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Extension of the λ curve of ^{4}He into the region of the metastable state of liquid helium

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Abstract. **A possible extension (λ') of the λ curve of liquid helium beyond the T, p boiling line, i.e., into the region of a metastable liquid state, is examined. The relation between the slope of the λ curve $\mathrm{d}p/\mathrm{d}T_\lambda$ and the sign of the thermal expansion coefficient of liquid helium α_p is discussed. Experimental data for α_p are analyzed using thermodynamic equations to reveal that the derivative $\mathrm{d}p/\mathrm{d}T_\lambda$ changes sign in the metastable region. As a result, the shape of the λ' extension becomes similar to that of the pressure-shifted Bose-condensation line of an ideal ^{4}He gas. The position of the λ' curve is not entirely arbitrary due to the spinodal nature of stretched liquid helium.**

In the T, p phase diagram of ^{4}He shown in Fig. 1, the λ line of the He I – He II transition is terminated at the melting curve above and at the liquid – vapor (boiling) curve below. Neither of these, however, represents a limit for the λ transition physically, and it is of interest therefore to see whether it is possible to extend the transition region to the metastable states of liquid helium lying above the melting curve and below the boiling curve shown in Fig. 1.

The superheating and supercooling of a liquid have been studied for different classes of materials [1 – 3]. A well defined state of metastability implies that a system has relaxed to its quasi-equilibrium state in all respects except for the formation of nuclei of a new phase more stable under the given external conditions. The average lifetime $\langle\tau\rangle$ of the system in a metastable state depends in a definite way on the temperature, pressure, and the sample volume V. Many laboratory experiments provide support for Volmer – Zel'dovich's homogeneous nucleation theory [4, 5], which allows the nucleation rate $J = J(T, p)$ ($\mathrm{s}^{-1}\,\mathrm{cm}^{-3}$) or the average lifetime of nuclei $\langle\tau\rangle$ to be calculated. In a simple case, these two quantities are related by $\langle\tau\rangle = (JV)^{-1}$. In thermodynamic terms, a metastable system may not differ from a stable one if the characteristic time of the experiment $t_{\exp}$ satisfies the inequalities

$$\{t_i\} < t_{\exp} < \langle\tau\rangle\,, \qquad (1)$$

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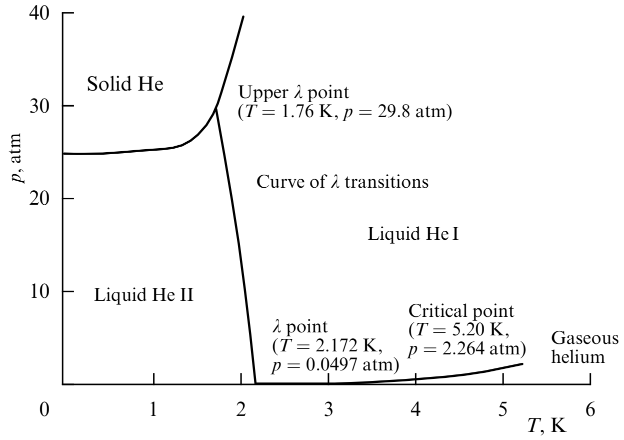

Figure 1. Phase diagram of ^{4}He.

where t_i is the relaxation time for the system's property i (temperature, pressure, etc.). The relations (1) express the condition for a well defined (pure) state of metastability, a state nicely exemplified by diamond.

In the present context, it may be said that the curve of the λ transition in ^{4}He does continue beyond the boiling curve into the region of the metastable state. But then the question arises of exactly how it continues as we go deeper into this region.

In the ordinary phase diagram shown in Fig. 1, the region of the He I – He II transition turns out to be open below, thus raising doubts about whether the theory of this phase transition is complete enough.

