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# Relative Molar Volume and Limiting Solubility of $^3$He in Superfluid $^4$He[†]

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The relative fractional molar volume $\alpha$ of $^3$He in a $^3$He-$^4$He solution has been determined by a capacitance-measurement technique with a solution which remained in a single phase down to the lowest temperature of measurement, 31.7 mK. The observed temperature dependence at saturated vapor pressure is in excellent agreement with theory. Accounting for $^3$He kinetic and interaction effects, we deduce that $\alpha_0$, the fractional molar volume of $^3$He at zero (concentration, temperature, and pressure) is $0.286 \pm 0.001$. Measurements on more concentrated solutions permitted redetermination of the limiting solubility at zero temperature, with the result $x_0 = 0.0660 \pm 0.0006$.

## INTRODUCTION

Adding $^3$He to superfluid $^4$He increases the molar volume of the liquid, since $^3$He atoms have a lighter mass, and therefore a greater zero-point motion in the solution than $^4$He. The molar volume $V_{34}$ of a dilute solution of the $^3$He in $^4$He can be written in the form

$$V_{34}(x, T, P) = V_4(T, P)[1 + x\alpha(x, T, P)] \quad , \qquad (1)$$

where $V_4$ is the molar volume of pure $^4$He at the same temperature $T$ and pressure $P$ as the solution; $x$ is the $^3$He mole fraction, or concentration. Recently, Edwards *et al.*,[1] reported the value $0.284 \pm 0.005$ for $\alpha_0 = \alpha(0,0,0)$, while, in a prior communication, Boghosian and Meyer[2] gave the value $0.308 \pm 0.010$ for the same quantity. In view of the important role of $\alpha_0$ in the theory of solutions as developed by Bardeen, Baym, and Pines,[3] we have redetermined $\alpha$ in an $x = 0.05535$ concentration solution, as a function of temperature[4] at saturated vapor pressure, employing the same techniques used by Edwards *et al.*[1] and by Boghosian and Meyer.[2] Combining these measurements with the theoretical $x$ dependence of $\alpha$, including $^3$He interaction effects, enables us to determine $\alpha_0$.

In addition, by enriching the solution used, we are able to redetermine the limiting solubility of $^3$He in $^4$He as a function of temperature.

## EXPERIMENTAL

The molar volume is related to the dielectric constant through the Clausius-Mossotti equation

$$(\epsilon - 1)/(\epsilon + 2) = \tfrac{1}{3} 4\pi\alpha_p / V \quad ,$$

where $\epsilon$ is the dielectric constant, $\alpha_p$ is the molar polarizability, and $V$ is the molar volume of the medium. It was shown by Peshkov[5] and by Boghosian and Meyer[6] that the molar polarizability of $^3$He and of $^4$He are equal. Thus, by measuring the capacitance of a system for which a $^3$He-$^4$He solution acts as the dielectric, it is possible to deduce the molar volume of the solution, and thereby the parameter $\alpha$.

In practice, there is likely to be stray capacitance in the system; therefore, it is necessary to measure the dielectric constant of a known material $^4$He in order to calibrate the system. One need not, however, calculate $V_{34}$ from the data – with the attendant loss in precision – in order to find $\alpha$. If one assumes that the vacuum value of the capacitor $C_v$ is given by a capacitance $C_0$ which is sensitive to the dielectric plus a stray capacitance $C_S$ which is independent of the medium between the plates, then

$$C_v = C_0 + C_S \, ,$$

$$C_4 = \epsilon_4 C_0 + C_S \, ,$$

$$C_{34} = \epsilon_{34} C_0 + C_S \quad ,$$

where the subscript 4 refers to pure $^4$He and the subscript 34 to the solution. Combining the above equations and the Clausius-Mossotti equation with Eq. (1) one finds by straightforward manipulation that

$$\alpha x = \frac{C_4 - C_{34}}{C_{34} - C_v} \times \frac{3(C_v - C_s)}{C_4 + 2C_v - 3C_s} \quad . \qquad (2)$$

