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Investigation of the Scaling Properties of a Binary Solution Near the Critical Vaporization State

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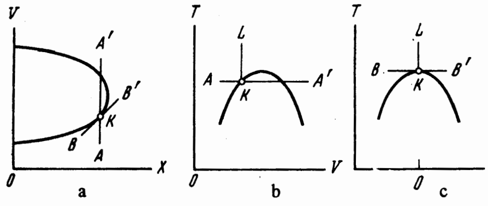
A simple and natural method of extending the hypothesis of homogeneity of the thermodynamic functions of a one-component system[1,2] to a binary mixture near the critical vaporization state is proposed. Data obtained by investigating the height and temperature behavior of the light scattering power on a n-pentane-cyclopentane solution at $T > T^{cr}$ are used for an experimental verification of the homogeneous properties of binary systems.

THE considerable recent progress in the knowledge of the properties of single-components systems near the critical liquid-vapor state is due primarily to the development of the so-called scaling theories, the Widom and Griffiths homogeneity hypothesis[1,2] and the Kadanoff-Pokrovskiĭ scalaing theory[3,4].

Several attempts were made[5-8] to generalize the main results of the scaling theories to include binary mixtures. Common to all these investigations is the assumed existence, in the three-dimensional space of independent variables, of a certain section containing the critical point, within which the thermodynamic properties of the solution can be described with the aid of a scaling equation of state which is a formal analog of the corresponding equation for single component system. Certain discrepancies in the definition of this isomorphic section are connected with the choice of the independent thermodynamic variables and the limiting conditions imposed on the additional degree of freedom. In addition, there is an acute shortage of experimental research data needed to verify the validity of the isomorphism assumption and to determine which of the schemes is preferable.

FUNDAMENTAL THERMODYNAMIC PREMISES

The liquid-vapor coexistence surface of a binary mixture has a saddle-like shape and goes over in the limiting cases $X \to 0$ and $X \to 1$ (X is the molar fraction of the second component) into the coexistence curves of the first and second components, respectively. The interactions of this surface with the planes $T = \text{const}$ and $X = \text{const}$ are shown in Figs. 1a and 1b.



a b c

FIG. 1. Sections of the T—V—X surface of the coexistence of a liquid—vapor binary solution: a) section $T = T^{cr}$; b) section $X = X^{cr}$; b) section parallel to the T axis and passing through the critical isotherm—isobar BB'. K—critical point; AA'—direction along which $T = T^{cr}$ and $X = X^{cr}$; KL—direction along which $X = X^{cr}$ and $V = V^{cr}$; $v = V - V^{cr} = (\partial V/\partial X)^{cr}_{P,T}(X - X^{cr})$.

A characteristic feature of both intersections is that the critical point K does not coincide with their vertices[10].

Let us consider the inclined section of the T—V—X surface (Fig. 1c), which is parallel to the T axis and passes through the critical isotherm—isobar which is asymptotically, near the critical state, a straight line (BB' on Fig. 1a) with slope $(\partial V/\partial X)^{cr}_{P,T}$. This section is remarkable in that its vertex corresponds to the critical state K, and that near the critical point this section coincides with the coexistence curve, i.e., the branches of this section describe the experimentally observed coexisting phases in which the pressure and temperature coincide, while the composition and the molar volume (density) are different and are connected by the relation $V_{liq} - V_{vap} = (\partial V/\partial x)^{cr}_{P,T}(X_{liq} - X_{vap})$. The plane of the inclined section under consideration contains the critical isochore—isoconcentrate. According to our assumption, it is precisely this direction which is the analog of the critical isochore of the pure substance.

When the solution is diluted ($X \to 0.1$), the section in question goes over into the T—V plane of the corresponding pure component.

Any binary-mixture state described by a point in the P—V—T--X space and located near the critical point can be regarded as specified if the following conditions are satisfied:

a) We know the value of X^{cr} through which the inclined section containing the point corresponding to the chosen state of the mixture passes.

b) The distance from the point under consideration to the critical state in the plane of the inclined section ($X - X^{cr}$ and $T - T^{cr}$) is known.