Note here that since there is experimental evidence for both liquid helium in the superheated (stretched) state [6 – 9] and the λ transition in this state [10, 11], we have not only theoretical arguments but also experimental grounds for addressing the formulated problem. [1] In Refs [10] and [11], an experimental liquid helium cell, with a volume of 0.72 cm^3 and 0.67 cm^3, respectively, was fitted with a heater and a resistance thermometer and connected by a capillary glass tube to a large bath with helium II, with the pressure in the latter maintained at a fixed level. The apparatus used in Ref. [11] allowed the visual monitoring of the sample. When the heater was on, a thermogram was recorded. At the λ-transition point, the strong reduction in the helium flow through the tube caused a dramatic increase in the rate of the temperature rise at fixed heater power. The He II → He I phase transition was reliably identified by a kink in the thermogram. Both phases were metastable if the pressure

[1] The author has no data on supercooled liquid ^{4}He in the crystallization region, so these states will not be considered here.

was maintained below the point of intersection of the λ curve with the saturation curve. The sequence of phase states in this case was from He II to superheated He II to superheated He I and then to boiling. The development and growth of vapor bubbles in the sample caused a sharp reduction in temperature. Figure 2 shows the data of Ref. [11] obtained in the pressure range 650 to 3.8 mmHg. In this range, it is seen that the λ curve continues into the region of metastable states of the liquid without having any singularities at the point where it intersects the liquid – vapor equilibrium curve.[2] Measurements in Ref. [10], while covering a narrower pressure range, agree well with the data of Ref. [11]. The methodology used in these studies prevented moving into the range $p < 0$. In order to extend the λ curve deeper, other factors, namely, those related to the slope $\mathrm{d}p/\mathrm{d}T_\lambda$ of this extension and the stability of the liquid, are to be considered.

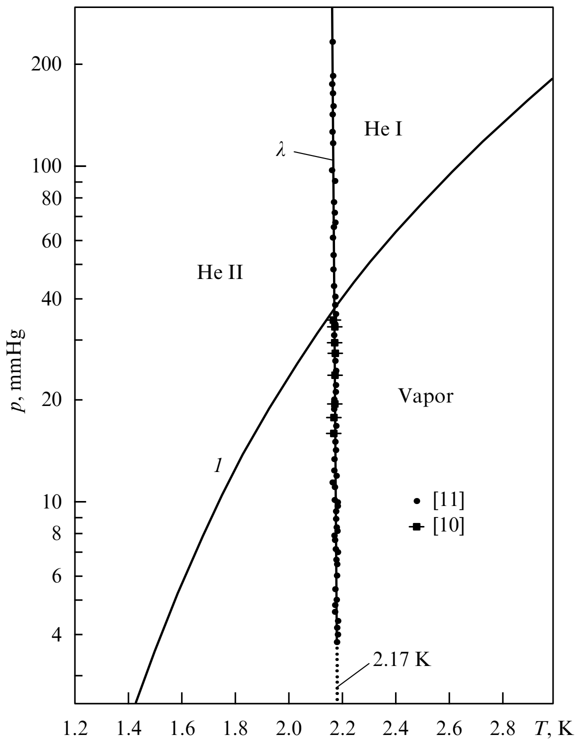

Figure 2. The He I – He II transition curve continued into the region of the metastable states of liquid helium. Points indicate the experimental results of Refs. [10] and [11]; solid line *1* represents the saturation curve.

It follows from general arguments that at negative pressures the continued λ curve must lie above the spinodal, the boundary of the region where the liquid is essentially unstable against small (homophase) perturbations. The spinodal is defined by the relation

$$\left(\frac{\partial p}{\partial v}\right)_T = 0\,, \tag{2}$$

it begins at the critical point C for the liquid – vapor equilibrium, and terminates at $T = 0$ (see Fig. 3, in which the spinodal *3* is constructed based on the data of Ref. [12]).

[2] There is a strong tradition in helium studies to measure pressure in mmHg and in atm (1 atm = 1.01 bar, 1 bar = 1×10^5 N m^{-2}). We willl use bars below.

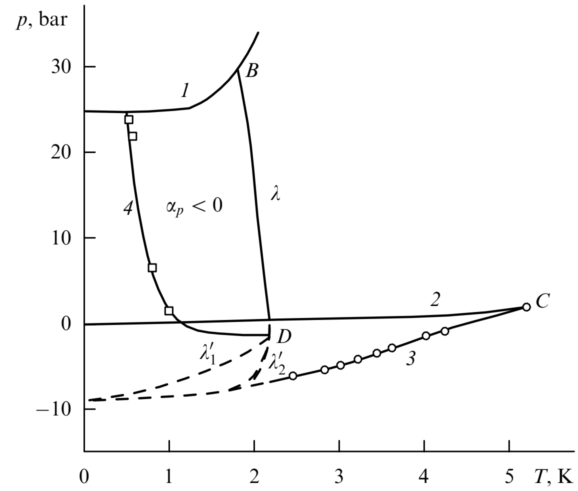

Figure 3. Extended phase diagram of ^{4}He: *1*, melting curve; *2*, liquid – vapor equilibrium curve; *C*, critical point; *3*, spinodal of the liquid; λ'_1, λ'_2, two possible continuations of the λ curve into the negative-pressure region; *4*, the lower boundary of the region of negative thermal expansion coefficient ($\alpha_p < 0$).