Two cells were constructed. The first, with the thermometric salt inside, had a very long equilibrium time (hours) when the $^3$He was added to the $^4$He at low temperatures; in addition, the equilibrium value indicated that approximately 20% of the $^3$He was not in the capacitor. The second cell was one that had been used for sonic measurements[7] but with a parallel plate capacitor replacing the sonic components. This placed the capacitor at the bottom of the cell so that the $^3$He light phase filled the capacitor. The capacitor was assembled from two coaxial back-up plates which had been used in the sonic apparatus to reduce ullage. These plates were cut from a stock material, described below, and optically polished on both sides so the resultant wafer was 0.793 cm thick. The stock material was prepared by bonding a stainless-steel rod (0.635 cm o.d.) to a concentric stainless-steel cylinder (1.167 × 0.953 cm i.d.) with Emerson and Cummings "Stycast" 2850. The plates were separated by a stainless-steel washer (1.167×0.794 cm i.d.) of 25 $\mu$ nominal thickness. The central button of each plate had three 0.0508-cm holes and the backs were grooved so that the liquid could enter the capacitor. From the published values of the molar volume[8] and of the dielectric constant[9] for $^4$He, $C_S$ was found to have the value 0.30072 and $C_0$ the value 11.2794 at 1.170 K. This capacitor was sufficiently stable to measure the capacitance to $\pm 0.2\times10^{-4}$ pF with a day-to-day stability of $\pm 1\times10^{-4}$ pF (one unit on the General Radio 1615A capacitance bridge). Since $(C_4 - C_{34})$ was the order of $100\times10^{-4}$ pF, it can be seen that an error of $1\times10^{-4}$ pF introduces an error of 1% in $\alpha x$. If $C_S$ in Eq. (2) were neglected, then an error of 0.05% would be introduced in $\alpha x$.

The cell was suspended from a chrome-alum-salt pill which supplied the refrigeration below 300 mK. The pill was constructed by compressing 162 g of millimeter-sized particles mixed with 136 g of powdered, reagent-grade silver chloride. The resultant cylinder was 3.5 cm diam by 12.7 cm long. A 0.635-cm hole was drilled along the axis so the pill could be suspended with a nylon rod; in addition three longitudinal grooves, at 90°, (0.318×0.318 cm) were milled on the side to receive the transmission lines and the $^3$He "switch" pump line. The $^3$He switch was used to remove the heat of magnetization and reduce the starting temperatures for demagnetization. The pill was coated heavily with "Apiezon N" grease, and then strips of "coil foil"[10] were placed around it; these provided thermal contact to the $^3$He switch and to the cell, and were held in place by tightly wound surgical tape. The pill was routinely demagnetized from 0.9–1.0 K in 10–15 min but 2 h were required before the cell came to the final temperature of 31 mK. The filling line from the cell to room temperature was of copper-nickel alloy (0.0406×0.0305 cm; a 30 cm length was used between the cell and the 300 mK $^3$He shield, and 41 cm between the shield and the 1.2 K $^4$He bath.

The experiment was performed by first obtaining the vacuum value $C_v$ as a function of temperature. The exact origin of the slight temperature dependence is not known but is probably due to residual contractions. These changes in $C_v$ are unimportant for the calculation of $\alpha$ but are important if values of the molar volume are desired.

After the vacuum value had been established, an accurately known quantity of $^4$He was condensed in the cell and $C_4$ was measured several times as a function of temperature. Finally, the cell was cooled to 500 mK with the $^3$He switch, the $C_4$ value determined again to obtain the capacitor shift, and an accurately known volume of $^3$He was condensed *without* warming the cell. The total volume of liquid in the experiment was about 0.7 cm$^3$ whereas the cell volume was 1.41 cm$^3$; this was done so that no material would be in the filling line. It had the disadvantage that at temperatures where the vapor pressure became appreciable the composition became uncertain. The vacuum line in which the gases were measured is equipped with a nominal 1000 cm$^3$ volume, a nominal 600 cm$^3$ volume in three sections, and a constant volume manometer. The standard volumes have been accurately calibrated by weighing outgassed water contained between reference marks. The mercury level in the manometer was read with a Wild cathetometer and all pressures were reduced to standard gravity and 0 °C. The greatest error in the quantity of gas, and therefore the composition, arises from temperature gradients between the manometer and the gas bulbs; these were on occasion as great as 0.3° or about 0.1%.

The first solution ($x = 0.05535$), which remained in a single phase to the lowest temperature, was used to determine $\alpha(T^*)$ from 31 mK to 1.2 K. The results are tabulated in Table I, where the entries have been terminated at $T^* = 525$ mK because of uncertainties in the composition due to the vapor phase. After these runs, the solution was enriched to $x = 0.09575$ for two series of limiting solubility measurements, and then diluted to $x = 0.07808$ for an additional series of solubility measurements.

