$$

$$M(t) = m(t) + \frac{1}{16} m(t/2) + \frac{2}{27} m(t/3)$$

$$G(z, T^*) = \int_0^{.30544} \exp(-12[z^4 L(y) - 2z^2 M(y)]/T^*) y^{1/2} dy$$

are functions which have been extensively tabulated by several workers.^{4,5,6}

Here, as in subsequent calculations, the integration interval is chosen to be $(0, .30544)$.

The iteration from this point on follows the scheme outlined above.

6. Unpublished calculations

INTEGRALS:

To carry out the proposed iterative solution of the integral equation, many difficult integrals must be calculated numerically. Recently, we have calculated $G(z, T^*)$ and several related integrals such as $\partial G(z, T^*)/\partial z$ and $\partial G(z, T^*)/\partial T^*$ using the University of Wisconsin Card Programmed Calculator, Model II. The integrands, $I(y)$, involved in these calculations, all had the same general characteristics shown in Figure 2.

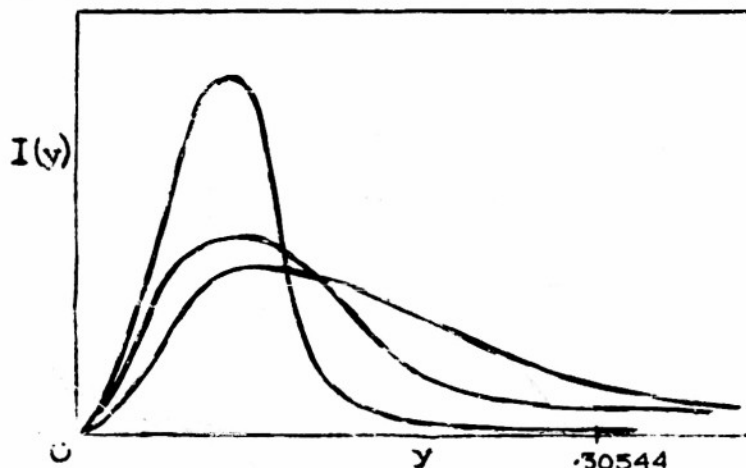


Figure 2

Owing to this behavior of the integrands, we adopted the following procedure for the integrations. Writing a typical integral in the form

$$\int_0^{.30544} I(y)dy = \int_0^{.0015} I(y)dy - \int_{.30544}^{.306} I(y)dy + \int_{.0015}^{.078} I(y)dy + \int_{.078}^{.156} I(y)dy + \int_{.156}^{.306} I(y)dy$$

we, (a) assumed the integrand was linear for the calculation of the first two terms.

(b) applied Simpson's rule to the last three terms using the Simpson's intervals .0015, .003, .006, respectively.

It is possible that a similar technique can be applied to the calculation of the more complicated integrals involved in the proposed iterative scheme.

CRITIQUE OF THE LENNARD-JONES AND DEVONSHIRE THEORY:

W. Fickett and Wm. W. Wood⁷ at Los Alamos have recently completed calculations for Lennard-Jones Devonshire liquids at very high densities and temperatures. They found that the Lennard-Jones and Devonshire equation is quite satisfactory under these conditions. However, under less severe conditions, this simplified theory is not nearly so accurate.

From experimental vapor pressure data on Xe, Ar, Kr, N₂, and O₂ we have calculated the density of a Lennard-Jones and Devonshire liquid in equilibrium with the vapor phase.⁶ In performing these calculations we adopted the critical constants $z_{crit} = .301$ and $T_{crit}^* = 1.3$ as suggested by Wentorf, Buehler, Hirschfelder and Curtiss.⁴ The results of these calculations, together with experimental "co-existence data", are presented in Figure 3.

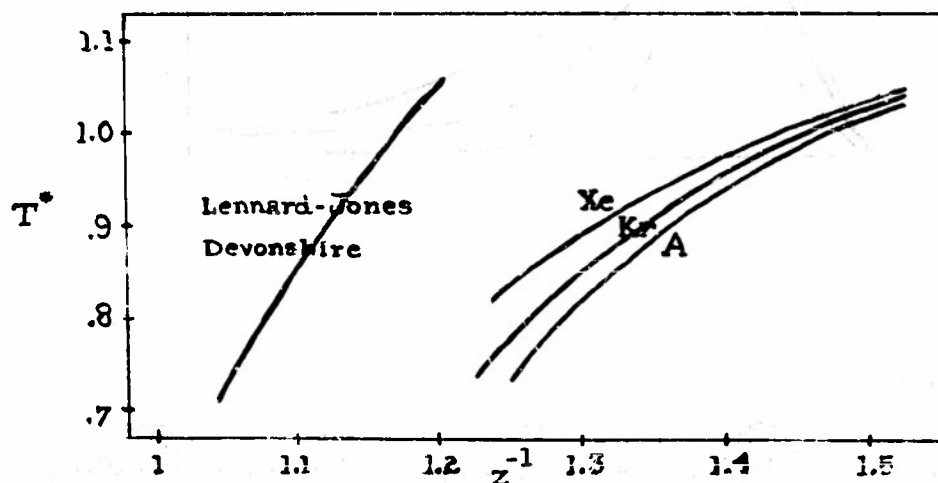


Figure 3.

From Figure 3 we see that a Lennard-Jones and Devonshire liquid is more dense than any of the real liquids considered. In addition, our calculations indicate that the Lennard-Jones and Devonshire value for pV/RT at the critical

7. Tables of the Lennard-Jones and Devonshire Equation of State at High Temperatures and Densities, W. Fickett and Wm. W. Wood, Los Alamos Scientific Laboratory, Los Alamos, New Mexico (unpublished)

point is .793 compared to the experimental value of about .292 for the rare gases. We also find considerable disagreement between experimental and Lennard-Jones and Devonshire values for the various thermodynamic properties.

CONCLUSION:

In view of the inadequacies of the Lennard-Jones and Devonshire theory mentioned above, more accurate calculations in the liquid-vapor co-existence region and near the critical point seem desirable. Subsequent calculations would depend upon these results.

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