In Fig. 3, which shows an extended phase diagram of ^{4}He, the assumed continuation of the λ curve beyond the liquid – vapor equilibrium curve *2* is shown dashed. We will call this portion the λ' curve. The striking thing about this curve is its turn at point D. The λ' curve closes the He I – He II transition region from below and obeys the Nernst theorem as $T \to 0$. Note also that the condition $\mathrm{d}p/\mathrm{d}T_{\lambda'} > 0$ is satisfied on the Bose condensation curve of the ideal ^{4}He gas [13, 14]. These, of course, are not sufficient arguments to accept the λ curve continuation of Fig. 3, with the sign reversal in the derivative $\mathrm{d}p/\mathrm{d}T_\lambda$. We therefore turn to a thermodynamic analysis of the behavior of this derivative based on the properties of liquid helium.

It is known [15] that the density $\rho = 1/v$ of ^{4}He near the λ curve has a maximum on isobars at temperatures $T_m(p)$. Since the difference $T_m - T_\lambda$ is positive and equals 6 mK [15] at the intersection point of the λ curve and the saturation curve *2*, and since it remains small at higher pressures, it follows that the λ curve passes through liquid helium states for which

$$\left(\frac{\partial \rho}{\partial T}\right)_p > 0\,, \quad \text{or} \quad \left(\frac{\partial v}{\partial T}\right)_p < 0\,. \tag{3}$$

The thermal expansion anomaly expressed by inequalities (3) persists up to curve *4* (see Fig. 3), at which the thermal expansion coefficient $\alpha_p = (1/v)(\partial v/\partial T)_p$ goes to zero, and the density on the isobars has a minimum [16]. To the left of and below curve *4*, normal thermal expansion behavior, $\alpha_p > 0$, is observed. (Because of the scale used in Fig. 3, the curve of density maxima cannot be separated from the λ curve).

The derivative $\mathrm{d}p/\mathrm{d}T_\lambda$ along the λ curve can be written as

$$\frac{\mathrm{d}p}{\mathrm{d}T_\lambda} = \left(\frac{\partial p}{\partial T}\right)_V + \left(\frac{\partial p}{\partial v}\right)_T \frac{\mathrm{d}v}{\mathrm{d}T_L}\,. \tag{4}$$

Note the signs of the terms on the right-hand side of this equation. For stable homogeneous states (including metastable states up to the spinodal), we have

$$\left(\frac{\partial p}{\partial v}\right)_T < 0\,. \tag{5}$$

From the thermodynamic relation

$$\left(\frac{\partial p}{\partial T}\right)_V \left(\frac{\partial T}{\partial v}\right)_p \left(\frac{\partial v}{\partial p}\right)_T = -1 \qquad (6)$$

and condition (5) we see that the derivatives $(\partial p/\partial T)_V$ and $(\partial v/\partial T)_p$ have the same sign. Close to the λ curve these derivatives are both negative for the reasons mentioned above. Since, further, the density of ^{4}He on the λ curve is known to decrease with increasing temperature, i.e., $\mathrm{d}v/\mathrm{d}T_\lambda > 0$, it follows that both terms on the right-hand side of Eqn (4) are negative, thus providing a thermodynamic explanation of the experimental fact $\mathrm{d}p/\mathrm{d}T_\lambda < 0$.

We will now focus our attention on the vicinity of point D in the phase diagram. Is it plausible that the λ curve changes its slope here as the derivative $\mathrm{d}p/\mathrm{d}T_\lambda$ changes sign?

As already noted, for liquid helium there exists a region where the variation of the isobaric density is not monotonic, showing first a minimum and then a maximum as the temperature is increased. The curve of density maxima adjoins the λ curve and is not separated from it in Fig. 3. In Ref. [16], where experimental minimal density points are compared with those of other workers, the $\alpha_p = 0$ curve in T, p variables encounters the melting curve *1* at $T = 0.59$ K and the boiling curve *2* at $T = 1.15$ K. In Ref. [16], the thermal expansion coefficient was calculated using the measured values of heat capacity and the values of the $\Delta T/\Delta p$ ratio for the quasi-adiabatic compression and expansion of the sample. In Refs. [17, 18], the density and α_p of liquid ^{4}He were determined from dielectric constant measurements. The authors of Ref. [17] point to the good agreement between their results and the T, p data of Ref. [16] for the $\alpha_p = 0$ curve.