The limiting solubility as a function of temperature was obtained by substituting the values of $C_{34}$ from the latter measurements in Eq. (2) along with the appropriate $C_4, C_v$, and $\alpha(T^*)$, obtained from measurements on the first solution. The re-

TABLE I. Experimental values of the cell capacitance: in vacuum, $C_v$; filled with $^4$He, $C_4$; and with $^3$He-$^4$He solution, $C_{34}$; and the derived fractional excess molar volume of $^3$He, $\alpha(.055, T)$.

| Temp (mK) | $C_{34}$ (pF) | $C_4{}^a$ (pF) | $C_v{}^b$ (pF) | $\alpha$ |
|---|---|---|---|---|
| Series A | | | | |
| 31.7 | 12.18218 | 12.19234 | 11.54829 | $0.284_1$ |
| 72.5 | 12.18221 | 12.19238 | 11.54833 | $0.284_4$ |
| 92.9 | 12.18224 | 12.19239 | 11.54835 | $0.283_8$ |
| 101.3 | 12.18227 | 12.19240 | 11.54836 | $0.283_2$ |
| 143.3 | 12.18230 | 12.19243 | 11.54838 | $0.283_2$ |
| 205.5 | 12.18239 | 12.19247 | 11.54842 | $0.281_8$ |
| 294.6 | 12.18250 | 12.19252 | 11.54847 | $0.280_1$ |
| 437.7 | 12.18267 | 12.19256 | 11.54852 | $0.276_4$ |
| Series B$^c$ | | | | |
| 37.6 | 12.18224 | 12.19235 | 11.54830 | $0.284_6$ |
| 88.8 | 12.18230 | 12.19239 | 11.54834 | $0.284_1$ |
| 128.4 | 12.18236 | 12.19242 | 11.54837 | $0.283_2$ |
| 189.6 | 12.18245 | 12.19246 | 11.54841 | $0.281_8$ |
| 191.4 | 12.18243 | 12.19246 | 11.54841 | $0.282_4$ |
| 276.8 | 12.18256 | 12.19251 | 11.54847 | $0.280_1$ |
| 524.6 | 12.18285 | 12.19259 | 11.54854 | $0.274_1$ |

[a] From a smooth curve of $C_4$ versus $T^*$.
[b] From a smooth curve of $C_v$ versus $T^*$.
[c] Adjusted by $-7\times10^{-5}$ pF for condenser shift.

sults plotted against $(T^*)^2$ are displayed in Fig. 1. The straight line through the data was obtained by a least-squares fit, for which the equation is

$$x^l(T^*) = (0.0660\pm0.0006)[1+9.3(T^*)^2] \ .$$

The constants are slightly different from those quoted by Edwards *et al.*[1] but the agreement is quite satisfactory. The temperature dependence is consistent with theoretical calculations based on the $^3$He-$^3$He effective interaction. The limiting solubility at absolute zero, $x_0 = 0.0660$, is almost the average of the value, $0.064 \pm 0.0006$, given by Edwards *et al.*, and the value $0.0684 \pm 0.0006$

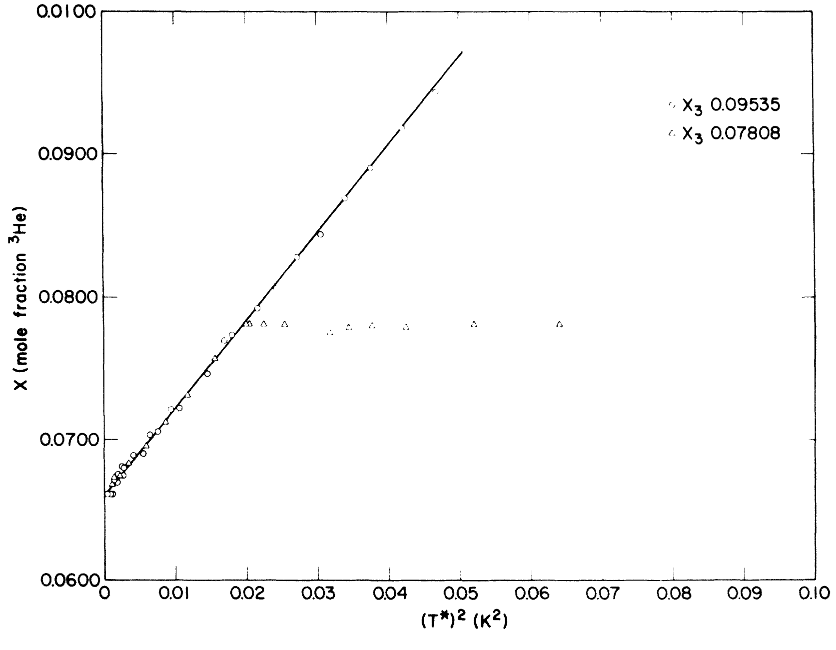

FIG. 1. The limiting solubility of $^3$He in $^4$He as a function of $(T^*)^2$, the square of the magnetic temperature.