The change of pressure resulting from a slight deviation from the critical state, within the limits of the considered inclined section, is described by the relation $P - P^{cr} = (\partial P/\partial T)^{cr}_{V,X}(T - T^{cr})$.

It seems to us that it is perfectly natural to expect the mixture states located in the considered inclined section to exhibit homogeneous properties in the sense of the Widom-Griffiths homogeneity hypothesis, i.e., the scaling equation of state of the solution, which is a formal generalization of the corresponding equation for the pure substances[2], should be written in the form

$$\Delta\mu = \mu(\tau, x) - \mu(\tau, 0) = x|x|^{\delta-1}f(\tau|x|^{-1/\beta}), \tag{1}$$

where $\mu = \mu_2 - \mu_1$ is the difference between the chemical potentials of the solution components, $\mu(\tau, 0)$ is the value of μ on the critical isochore—isoconcentrate, x is the deviation of the composition from the critical value, $\tau = t + C(\partial P/\partial T)^{cr}_{V,X}$, t is the thermodynamic shift and constitutes the sum of the temperature deviation and the associated deviation of the pressure from the critical value, δ and β are critical exponents describing, respectively, the shapes of the critical isotherm—isobar and of the coexistence curve lying in the plane of the inclined section considered above, C is a coefficient that depends on X^{cr}, and f is a universal function of the argument $\tau|x|^{-1/\beta}$ with known asymptotic values[2].

An analysis carried out within the framework of the classical theory of critical phenomena[10] shows that $1 + C(\partial P/\partial T)^{cr}_{V,X} > 0$ even though the coefficient $C(X^{cr})$ can be positive as well as negative.

On approaching the critical isochore—isoconcentrate we have $\tau|x|^{-1/\beta} \to \infty$, and the function $f(\tau|x|^{-1/\beta})$ diverges in this case like $(\tau|x|^{-1/\beta})^{\beta(\delta-1)}$. It is then more convenient to use the scaling equation of state in the form proposed in[11].

Dividing (1) by $|\tau|^{\beta\delta}$, we obtain

$$\Delta\mu/|\tau|^{\beta\delta} = m(y), \tag{2}$$

i.e., the scaling chemical potential of the solution, $m = \Delta\mu/|\tau|^{\beta\delta}$, is a function of only the scaling concentration $y = x|\tau|^{-\beta}$. In the limiting cases $y \to 0$ and $y \to \infty$, the function m(y) can be represented in the form of certain power series. For the vicinity of the critical isochore—isoconcentrate, where $y \to 0$, we have

$$m(y) = a_1 y + a_3 y^3 + a_5 y^5 + \dots \tag{3}$$

Near the critical isotherm—isobar we have $y \to \infty$ and in this case

$$m(y) = b_1 y^\delta \pm b_2 y^{\delta-1/\beta} + b_3 y^{\delta-2/\beta} \pm \dots \tag{4}$$

The minus sign corresponds to the case $\tau < 0$.

Differentiating Eq. (2) with respect to the concentration, we obtain

$$(\partial\mu/\partial X)_{P,T}|\tau|^{-\gamma} = m'(y), \tag{5}$$

where $\gamma = \beta(\delta - 1)$. Thus, the product of the susceptibility by the thermodynamic shift raised to the power γ is a universal function of the scaling concentration γ.