As the pressure decreases ($p < 5$ bar), the $\alpha_p = 0$ and λ curves rapidly approach each other. Allowing for the continuation of both curves into the $p < 0$ region, their meeting point D can be determined by extrapolation. This point terminates the region of the anomalous behavior ($\alpha_p < 0$) of the thermal expansion coefficient, and instead of two density extrema a point of inflection appears on the isobars for $p > p_D$. Outside of the closed region with $\alpha_p < 0$ shown in Fig. 3, the derivatives $(\partial v/\partial T)_p$ and $(\partial p/\partial T)_V$ are positive and the λ curve continues with a different slope, $\mathrm{d}p/\mathrm{d}T_\lambda > 0$. The positive sign of the derivative λ' follows from Eqns (4)–(6) and the equality

$$\frac{\mathrm{d}v}{\mathrm{d}T_\lambda} = \left(\frac{\partial v}{\partial T}\right)_p + \left(\frac{\partial v}{\partial p}\right)_T \frac{\mathrm{d}p}{\mathrm{d}T_\lambda}\,.$$

Thus, the transition from the λ to λ' curve with the reversal of the sign of the slope is found to be related to the disappearance of the thermal expansion anomaly in He II below the point D.[3]

The proposed approach was first discussed in the present author's earlier publication [19]. Note that the specific volume of liquid helium on the λ' curve decreases with increasing temperature, i.e., $(\mathrm{d}v/\mathrm{d}T)_{\lambda'} < 0$. This can be shown by considering a series of isochores in the T, p plane and the way they successively 'attach themselves' to the λ' curve. On the λ curve, we have $(\mathrm{d}v/\mathrm{d}T)_\lambda > 0$, but the specific volume increases monotonically as one moves from point B to point D (see Fig. 3) and further along the λ' curve.

[3] Note that, apart from helium, water has a region where $\alpha_p < 0$.

We turn next to the theory of the Bose condensation of an ideal gas of ^{4}He. Although the link between the phase transition in liquid helium and the Bose condensation phenomenon has frequently been noted in the literature, the observed behavior of ^{4}He differs considerably in detail from the case of an ideal gas [20]. The pressure on the Bose-condensation curve can be written as [13, 14]

$$p = 0.0852\,\frac{gm^{3/2}(k_\mathrm{B}T)^{5/2}}{\hbar^3}\,. \qquad (7)$$

For ^{4}He, $g = 1$, and $m = 6.648 \times 10^{-24}$ g.

The table below lists the pressure values calculated with Eqn (7) for several values of temperature. Comparing the λ' curve with the Bose-condensation curve shows that the starting point of the λ curve at $T = 0$ is shifted by about 9 bar into the region of the stretched state of liquid helium ($p < 0$). This may be due to the neglect of molecular interactions in deriving the Bose-condensation theory result (7). With a proper shift, the two curves do not move apart strongly before the point of reversal of the λ' curve. If we bring them together at $T = 0$, $p = 0$, then at $T = 2.17$ K the pressure on the Bose-condensation curve is 19.3 bar and on the shifted λ' curve, about half that value. At the same time, the λ curve has a qualitatively different shape.

Table. Values of pressure as calculated with Eqn (7), on the Bose-condensation curve for the ideal gas of ^{4}He, $p = 2.788T^{5/2}$ bar.

T, K	0	0.5	1.0	1.5	1.8	2.0	2.17
p, bar	0	0.493	2.79	4.13	12.1	15.8	19.3

Actually, the only conclusions drawn from the thermodynamic analysis are the change in the slope of the λ curve for $p < 0$ and the qualitative correspondence between the λ' curve and the curve of the Bose condensation of ideal gas. To reach these conclusions, the curves of density extrema had to be extrapolated beyond the zero isobar. In extrapolating curve *4*, the approximation relation (7) of Ref. [16] with extrapolated values of $a(T)$ and $b(T)$ was employed, yielding $T_D = 2.18$ K and $p_D = -0.98$ bar as the coordinates of the point D. Alternatively, curve *4* was also extrapolated by rectifying the curve $\alpha_p(T, p) = 0$ using the coordinates p, $y = (T - T_0)^{-n}$: $p = Ay + B$. For $T_0 = 0.4$ K and $n = 1.5$, the values $p_D = -2.76$ bar and $T_D = 2.20$ K, not too far from the previous estimate, are found.

