# Connection between the Isobaric Thermal Expansion Coefficient with the Zeno-Line and Critical-Point Parameters for Liquids

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# Connection between the Isobaric Thermal Expansion Coefficient with the Zeno-Line and Critical-Point Parameters for Liquids

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**ABSTRACT:** We have found the expression which connects the values of volumetric thermal expansion coefficients under low temperatures and pressures with the critical-point and Zeno-line parameters. The calculations based on this expression for different substances ($NH_3$, $CO_2$, hexane, Hg, and Cs) are in good agreement with experimental data at relatively low temperatures.
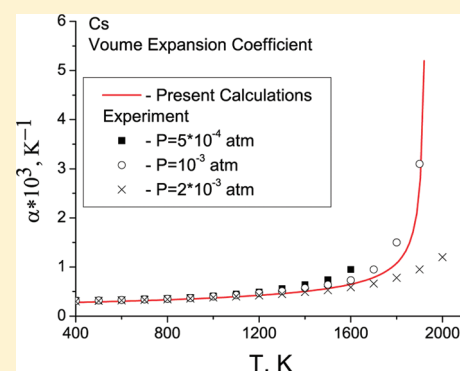
## INTRODUCTION

The search for the similarity and the unifying principles in the description of the variety of the thermodynamic properties of complex matter is the key point of statistical physics.[1] Different substances can exhibit general well-known features, such as the principle of corresponding states or the law of rectilinear diameter. These features are referred to as similarity laws. Among different similarity laws the regularity concerning the Zeno line (ZL) has been widely studied recently as far as it appears to be valid for a wider class of real substances than others.[2-9] This regularity characterizes the states where the compressibility factor is unity, i.e., $Z = P/(\rho k_B T) = 1$, where $P$ is the pressure, $\rho$ is the particle density, $T$ is the temperature in K, and $k_B$ is the Boltzmann constant. The states belonging to ZL can form a straight line on the density–temperature plane.

The similarity laws allow one to set a correspondence between different thermodynamic values without explicit use of the equation of state. Thus, the universal expression for the description of the liquid branch of the binodal line for a wide group of real substances has been obtained in refs 2 and 3 on the basis of ZL similarity. It was shown that the extension of the binodal in the $T \rightarrow 0$ domain has to tend to the Zeno line asymptotically; i.e., the Zeno line is the tangent to the liquid binodal at $T \rightarrow 0$. This fact together with the three-term Guggenheim equation gave rise to the binodal equation mentioned above. The Zeno-line parameters can be determined from the condition that the binodal, thusly constructed, deviates from experimental binodal data (or their low temperature part) minimally.

Another well-known fact is that at low temperatures the liquid binodal branch practically coincides with the set of isobaric curves in the density–temperature plane. This behavior of isobars can be explained by the low compressibility, which is

characteristic of many substances in the liquid state. It also allows one to calculate the volumetric expansion coefficients (VEC) (which are also referred to as the isobaric thermal expansivities) for liquids if some expression for the liquid branch of the binodal is known. As far as we have the binodal equation obtained on the basis of ZL regularity, it is interesting to calculate VEC by means of such equation.

These calculations are the main goal of this study. Below we will show that the above considerations give the opportunity to set a connection between the VEC and parameters characterizing the Zeno line and the critical point. We will apply this unconventional method for VEC calculations first for van der Waals equation and then at room temperature for some nonmetallic substances ($NH_3$, $CO_2$, hexane) and metals (Hg, Cs). We will compare the calculated values of the VEC at room temperature with experimental data. We will also compare our VEC for Cs and hexane with the measurements at different temperatures.

## BASIC EQUATIONS

The volumetric expansion coefficient $\alpha$ can be defined as

$$\alpha = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P \tag{1}$$

The derivative in eq 1 is taken along the isobar line in the density–temperature plane. Consequently, in general, to calculate $\alpha$, it is necessary to know the isobar equation for different substances and different pressures. But there is empirical fact,

mentioned in the Introduction, that the set of the isobar curves goes very close to the liquid binodal line at the temperatures, exceeding the triple point temperature ($T_t$). Practically, for a very wide wealth of substances, the isobars of various liquids have the segments almost coinciding with the liquid binodal line. Consequently, the derivative in eq 1 in this case can be taken along the coexistence curve. Therefore, if one has some general relation for the liquid binodal branch $\rho_{binodal} = \rho(T)$, it is possible to substitute it in eq 1. Guggenheim[10] was possibly the first, who offered such a relation in a form of three-term equation. For liquid branch it is