CONNECTION BETWEEN THE SCATTERING ABILITY AND THE THERMODYNAMIC PROPERTIES OF A SOLUTION. VERIFICATION OF THE HOMOGENEITY HYPOTHESIS

Comparing the equation for the equilibrium of a solution in a gravitational field[10]

$$(\partial\mu/\partial X)_{P,T}dX = -V(\partial\rho/\partial X)_{P,T}g dZ$$

with the expression for the constant of scattering by composition fluctuations in the Rayleigh-Einstein approximation

$$R_{RE} = \frac{\pi^2 k_B T}{2\lambda^4}\frac{V}{(\partial\mu/\partial X)_{P,T}}\left(\frac{\partial\varepsilon}{\partial X}\right)^2_{P,T},$$

we obtain

$$R_{RE} = -\frac{\pi^2 k_B T}{2\lambda^4 g}\left(\frac{\partial\rho}{\partial X}\right)^{-1}_{P,T}\left(\frac{\partial\varepsilon}{\partial X}\right)^2_{P,T}\frac{dX}{dZ} \tag{6}$$

(ρ is the density of the solution, ϵ is the dielectric constant, Z is the vertical coordinate, g is the acceleration due to gravity, and λ is the wavelength), i.e., in the Rayleigh-Einstein approximation the intensity of the light scattered by the composition fluctuations is proportional to the macroscopic composition gradient produced under the influence of the gravitational field.

We used the previously described experimental setup[12] to investigate the height dependence of the intensity of light scattering in a solution containing a 0.182 molar fraction of cyclopentane in n-pentane at 15 positive temperature deviations from the critical temperature $T^{cr} = 475.8°K$. The chamber was filled in such a way that at $T = T^{cr}$ the meniscus separating the liquid and vapor phases vanished in the middle of the chamber. At $T > T^{cr}$ the layer located at the level where the meniscus vanished had maximum scattering ability and possessed the critical values of the density and composition.

The measurement results were corrected for the attenuation of the incident and scattered light fluxes and also for the contribution of the secondary scattering. The secondary scattering was calculated with allowance for the polarization properties of the incident and scattered light[13]. The error in the determination of the intensity of single scattering did not exceed 4—5%. The measurement results are presented in the table.

When the critical state is approached, the quantity $(\partial V/\partial P)_{T,X}$, which determines the scattering of light by the pressure fluctuations[1], diverges weakly like $\tau^{-\alpha}$ (α is the exponent describing the temperature behavior of the specific heat and its value is 0.125 in the pure substance[14]). At the same time, the susceptibility $(\partial\mu/\partial X)^{-1}_{P,T}$ increases like $\tau^{-\gamma}$ ($\gamma \approx 5/4$), i.e., the decisive contribution to the total intensity of light scattering in the solution near the critical state is made by scattering from composition fluctuations.

To describe the experimentally observed height and temperature behavior of the intensity of light scattering, we use the Ornstein-Zernike formula[2], which can be represented in the form

$$I_{OZ}^{-1}(h, \tau) = I_{RE}^{-1}(h, \tau) + I_{cr}^{-1}(h, \tau), \tag{7}$$

where h is the deviation, in height, from the level with maximum scattering ability. Near the critical state we can put

$$\left(\frac{\partial\rho}{\partial X}\right)_{P,T} \approx \left(\frac{\partial\rho}{\partial X}\right)^{cr}_{P,T}, \quad \left(\frac{\partial\varepsilon}{\partial X}\right)_{P,T} \approx \left(\frac{\partial\varepsilon}{\partial X}\right)^{cr}_{P,T}, \quad I_{cr}(h,\tau) \approx I_{cr}(0,0). \tag{8}$$

By investigating the temperature behavior of the quantity $I_{OZ}^{-1}(0, \tau)$, i.e., the critical isochore—isocon-

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[1] The intensity of scattering by pressure fluctuations is proportional to the quantity.

$$\left(\frac{\partial\varepsilon}{\partial P}\right)^2_{T,X}\left(\frac{\partial V}{\partial P}\right)^{-1}_{T,X} = \left(\frac{\partial\varepsilon}{\partial V}\right)^2_{T,X}\left(\frac{\partial V}{\partial P}\right)_{T,X}$$

[2] The deviations from the Ornstein-Zernike theory predicted in[15] have not been observed to date. This allows us to assume that the use of formula (7) does not lead to significant errors in the subsequent results.