That the λ curve has a temperature maximum ($T = T_D$) follows from the theoretical estimates [21] of its behavior in density–temperature variables (with the density being given by $\rho = 1/v$). While pressure is less suitable than density as a physical parameter, it provides a clearer distinction between the stable and metastable states of liquid helium. The variables T, p, ρ are related by the equation of state, which, as is indicated by the change in the sign of the thermal expansion coefficient, is rather complicated in the region of interest here.

Some remarks about the shape of the λ' curve are appropriate here. There is an unresolved arbitrariness in the way the curve is located in Fig. 3. It is natural to assume that the possible position of the λ' curve on the pressure axis is limited from below by the spinodal of liquid ^{4}He, i.e., by the boundary of the region where liquid helium is stable against small homophase perturbations. We assume that as $T \to 0$, the limiting values for the λ' and the spinodal are the same. [4] The determination of the spinodal of ^{4}He, while of interest in itself, is also directly relevant to our problem. The response of a homogeneous system when beyond the spinodal is to increase small deviations from equilibrium, with thermal motion enhancing the instability. The spinodal as the stability boundary can be determined by the behavior of the elasticity $(\partial p/\partial \rho)_T$ or of the reciprocal heat capacity $c_p^{-1} = (\partial T/\partial s)_p$, both of which tend to zero as the spinodal is approached. Ref. [12] uses the experimental data of Ref. [23] on the sound speed w_s. The isothermal sound speed w_T is expressed directly in terms of the derivative $(\partial p/\partial \rho)_T$, $w_T = (\partial p/\partial \rho)_T^{1/2}$, and the adiabatic sound speed w_s measured for $p > 0$ is recalculated into the isothermal speed using data for other thermodynamic properties,

$$w_T^2 = w_s^2 - \frac{T}{\rho c_V}\left(\frac{\partial p}{\partial T}\right)_V^2 ,$$

with c_V the heat capacity per unit volume at constant volume. The spinodal is approximated by assuming a linear pressure dependence for w_T^3 along the isotherms, $w_T^3 = ap + b$, where the parameters a and b are temperature-dependent. Then for the spinodal pressure we have $p_{sp} = -b/a$ for each temperature. For $T = 0$ K this approximation yields $p_{sp}(0) = -9$ bar. As already mentioned, the spinodal in Fig. 3 is consistent with the results of Ref. [12].

Experiments on the spontaneous boiling-up and cavitation strength of a liquid provide another way to estimate the pressure position of the spinodal from above. Although the curve of achievable superheated (stretched) states for a given nucleation rate lies above the spinodal (in pressure), there is a definite correlation between them. For ordinary liquids it has been established experimentally [1] that the predicted [4, 5] and observed values of the nucleation rate J as measured in $s^{-1}\,cm^{-3}$ agree well over a wide range of values of J, from $\log J \simeq 2$ to $\log J \simeq 22$. Experiments on ^{4}He have revealed that theory somewhat overestimates the degree of metastability of a liquid for a given nucleation rate [6 – 8]. A possible explanation is that the radiation background and cosmic radiation have an initiating effect given a time of experiment of the order of tens of seconds and a sample volume of the order of 1 cm^3. The degree of superheating may be smaller or the cavitation threshold lower if there are 'weak' places at the chamber walls or on solid inclusions. For superfluid helium, it is also suggested [24] that quantum vortices may act to lower the nucleation barrier.

To provide a better match between the curve of observed elongations in T, p variables and the calculated curve $J(T, P) = \text{const}$, the method of a focused acoustic field is employed [9]. This reduces the cavitation zone to $(\lambda/2)^3$, λ being the acoustic wavelength at a characteristic frequency of 1 MHz and a duration of the exciting radio pulse of 1 ms. It is worth noting here that the position of the spinodal in Fig. 3 is qualitatively consistent with that of the cavitation boundary of ^{4}He for $J \sim 10^{15}$ to 10^{20} s^{-1} cm^{-3}. The spinodal corresponds to greater elongations than the $J(T, p) = \text{const}$ curve drawn for the largest allowable J values in the homogeneous nucleation model. Note that the λ_1' curve in Fig. 3 lies in the region of achievable metastability of ^{4}He.