$$\rho(T) = \rho_c + A\tau + B\tau^\beta \qquad \tau = 1 - T/T_c \qquad (2)$$

The subscript "c" refers to the critical point. The critical index $\beta$ can be different. For instance, for van der Waals model it is 1/2 (see ref 11). But for many real substances it is approximately 1/3 according to the measurements and critical phenomena theory ("Ising class", ref 11, more exact value $\beta = 0.326$). Various techniques[12,13] were developed to determine the coefficients $A$ and $B$. It was shown that for a variety of substances it is possible to fit $A$ and $B$ to get excellent agreement with experiments (108 substances were checked in ref 12). So the three-term eq 2 can be considered as the required general relation to substitute it into eq 1. The only problem is an arbitrariness of the coefficients $A$ and $B$. But the fundamental fact, mentioned in the Introduction (see refs 2, 3, and 14) is that the Zeno line is the tangent to the liquid branch of the binodal at $T \rightarrow 0$. This fact allows us[2,3] to remove the uncertainty of $A$ and $B$, as far as eq 2 at $T \rightarrow 0$ should have ZL as the asymptote. Therefore

$$A = \cfrac{\dfrac{T_c}{T_B}\rho_B - \beta\rho_B + \beta\rho_c}{1 - \beta}$$

$$B = \cfrac{\left(1 - \dfrac{T_c}{T_B}\right)\rho_B - \rho_c}{1 - \beta} \qquad (3)$$

The subscript "B" is related to the ZL parameters. (ZL equation is $\rho/\rho_B + T/T_B = 1$; see eq 4 below and refs 2 and 3). So now we have the general relation to describe the liquid branch of binodal independently of a particular substance. This expression is exact at $T \rightarrow 0$ (see refs 2, 3, and 14). Consequently, it can be used in eq 1 as discussed above. Using eqs 1−3, one can easily obtain

$$\alpha = \frac{A + B\beta\tau^{\beta-1}}{T_c(\rho_c + A\tau + B\tau^\beta)} \qquad (4)$$

At critical point, where $\tau \rightarrow 0$ ($T \rightarrow T_c$), eq 4 gives $\alpha \rightarrow \infty$, as far as usually $\beta < 1$. For the opposite case when $T \rightarrow 0$ eq 4 can be simplified. In this case, the binodal transforms into ZL equation and for $\alpha$ we have

$$\rho(T) = \rho_B(1 - T/T_B)$$
$$\alpha(T) = \frac{1}{T_B(1 - T/T_B)} \qquad (5)$$

Equation 5 allows one to estimate the Boyle temperature using the values of $\alpha$ measured under low temperatures $T$. It follows from eq 5 that

$$T_B = \frac{1 + \alpha T}{\alpha} \qquad (6)$$

The triple point temperature is $T_t \approx 0.2T_B$ for the substances obeying the principle of corresponding states.[5,6] So the value of the VEC at the triple point is $\alpha \approx 1.25/T_B$. Formally, we can also consider the limit $\tau \rightarrow 1$ ($T \rightarrow 0$), when the ZL tends asymptotically to the liquid branch of binodal. This is a non-physical limit because a liquid solidifies when $T < T_t$, where $T_t$ is the triple point temperature. Nevertheless, consideration of this limit gives the opportunity to estimate a lower limit value of the VEC for liquid, when $T \rightarrow 0$ and $P \rightarrow 0$. It follows from eqs 5 that

$$\alpha_0 = \lim_{\substack{T \rightarrow 0 \\ P \rightarrow 0}} \alpha(T) = 1/T_B \qquad (7)$$

This important relation sets a connection between the limit value of the VEC and the inverse Boyle temperature.