Height dependences of the intensity of light scattering in an n-pentane–cyclopentane solution at different temperatures

h, mm	$T-T^{cr}$, deg														
	0,02	0,09	0,17	0,19	0,34	0,48	0,57	0,72	0,91	1,25	1,64	2,14	2,68	3,15	3,79
−18	44	44	45	46	49	52	54	58	65	70	71	70	68	59	51
−16	48	48	50	51	54	58	61	65	73	79	80	78	71	62	52
−14	54	54	57	58	62	67	71	76	84	92	91	84	74	64	53
−12	63	63	66	66	71	78	83	90	97	107	101	91	76	66	55
−10	75	75	77	77	83	94	100	104	114	124	111	96	78	67	57
−8	90	91	96	95	102	117	123	126	135	141	121	102	80	69	57
−6	118	118	125	122	133	140	152	162	166	159	132	107	82	70	58
−4	164	175	180	180	190	203	217	216	203	178	142	109	83	71	58
−2	322	375	390	382	346	330	320	295	247	195	150	111	84	72	59
0	1890	1510	1121	1071	710	531	456	369	289	204	153	112	84	72	59
+2	323	390	394	392	356	328	332	298	255	196	149	110	83	71	58
+4	153	154	165	161	180	186	200	206	202	172	138	106	81	70	57
+6	108	111	110	109	125	132	146	144	152	149	127	100	77	68	55
+8	85	88	86	86	96	101	111	110	123	124	116	94	74	67	53
+10	73	74	72	72	80	83	88	90	98	107	104	88	71	65	52
+12	63	62	62	61	70	69	75	76	85	94	93	81	69	62	50
+14	54	54	54	54	59	61	65	70	76	81	84	74	66	60	49
+16	47	47	48	48	51	53	58	62	68	71	74	69	64	57	48
+18	42	42	43	42	45	48	49	55	60	62	66	64	61	55	47

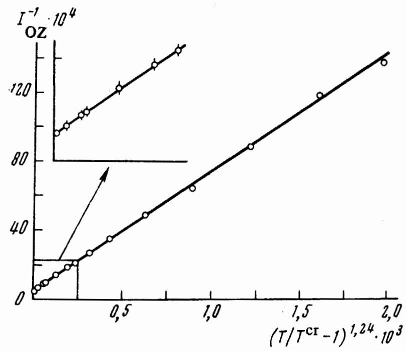

FIG. 2. Dependence of reciprocal intensity of light scattering on the critical isochore–isoconcentrate on $(T/T^{cr}-1)^\gamma$. The experimental points in the temperature interval $0 \leqslant T-T^{cr} \leqslant 0.6°$ are shown on an increased scale.

centrate, we have determined the limiting value $I^{-1}_{cr}(0, 0) = 5.0 \times 10^{-4}$, and we have also established that the quantity $I^{-1}_{RE} \sim (\partial\mu/\partial X)_{P,T}$ varies with temperature in the interval $4.2 \times 10^{-5} \leq (T/T^{cr} - 1) \leq 7.97 \times 10^{-3}$ in accordance with the law $I^{-1}_{Re} \sim \tau^{1.24\pm0.5}$ (see Fig. 2). The obtained value of the exponent $\gamma = 1.24$ is somewhat higher than the vaule $\gamma = 1.20$ obtained for n-pentane[16]. This result is due to a considerable degree to our additional allowance for the secondary scattering produced in the selected layer of matter when the latter is illuminated with light scattered once in the same layer. The corresponding correction at the level h = 0 increases on approaching the critical state, and for layers of matter 1 mm thick it reaches at λ = 5461 Å the value 7–8% at a total relative intensity of secondary scattering 12–13%.