given by Schermer *et al.*[11] It should be noted that in these calculations we used $\alpha = \alpha(0.055, T)$ instead of $\alpha(x, T)$; this could produce deviations at $x = 0.095$. The values of $\alpha(T^*)$ are in reasonably good agreement with those quoted by Edwards *et al.*, and $\alpha(0.055, 0)$ is in exact agreement with Edwards *et al.*[1]

## THEORETICAL

The molar volume of a dilute solution can be calculated from the Gibb's free energy per particle $\mu$, by

$$V_{34} = (\partial\mu/\partial P)_{x,T} \,, \tag{3}$$

where $\mu$ is given in terms of the $^3$He and $^4$He chemical potentials by

$$\mu(x, T, P) = x\mu_3(x, T, P) + (1 - x)\mu_4(x, T, P) \,. \tag{4}$$

The Gibb's-Duhem relation allows us to write

$$\left(\frac{\partial\mu_4}{\partial x}\right)_{T,P} = -\frac{x}{1-x}\left(\frac{\partial\mu_3}{\partial x}\right)_{T,P} \,,$$

so that by integration we have

$$\mu(x, T, P) = (1 - x)\mu_4(0, T, P) + x\mu_3(x, T, P) - (1 - x)\int_0^x dx' \frac{x'}{1-x'}\frac{\partial\mu_3(x', T, P)}{\partial x'} \,. \tag{5}$$

Now the $^3$He quasiparticle energy can be written in the form

$$\epsilon_p = \epsilon_0(T, P) + p^2/2m + \textstyle\sum_{p'} f_{pp'} n_{p'} \,, \tag{6}$$

where $\epsilon_0$ (a negative number) is the binding energy of a single $^3$He atom to pure $^4$He; $m (\approx 2.34\, m_3$ at $T = 0$ and $P = 0)$ is the effective mass[3] of a single $^3$He atom in pure $^4$He;

$$n_p = (e^{(\epsilon_p - \mu_3)/KT} + 1)^{-1}$$

is the $^3$He quasiparticle distribution function; and $f_{\vec{p}\vec{p}'}$ is the Landau parameter describing the $^3$He-$^3$He effective interaction.[3] Thus, for a dilute solution $\mu_3$ is of the form

$$\mu_3(x, T, P) = \epsilon_0(T, P) + \mu_F(x, T, P) + x\epsilon_1(x, T, P) \,, \tag{7}$$

where $\mu_F$ is the chemical potential of a free Fermi gas of mass $m$ at temperature $T$ and density $n_3$ equal to that of the $^3$He in solution, and $\epsilon_1$ is a correction roughly proportional to the strength of the $^3$He-$^3$He effective interaction.

We now expand $\mu$ for small $x$. It is sufficient at present to replace $\epsilon_1(x, T, P)$ by $\epsilon_1(O, T, P)$. On the other hand, $\mu_F$ is of the form $KT_F g(T/T_F)$, where $g$ is a known dimensionless function obeying $g(0) = 1$;

$$T_F = p_F^2/2mK$$

is the $^3$He Fermi temperature; and $p_F = \hbar(3\pi^2 n_3)^{1/3}$. Thus, $\mu_F$ cannot be expanded directly in integer powers of $x$. Instead, we use the fact that for a free Fermi gas at fixed $T$

$$d\mu_F = \frac{3}{2}\left(\frac{\partial P_F}{\partial n_3}\right)_m \frac{dT_F}{T_F} \,, \tag{7'}$$

where $P_F$ is the pressure of a free Fermi gas of density $n_3$, mass $m$, and temperature $T$. Equation (7′) includes any variation of $m$. Then by a short calculation we find that up to terms of the order of $x^2KT/m_4s_0^2$, for $T \gg T_F$, and $x^2KT_F/m_4s_0^2$, for $T \ll T_F$

$$\mu(x, T, P) = \mu_4(0, T, P)(1 - x) + x[\epsilon_0(T, P) + \mu_F(x) - P_F(x)/n_3] + \tfrac{1}{2}x^2\epsilon_1(0, T, P) \,. \tag{8}$$

$m_4$ is the $^4$He atomic mass, and $s_0 = 238$ m/sec is the first sound velocity of pure $^4$He at $T = 0$ $[m_4s_0^2/K = 27.2\text{ K}]$. The remaining terms $\sim x^{5/3}$ not included in (8) contribute $\lesssim 0.001$ to $\alpha$ in a 5.5% solution.