We should note that, although three-term eq 2 allows one to describe the binodal of very different substances, it has no rigorous theoretical grounds. But it is possible to show that eq 7 can be also obtained under some suggestions by means of recently developed isomorphism[8,9] between the lattice gas (LG) and real fluids. In refs 8 and 9 it was offered to set correspondence between the phase diagram in $\rho-T$ plane of real fluids and the phase diagram of LG in $x-t$ plane ($x$ is the LG density, $t$ is the LG temperature). Namely, the isomorphic relations, proposed in ref 8 are

$$\rho(x,t) = \frac{\rho_B zx}{1 + zt} \qquad T(x,t) = \frac{ztT_B}{1 + zt}$$
$$z = \frac{T_B}{T_B - T_c} \qquad (8)$$

Then, using eq 8, the derivative $d\rho/dT$ can be expressed as

$$\frac{d\rho}{dT} = \frac{\rho'_x\left(\dfrac{dx}{dt}\right) + \rho'_t}{T'_x\left(\dfrac{dx}{dt}\right) + T'_t} \Longrightarrow -\frac{1}{\rho}\frac{d\rho}{dT}$$

$$= \frac{1 + zt}{T_B} + \frac{(1 + zt)^2}{zT_B}\left(-\frac{1}{x}\frac{dx}{dt}\right) \qquad (9)$$

At $T \rightarrow 0$ we have $t \rightarrow 0$ and

$$-\frac{1}{\rho}\frac{d\rho}{dT} = \frac{1}{T_B} + \frac{1}{zT_B}\left(-\frac{1}{x}\frac{dx}{dt}\right) \qquad (10)$$

So to get VEC we need to take derivatives in eq 10 at constant pressure. The third law of thermodynamics gives for LG that $(dx/dt)_{\pi=const} = 0$, $t \rightarrow 0$, where $\pi$ is the lattice gas pressure. (This derivative is not zero in the case of some other system, for instance van der Waals.) Now it is necessary to suppose that the condition $\pi = const$ give rise to $P = const$ ($P$ is the pressure of real fluids). In ref 9 the transformation $\pi \rightarrow P$ between the pressures was offered. The equation of states (EOS) for $\pi$ and $P$ were also considered. But both the transformation and EOS are not universal. Earlier we have made an analogous procedure[24] and have found the disagreement between the calculated and experimental values of the pressures at isotherms of real substances. So the relation $\pi = const \Longrightarrow P = const$ remains supposition. Under this condition eq 7 directly follows from eq 10.

As far as the binodal curve, the isobars and ZL (ZL is the binodal asymptote at $T \rightarrow 0$) coincide only at low
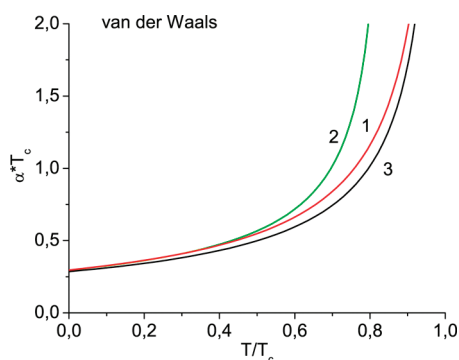
**Figure 1.** Reduced VEC $\alpha T_c$ versus reduced temperature $T/T_c$ for the van der Waals equation. Line 1 is calculated by eq 17; lines 2 and 3 are calculated by exact eq 14 at $P = 0$ and $P = P_c$, respectively.

temperatures, the applicability of our results (eqs 4−7) is limited at high temperatures region. Below we confirm this statement considering model van der Waals system and real liquids.

## ■ VAN DER WAALS EQUATION

The van der Waals equation and corresponding compressibility is

$$P = \frac{\rho T}{1 - \rho b} - a\rho^2$$

$$Z = P/\rho T = \frac{1}{1 - b\rho} - \frac{a\rho}{T} \qquad (11)$$

Let us introduce the Boyle units: $\rho_B = 1/b$ ; $T_B = a/b$ ; $P_B = a/b^2$. Then the reduced pressure, density and temperature are: $P' = P/P_B$ ; $\rho' = \rho/\rho_B$ ; $T' = T/T_B$. In these reduced units the critical parameters are $P'_c = 1/27$; $\rho'_c = 1/3$; $T'_c = 8/27$. Then eq 11 can be transformed as

$$P' = \frac{\rho' T'}{1 - \rho'} - (\rho')^2 \qquad Z = \frac{1}{1 - \rho'} - \frac{\rho'}{T'} \qquad (12)$$