We have converted the experimentally determined values of $I_{OZ}(h, \tau)$ into values of $I_{RE}(h, \tau)$ and integrated the functions $I_{RE}(h, \tau)$ numerically. This enabled us to determine, accurate to within the coefficient

$$k \sim \frac{\pi^2 k_B T}{2\lambda^4}\left(\frac{\partial\rho}{\partial X}\right)^{-1}_{P,T}\left(\frac{\partial\varepsilon}{\partial X}\right)^2_{P,T}$$

the height dependences of the deviations of solution composition from the critical value

$$kx(h,\tau) = \int_0^h I_{RE}(Z,\tau)\,dZ$$

(see formulas (6)--(8)).

To estimate the critical exponent δ, we have investigated the temperature behavior of the dependence of kx on h and the dependence of $I^{-1}_{RE} \sim (\partial\mu/\partial X)_{P,T}$ on kx. The investigation has shown that in the case of the solution investigated by us the numerical value of the exponent δ ranges from 4.8 to 5.2. A detailed description of the procedure used to determine the exponents γ and δ is of independent interest and is the subject of a separate article[13].

In a gravitational field we have $\Delta\mu \sim (M_2 - M_1)h$ (M is the molecular weight), so that we can interpret the obtained height distributions of the deviations of the composition from the critical value as the dependences of the difference between the chemical potentials $\Delta\mu = \mu(\tau, x) - \mu(\tau, 0)$ on the composition deviations $x = X - X^{cr}$, and verify on this basis the validity of our generalization of the homogeneity hypothesis, originally formulated for the thermodynamic functions of pure substances, to include the case of binary mixtures. To this end, we have calculated, accurate to within a constant coefficient, the values of the scaling concentration $kx/t^{\nu/(\delta-1)}$ for heights |h| equal to 2, 4, 6, ..., 18 mm and for 11 temperatures $T - T^{cr} > T^{cr}$ in the interval from 0.34 to 3.79°. The corresponding values of the scaling chemical potentials were also determined, accurate to a constant coefficient, in the form $h/t^{\gamma\delta/(\delta-1)}$. The dependences of the scaling chemical potentials on the scaling concentrations obtained in this manner for different isotherm–isobars were plotted in a single coordinate system (Fig. 3). These dependences coincided within the limits of experimental error and formed a single curve corresponding to the function m(y). The results shown in Fig. 3 were obtained under the assumption that δ = 5. We have also performed the calculations assuming δ = 4.8 and 5.2. Such a variation of the exponent δ does not lead to a noticeable increase of the scatter of the experimental points, and only deforms slightly the entire m(y) curve.