The molar volume can be calculated from (8) and (3). Using (7), we find

$$V_{34}(x, T, P) = V_4 + x\left(V_3 - V_4 + \frac{3}{2}\frac{P_F}{n_3}\frac{1}{T_F}\frac{\partial T_F}{\partial P}\right) + \frac{x^2}{2}\frac{\partial\epsilon_1(0, T, P)}{\partial P} \,, \tag{9}$$

where $V_4(T, P)$ is the molar volume of pure $^4$He, and $V_3(T, P) \equiv \partial\epsilon_0(T, P)/\partial P$. Comparison of Eq. (9) with Eq. (1) shows that to the present order in $x$,

$$\alpha(x, T, P) = \alpha(0, T, P) + \frac{P_F}{n_3}\frac{\partial n_4}{\partial P}\left(1 - \frac{3}{2}\frac{n_4}{m}\frac{\partial m}{\partial n_4}\right) + \frac{x}{2}n_4\frac{\partial\epsilon_1(0, T, P)}{\partial P} \,, \tag{10}$$

where $n_4 = L/V_4$. Now, $\alpha(0, T, P) = (V_3/V_4) - 1$ can, to a high order of accuracy, be replaced by its $T = 0$ value, and $\partial n_4/\partial P$ replaced by $1/m_4s_0^2$. It is convenient to define $Y(T)$ by

$$Y(T) = P_F(T, T_F)/P_F(0, T_F) = P_F(T, T_F)$$

$$\times (5m/n_3 p_F{}^2);$$

then $\alpha(x, T, P) = \alpha_0(P) + Y(T)$

$$\times \frac{P_F{}^2}{5mm_4 s_0{}^2}\left(1 - \frac{3}{2}\,\frac{n_4}{m}\,\frac{\partial m}{\partial n_4}\right) + \frac{x}{2}\,n_4\,\frac{\partial\epsilon_1(0, T, P)}{\partial P}\ . \tag{11}$$

Aside from the $\epsilon_1$ term, this expression agrees with that given in Refs. 1 and 2 to lowest order in $x$ (e.g., $x^{2/3}$ at $T = 0$). We emphasize that Eq. (11) includes the $x$ dependence of the phonon frequencies. The principal temperature dependence is in $Y(T)$.

Figure 2 shows $\alpha(0.055, T, 0)$ as listed in Table I. The solid line represents Eq. (11), neglecting the minor contribution from temperature dependence of $\epsilon_1$, and drawn through the point at 128.4 mK. The value $(n_4/m)(\partial m/\partial n_4) = 1.12$, which gives a good theoretical fit[12] to the temperature-dependence measurements[13] of the first sound velocity in dilute solutions of $^3$He in $^4$He, was used here; it happens also to give the best fit to the presently observed temperature dependence of $\alpha$. This value is 10% less than that given in Ref. 1. The excellent agreement between theory and experiment, which extends to $T > T_F$, provides a further confirmation of the description of the $^3$He as a gas of quasiparticles obeying Fermi-Dirac statistics.

Extrapolating back to $T = 0$ we conclude that

$$\alpha_0 + \tfrac{1}{2}\,x\,n_4\,\frac{\partial\epsilon_1}{\partial P} = 0.2882\ . \tag{12}$$

To deduce $\alpha_0$ we must know the pressure dependence of the Fermi-liquid correction $\epsilon_1$. The main Fermi-liquid correction to $\mu_3$ can be approximated by $\epsilon_1 = \frac{1}{2}\,n_3 v_0$, where $v_0 \approx -\alpha_0{}^2 m_4 s_0{}^2/n_4$ is the $^3$He-$^3$He effective interaction at zero momentum transfer. Then

$$\tfrac{1}{2}\,x n_4\,\frac{\partial\epsilon_1}{\partial P} \approx -\tfrac{1}{2}\,x\alpha_0{}^2\left(\frac{n_4}{s_0}\,\frac{\partial s_0}{\partial n_4} + \frac{n_4}{\alpha_0}\,\frac{\partial\alpha_0}{\partial n_4}\right)\ , \tag{13}$$

which is ~ – 0.0026 for a 5.5% solution. Exchange contributions to $\epsilon_1$ could modify this result by ~30%, but since little is known about the pressure dependence of the $^3$He-$^3$He effective interaction at finite momentum transfer, we cannot estimate them accurately. We can conclude then that

$$\alpha_0 = 0.286 \pm 0.001\ ,$$

in excellent agreement with Ref. 1.