The equation of the line where $Z = 1$ (Zeno line) can be written as

$$\rho' + T' = 1 \qquad (13)$$

From eq 11 one can easily obtain two parametric equations which determine the dependence of the VEC on temperature

$$\alpha = \frac{\rho'}{(P' + \rho'^2) - 2\rho'^2(1 - \rho')}$$

$$T' = \frac{(P' + \rho'^2)(1 - \rho')}{\rho'} \qquad (14)$$

If $\rho' \rightarrow 1$, then $\alpha \rightarrow 1/(P' + 1)\alpha \rightarrow 1/(P+1)$ and $T' \rightarrow 0$. The pressure does not affect the dependence $\alpha(T')$, if the inequality

$$P' \ll (\rho')^2 \qquad (15)$$

is valid. This means that the thermal and potential parts of the pressure are almost equal. The thermal part is slightly greater than the potential part. Such compensation takes place along the binodal line. So we can calculate the low temperature part of the VEC using the latter. Binodal equation (eqs 2 and 3 with $\beta = 0.5$)

for the van der Waals system looks like ($\tau = 1 - T'/T'_c = 1 - 27T'/8$)

$$\rho'(T') = 1/3 - 2\tau/27 + 20\tau^{1/2}/27 \qquad (16)$$

Substitution of eq 16 to eq 1 gives

$$\alpha(T') = -\frac{3}{4} \frac{1 - 5/\sqrt{\tau}}{1 - 2\tau/9 + 20\sqrt{\tau}/9} \qquad (17)$$

When $T' \rightarrow 0$ ($\tau \rightarrow 1$), we can obtain from eq 17

$$\alpha(T') = \frac{1}{1 - T'} \qquad (18)$$

Or in ordinary units

$$\alpha(T) = \frac{1}{T_B - T} \qquad (19)$$

The limit value of the VEC is equal to $\alpha_0 = 1/T_B$, where $T_B/T_c = 27/8$. Now we can compare the approximate solution eq 17 with the exact one (eq 14). Such comparison is presented in Figure.1 for dimensionless value $\alpha^* T_c$.

The VEC calculated according to eq 17 (line 1) and according to exact eq 14 (line 2, 3) are in good agreement at low temperatures, at $T/T'_c < 0.4$ and different pressures ($P = 0$, $P = P_c$). At higher temperatures we can see that our equations are no more applicable.

We should note that besides high temperatures our regularities can have two more limitation. In ref 5 we have investigated the behavior of ZL in various model systems, like square-well potentials, generalized Lennard-Jones (or Mie m-n) potentials, etc. We have found that the similarity relations concerning ZL, which is used in present investigation of VEC, are invalid in two cases. First one arises, when the liquid−vapor transition in some system becomes metastable with respect to freezing. It is the case when the potential is too "short-ranged". For instance, for the square-well potential, the width of well becomes too narrow (if the hard wall is located at $r = \sigma$, then at a width less than $1.25\sigma$ there is metastability; see ref 5). Another case is connected with the opposite situation when the potential is too "long-ranged". In this case the triple point is located too close to the critical point. So the liquid region becomes too narrow and the ZL is no more the straight line. For real substances these "exotic" potentials can arise in so-called "soft matter", where the range of potentials can be "tuned". Colloids are excellent examples. In the next section we will not study soft matter but will consider only usual substances. So we will have only one limitation: high temperatures.

## ■ REAL SUBSTANCES

Most experimental data describing VEC for different substances are given in handbooks at room temperature and atmospheric pressure. Analysis fulfilled in the previous section can be used to compare some of these reference data with predictions following from eq 4. The results of such comparison are collected in Table 1.

In Table 1, columns 2−4 correspond to the critical and Zeno-line parameters taken from ref 4. Columns 5−7 contain experimental data: temperature ($T$), pressure ($P$), and experimental value of VEC ($\alpha_{exp}$) at these ($T$, $P$) according to different works (references presented in column 7). Columns 8 and 9 show the calculated values of VEC: $\alpha$ is calculated according to eq 4 with