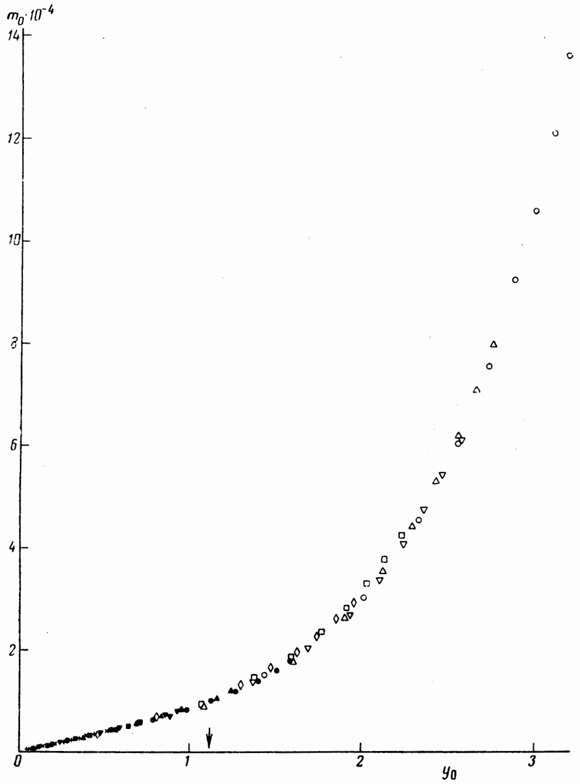

FIG. 3. Dependence of the scaling chemical potential m - $C_1 m_0$, where $m_0 = h/t^{\gamma\delta/(\delta-1)}$ on the scaling concentration $y = C_2 y_0$, where $y_0 = kx/t^{\nu/(\delta-1)}$ at $T > T^{cr}$. The experimental points correspond to the following values of $T - T^{cr}$: ○–0.34, △–0.48, ▽–0.57, □–0.72, ◇–0.91, ●–1.25, ▲–1.64, ▼–2.14, ■–2.68, ×–3.15, +–3.79. Here and in the succeeding figures the arrow designates the value of y_0 corresponding to the condition y = R.

The intensity of light scattering determined directly from the experimental data is proportional in the Rayleigh-Einstein approximation to the susceptibility $(\partial\mu/\partial X)^{-1}_{P,T}$, which makes it possible to verify the scaling law written in the differential form (5). The plots of $(I_{RE}t^{\gamma})^{-1}$ against kx/t^{β} for 11 transcritical temperatures, drawn in a single coordinate system, are shown in Fig. 4. These plots form a single scaling curve m′(y), in full agreement with the hypothesis (5). The scatter of the points does not exceed the experimental errors. It is seen from Figs. 3 and 4 that at small values of y the m(y) dependence is linear, while m′(y) tends to a constant limit as y → 0 (see the expansion (3)).

At small $y \sim kx/t^{\beta}$, the m′(y) dependence, in accordance with assumption (3), takes the form $a_1 + 3a_3y^2 + \ldots$, i.e., it is asymptotically quadratic. To verify this statement, we have plotted the quantity $[I^{-1}(h, \tau) - I^{-1}(0, \tau)]t^{-\gamma}$ against kx/t^{β} for ten temperatures in a doubly logarithmic scale (Fig. 5). Such a choice of variables is equivalent to an upward transfer of the coordinate axis by an amount equal to the intercept of m′(y) at y = 0. Although the scatter of the experimental points increases noticeably with increasing kx/t^{β}, owing to the larger relative errors resulting from the determination of differences of similar quantities, it is clearly seen that at small y the investigated dependence is linear in a double logarithmic scale, with a slope close to 2. At sufficiently large values of y, an increase of the slope is observed, this being a manifestation of the transition from the asymptotic expansion (3), which holds by assumption when

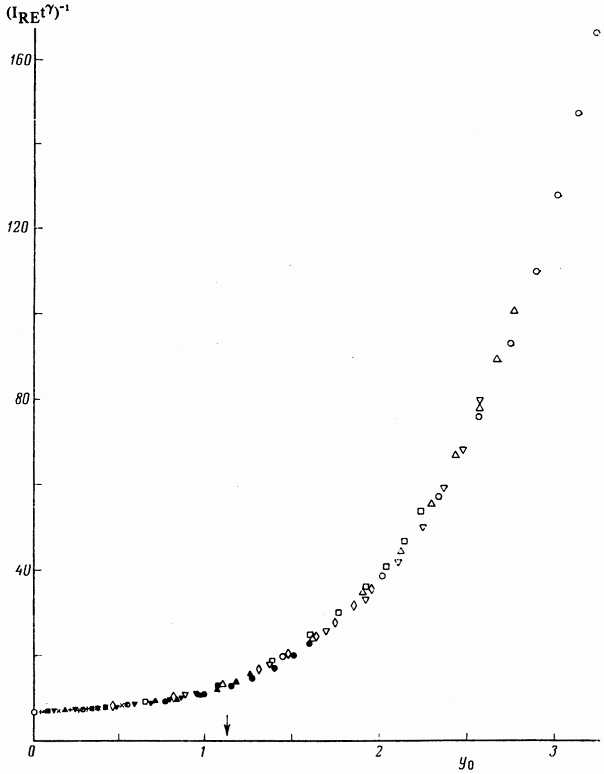

FIG. 4. Dependence of the reciprocal scaling susceptibility $m' \sim (\partial\mu/\partial x)_{P,T}t^{-\gamma}$ on the scaling concentration $y = C^2 y_0$. The symbols for the experimental points are the same as in Fig. 3.