[4] Here the analogy with the crystal – liquid phase transition in simple substances may be employed. Upon the metastable continuation of the melting curves into the region of $p < 0$ and low temperatures, the coexisting liquid and crystal are seen to approach their spinodal state [22].

The curve (λ_1') in Fig. 3 illustrates the case in which the λ' curve and the spinodal approach each other gradually. We cannot rule out the possibility that after the turn at point D the λ' curve will terminate at the spinodal at a finite temperature T_1, $0 < T_1 < T_D$. This situation deserves special consideration. From Eqn (4) it follows that at the point where the λ' curve meets the spinodal (and where $(\partial p/\partial v)_T = 0$) we have

$$\frac{dp}{dT_\lambda} = \left(\frac{\partial p}{\partial T}\right)_v , \tag{8}$$

which means that the λ' curve here has a common tangent with the isochore passing through the point where the λ' curve meets the spinodal. But this is exactly the property the spinodal itself possesses: it envelopes the family of isochores in the T, p plane [1]. [5] To see this, we write the slope of the spinodal dp/dT_{sp} in a form analogous to Eqn (4),

$$\frac{dp}{dT_{sp}} = \left(\frac{\partial p}{\partial T}\right)_v + \left(\frac{\partial p}{\partial v}\right)_T \frac{dv}{dT_{sp}} . \tag{9}$$

Since the derivative dv/dT_{sp} is finite everywhere except for the liquid – vapor critical point, we are left with only one term on the right, i.e.,

$$\frac{dp}{dT_{sp}} = \left(\frac{\partial p}{\partial T}\right)_V . \tag{10}$$

Owing to conditions (8) and (10), the λ' curve and the spinodal have a common tangent at their meeting point $T = T_1$, which coincides with that to the isochore passing through this point. Consequently, the λ curve smoothly joins the spinodal rather than being stopped by it.

The spinodal (2) forms the boundary of the region of mechanical stability of the homogeneous state and corresponds to a strong thermodynamic singularity, whose effect is to destroy the correlations which allow the existence of He II. Beyond the spinodal, the mechanism of the spinodal decomposition of the liquid becomes operative [25], implying that the λ' curve cannot be ascribed any meaning there. For $T < T_1$, the spinodal plays the role of the boundary of the He II phase. Given all this, improving the accuracy of the position of the spinodal becomes important. [6]

To summarize, we could say that, thermodynamically, the negative slope of the λ curve in T, p variables is due to the unusual behavior of the liquid helium density — to the existence of a minimum and a maximum on the density isobars, between which the thermal expansion coefficient is negative, $(\partial v/\partial T)_p < 0$. The curve of density maxima follows the λ curve with a small shift upward in temperature. Upon extrapolating to $p < 0$, the extremum curves merge at point D (see Fig. 3), the extrema degenerating to an inflection point on subsequent isobars. Beyond point D there are no states with $\alpha_p < 0$, and further along the λ curve its slope changes sign, so that $dp/dT_{\lambda'} > 0$.

[5] Note that in the three-dimensional T, p, v space neither the λ curve nor the spinodal are plane.

[6] The referee drew my attention to a recent paper [26] which uses the path integral Monte Carlo method to study the behavior of helium at negative pressures. The spinodal points in the range 0.50 – 4.00 K are compared with the estimates of other researchers. The deviation from curve *3* in Fig. 3 of the present work is insignificant.

In this approach, the λ' curve in the metastable — but pre-spinodal — state of liquid helium is treated as a real, physical, lower-pressure boundary for the He I – He II phase transition. Negative pressure may be achieved in a liquid by, for example, applying a focused acoustic field [9]. While the exact position of the λ' curve has remained undetermined, two possibilities, λ'_1 and λ'_2, are shown in Fig. 3. A comparison of the λ' curve and the Bose condensation curve of an ideal gas reveals that they have the same sign for the derivative $\mathrm{d}p/\mathrm{d}T$ and show a pressure change of the same order over the temperature range from 0 to 2.17 K. This strongly suggests the use of the λ' curve for comparison with the theory of the Bose condensation of an ideal gas and for elucidating the relevance of this theory to the He I – He II transition.

Finally, the equation of state of superfluid helium depends on the energy spectrum of its (phonon – roton) elementary excitations [5, 17, 27], a spectrum whose form near the phase transition curve is not known with sufficient accuracy to yield the behavior of the density of liquid helium in fine detail.

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