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**Table 1. Temperatures ($T$, K), Densities ($\rho$, g/cm$^3$), and VEC ($\alpha$, K$^{-1}$)[a]**

|  | col 1 | col 2 | col 3 | col 4 | col 5 | col 6 | col 7 | col 8 | col 9 | col 10 |
|---|---|---|---|---|---|---|---|---|---|---|
| subs. | $T_B$ | $\rho_B$ | $T_c$ | $\rho_{cr}$ | $T$, K | $P$, atm | $\alpha_{exp}\cdot 10^3$ | $\alpha\cdot 10^3$ | $\alpha_0\cdot 10^3$ | $T^*_B$ |
| NH$_3$ | 936 | 0.95 | 405 | 0.23 | 293 | 1 | 2.45[15] | 2.5 | 1.1 | 710 |
| C$_6$H$_{14}$ | 1230 | 0.9 | 508 | 0.23 | 303,1 | 0.0162 | 1.35[16] | 1.5 | 0.81 | 1044 |
| CO$_2$ | 741 | 1.8 | 304 | 0.47 | 298 | 1 | 20.7[17] | 20.6 | 13.5 | 346 |
| Hg | 6300 | 14.4 | 1750 | 5.8 | 298 | 1 | 0.18[18] | 0.18 | 0.16 | 5854 |
| Cs | 4120 | 1.96 | 1930 | 0.39 | 400 | 0.006 | 0.27[19] | 0.28 | 0.24 | 4104 |

[a] $T^*_B$ is obtained by inverting eq 7.
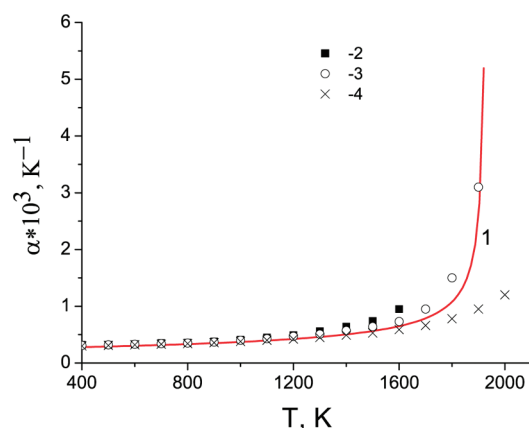


**Figure 2.** VEC versus temperature for Cs. Line 1 is calculation by eq 4, and symbols are the experimental data (ref 19): 2, $P = 5 \times 10^{-4}$ atm; 3, $P = 1 \times 10^{-3}$; 3, $P = 2 \times 10^{-3}$.
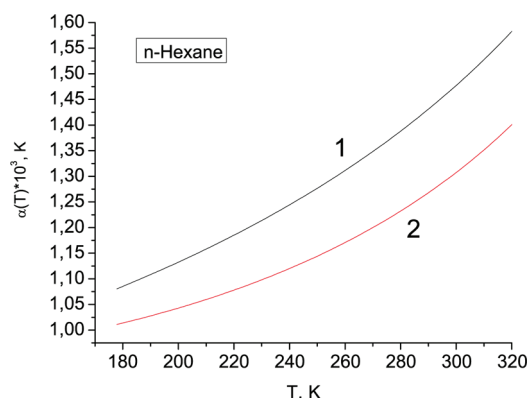


**Figure 3.** VEC versus temperature for $n$-hexane. Line 1 is calculation by eq 4; line 2 is calculation by eq 23.

$\beta = 1/3$; $\alpha_0$ is the limit value (eq 7) when $T \to 0$. Column 10 gives the value of the Boyle temperature obtained by inverting eq 7. The latter are in reasonable agreement with the earlier calculated values with the exception of CO$_2$. The temperature of the VEC measured for CO$_2$ is very close to the critical one, i.e., $T/T_c \sim 1$ (Table 1) and eq 7 becomes inapplicable in this case. However, the more complicated eq 4 gives a good result for CO$_2$. One can see from Table 1 that the calculated values of the VEC are in good agreement with experiment.

The values of the VEC for Cs in wide range of temperatures and pressures have been measured in ref 19. Some of these data are presented in Figure 2. Symbols 2−4 correspond to the experimental isobars at pressures $5 \times 10^{-4}$, $1 \times 10^{-3}$, and $2 \times 10^{-3}$ atm, respectively. In Figure 2, the solid line presents the calculation according to the method discussed above. The latter agrees well with the experimental data at low temperatures.