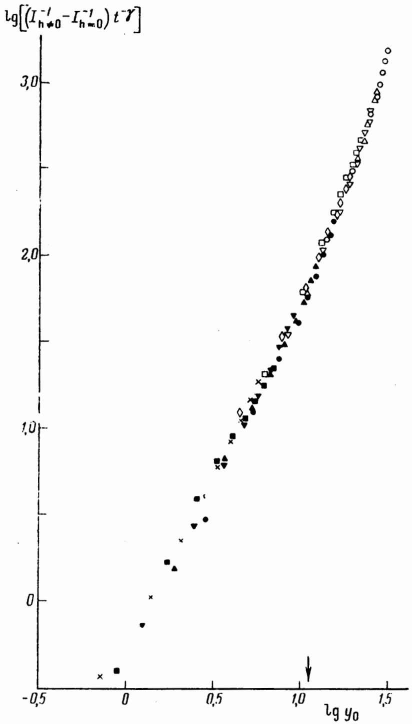

FIG. 5. Plot of $\log[(I^{-1}_{h\neq0} - I^{-1}_{h=0})t^{-\gamma}]$ against $\log y_0$. The symbols for the experimental points are the same as in Fig. 3.

$y \ll R$, to the expansion (4) which is valid when $y \gg R$ (R is the quantity determining the region of convergence of the series (3) and (4)).

Let us see how to determine, without knowing the numerical value of the coefficient k in the expression kx/t^{β}, whether the experimental data lie in the region of "small y" or "large y." Using (5) and the expansions (3) and (4), and recognizing also that $I^{-1} \sim (\partial\mu/\partial X)_{p,T}$, we can show that in the region of applicability of the expansion (3) the quantity $(\partial I/\partial\tau)_{\mu} < 0$, whereas in the region of applicability of the expansion (4) we have $(\partial I/\partial\tau)_{\mu} > 0$. It is natural to expect $(\partial I/\partial\tau)_{\mu} = 0$ at $y = R$. From an examination of the data in the table it can easily be seen that the scattering intensity at a certain fixed height $h \neq 0$ (i.e., at $\Delta\mu = \text{const}$) actually goes through a maximum at a certain temperature $T_{\max}(h) > T^{\mathrm{cr}}$. The value of $T_{\max}(h)$ increases with increasing h. As shown by calculations the value $(kx/t^{\beta})_{\max}$ corresponding to $T_{\max}(h)$ is practically constant for all the isotherm—isobars investigated by us, whereas the ratio $(kx/t^{\beta})/(kx/t^{\beta})_{\max}$ takes on values from 0.05 to 2.9.

We have used the quantities P, T, and X in the present paper as independent thermodynamic variables. From an examination of the conditions for the stability of solutions it follows that the properties of a binary mixture near the critical state of vapor production can be described just as easily by the variables T, V, μ as by the variables P, T, X (see, for example,[17,18]). It is easy to show that $(\partial V/\partial X)^{\mathrm{cr}}_{P,T} \equiv (\partial V/\partial X)^{\mathrm{cr}}_{\mu,T}$, i.e., the isomorphic section chosen by us does not change on going from one set of independent variables to the other. Such a transition reduces simply to a replacement of Eq. (1) by the equivalent equation

$$P(\Theta, v) - P(\Theta, 0) = v|v|^{\delta-1} f(\Theta|v|^{-1/\beta}), \qquad (9)$$

where the thermodynamic shift Θ is equal to $t + C'(\partial\mu/\partial T)^{\mathrm{cr}}_{V,X}t$. The coefficient C', which depends on the choice of the critical point on the critical curve of the solution, tends to zero like $X \ln X$ when the solution is diluted[18], i.e., (9) goes over in natural fashion into the corresponding equation of state of the pure substance.

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