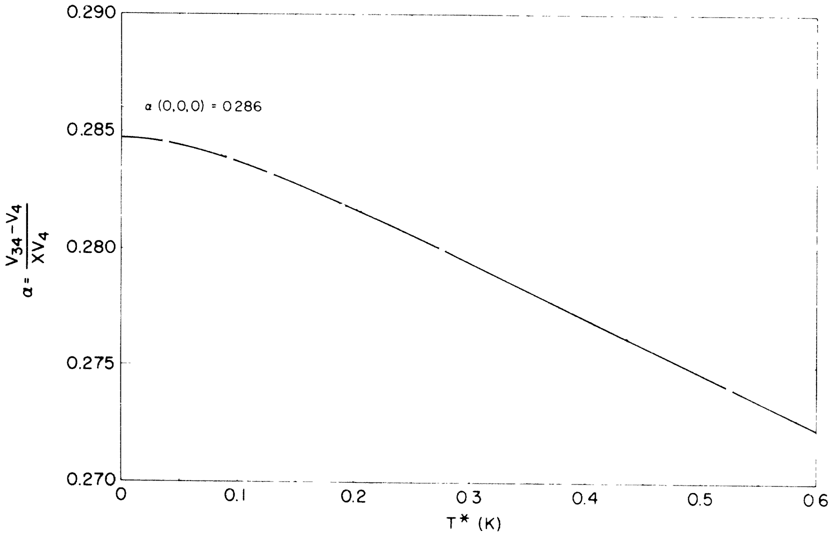

FIG. 2. The fractional relative molar volume $\alpha$ of $^3$He in $^4$He as a function of $T^*$, the magnetic temperature, for a solution containing 5.535 mole% $^3$He at saturated vapor pressure. The solid line is the theoretical fit normalized to the point at 128.4 mK.

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# Quantum Corrections to the Coexistence Curve of Neon near the Triple Point

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Quantum corrections to the free energy of a classical Lennard-Jones system are calculated, using the well-known Wigner expansion in powers of $\hbar$. In the vicinity of the triple point of neon, the $\hbar^4$ term turns out to be about 40 times smaller than the $\hbar^2$ term, which justifies truncation of the series after the first correction term. The resulting changes in the densities and pressures of melting and condensation transitions give rise to a phase diagram which is in good agreement with experimental neon results.

## I. INTRODUCTION

In a recent paper,[1] Hansen and Verlet have studied the gas-liquid and fluid-solid phase changes of a classical system of particles interacting through the Lennard-Jones interatomic potential

$$v(r)=4\epsilon\left[(\sigma/r)^{12}-(\sigma/r)^{6}\right]. \qquad (1)$$

The basic idea of that work was to calculate the free energies of the condensed and solid phases by integrating the equation of state obtained by the Monte Carlo method along an isothermal reversible path devised for that purpose. A Maxwell double-tangent construction allowed them to obtain the transition densities and pressures which turned out to be in good agreement with experimental argon data.

It is well known that the coexistence curve of neon and, in particular, its triple point differ sensibly from those expected by the law of corressponding states.[2] Since the neon atomic mass is about half the argon atomic mass, and as the triple-point temperature is 24.56 °K compared to 83.81 °K for argon,[3] it is natural to expect that the deviations from the law of corresponding states are due to quantum effects. This is confirmed by the fact that under triple-point conditions the thermal wavelength in solid neon is only about 4 times smaller than the average interparticle distance; in reduced units (i.e., $\sigma=1$, $\epsilon/k=1$), we found $\lambda=(2\pi\hbar^2/mkT)^{1/2}$ to be equal to 0.265, whereas the nearest-neighbor distance is approximately $d=1.1$.

In our calculations, we used the values of the potential parameters determined by Brown in the solid phase under ground-state conditions,[4] i.e.,