We should note that over time, VEC in compressed liquids was investigated for a variety of substances. Presently, many experimental and theoretical works provide a variety of fitting expressions for VEC data under different conditions.[20−22] In particular, for $n$-hexane the formula adequately describing the $\alpha(P)$ isotherms ($P$ is the pressure) was suggested by Pruzan[21]

$$\alpha(P) = \frac{\alpha^*}{\sqrt{P - P_{sp}}} \quad (20)$$

Here $P_{sp}$ is pressure along the spinodal curve and $\alpha^*$ is the parameter depending on the temperature along the isotherm only. This expression was generalized for other substances

and for nonisothermal states in ref 22 as

$$\alpha = \alpha(P, T) = \frac{\alpha^*(T)}{\sqrt{P - P_{sp}(T)}}$$

$$\alpha^*(T) = \alpha_0 \left( 1 + A \left[ \frac{P_{sp}(T) - P}{p^*} \right]^\omega \right) \quad (21)$$

Equation 21 was used in ref 23 for a number of substances. Here we consider only $n$-hexane, for which $\alpha_0 = 6.89$ (MPa)$^{0.5}$/kK, $A = 1$, $p^* = -201.6$, $\omega = 0.78$ (see ref 23). For the pressure along the spinodal $P_{sp}(T)$ there is an accurate expression, which correctly describes extensive experimental data (see ref 24)

$$P_{sp}(T) = P_{vap}(T) + p^* \left[ \frac{(1 - T/T_c)^{3\varepsilon}}{(T/T_c)} \right]^{1/2} \quad (22)$$

Here $P_{vap}(T)$ is the pressure along the liquid branch of the binodal, $T_c$ is the critical temperature ($T_c = 507.82$ K for $n$-hexane[15]), $p^*$ is the same parameter as in eq 21, $3\varepsilon = 3.65$ for $n$-hexane.[22] eq 22 was used in ref 22 together with eqs 20 and 21 to describe VEC for various substances. It was pointed out in ref 22 that the term $P_{vap}(T)$ is important only near the critical point. For example, for $n$-hexane near the triple point $T_t = 177.83$ K, $P_{tr} \approx P_{vap} = 1.28 \times 10^{-6}$ MPa. At the same time the second term in eq 22 gives us $P_{sp}(T_{tr}) - P_{vap}(T_{tr}) = -155$ MPa. Even at $T = 303.15$ K (it is the temperature of the isotherm where the VEC of $n$-hexane was measured in ref 16) $P_{sp}(303.15) - P_{vap}(303.15) = -49.5$ MPa, while $P_{vap}(303.15) = 0.025$ MPa (see ref 15). So we can claim that

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the second term in eq 21 is much greater (by absolute value) than the first one in the temperature range from the triple point temperature $T_t$ to room temperature $T \approx 300$ K. On the other hand, if we look at eqs 21, we can see that for pressures $P \leq P_0 \sim 1$ MPa the coefficient $\alpha = \alpha(P,T)$ is practically independent of the pressure as far as $P \ll |P_{sp}(T)|$, i.e.

$$\alpha(P,T) = \frac{\alpha^*(T)}{\sqrt{P - P_{sp}(T)}} \approx \frac{\alpha^*(T)}{\sqrt{-P_{sp}(T)}}$$

$$P < 1 \text{ MPa}, \ T_t \leq T \leq 300 \text{ K} \tag{23}$$

This VEC also depends only on temperature, similarly to the VEC described by eq 4 of this paper. Equation 23 at room temperature for $n$-hexane gives the VEC that is 10% greater than the experimental one (see Table 1). It is interesting to compare the values of the VEC calculated according to eqs 4 and 23 at low temperatures. In Figure 3 these two values of the VEC are presented at $T_t \leq T \leq 320$ K for $n$-hexane. At $T = T_t = 177.83$ K the difference between them is about 6%. The difference slightly increases with temperature. This behavior is similar to that of the VEC for Cs in Figure 2.

## ■ CONCLUSION

In this work, a new step is made toward the better understanding of the similarity laws and thermodynamics of real liquids. We have shown that the volumetric expansion coefficient of real liquid at low temperatures and pressures can be calculated on the basis of its binodal expressed through the critical-point and Zeno-line parameters. Moreover, eq 4 sets a correspondence between low temperature thermodynamic functions (VEC) accessible from experiment and the critical-point and Zeno-line parameters. The latter often lie in the domain of the phase diagram inaccessible for direct experimental measurements. This allows us to use these low temperature thermodynamic data for more precise definition of the critical-point and Zeno-line parameters.

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