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The Behavior of Thermodynamical Quantities Near the Critical Line of an "Incompressible" Liquid Mixture

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The specific heat at constant volume of a weakly compressible liquid mixture is measured near the critical liquid—liquid equilibrium line. By using the results of previous measurements of the specific heat under constant pressure in the same mixture, other thermodynamical quantities are calculated near the critical line, and it is found that these other quantities exhibit a behavior similar to that of the analogous quantities near the λ transition point in liquid helium. The relations between the singular and regular parts are determined, just as in helium, by the small parameter $R\rho_c dT_K/dP$ (ρ_c is the critical density, T_c is the critical temperature, P is the pressure, and R is the gas constant), whose presence is related to the "incompressibility" of the liquid.

It is well known[1-3] that in systems having second-order phase transition lines (the λ-transition lines in helium, the critical lines of binary mixtures, etc.), the specific heat C_V at constant volume ($C_{V,x}$ in mixtures, where x denotes the impurity concentration) must remain a finite quantity on the transition line. At the same time the concept of the isomorphism of critical phenomena[3] leads to the conclusion that the specific heat C_P at constant pressure ($C_{P,x}$ in binary mixtures) increases without limit at all points on the transition line of such systems (if arguments[4] connected with the presence of a shear modulus in solids are excluded). However, owing to the presence of the small parameter $R\rho_\lambda dT_\lambda/dP \sim -3\times 10^{-2}$ (ρ_λ denotes the density along the λ-line and T_λ is the λ-transition temperature) in the case of the λ-transition, the specific heat C_V "follows" the logarithmic singularity of C_P in the entire experimentally accessible region $\tau(P) = |T - T_\lambda(P)|/T_\lambda(P)$, and the difference $C_P - C_V$ is very small.[5] The condition $R\rho_\lambda dT_\lambda/dP \ll 1$ also leads to singularities, near the λ-line, in the behavior of $(\partial P/\partial T)_V$, the thermodynamical velocity of sound, and a number of other quantities.[6-8]

In an object of a different nature—namely, binary mixtures which separate into two liquid phases—the analogous parameter $R\rho_c dT_c/dP$ (ρ_c is the critical density and T_c is the critical temperature) usually turn out to be close to $R\rho_\lambda dT_\lambda/dP$ in absolute magnitude; therefore here one should anticipate a similar behavior of the thermodynamical quantities.

The results of the first measurements of the specific heat $C_{V,x}$ near the critical line of demixing of a mixture of methanol—cyclohexane are presented in the present communication. Our measurements of the specific heat $C_{P,x}$ in this same mixture (at the saturated-vapor pressure[9] indicated that the following relation holds near the critical point:

$$-\left(\frac{\partial S}{\partial T}\right)_{P,x} = \frac{C_{P,x}}{T} = A\tau(P)^{-\alpha} + B_1, \qquad (1)$$

where $\tau(P) = |T - T_c(P)|/T_c(P)$, $0 < \alpha \leq 0.2$ for $T > T_c$, and $0 \leq \alpha < 0.2$ for $T < T_c$. If one assumes $\alpha = 1/8$, then $At_c = 9$ J/mole-deg for $T < T_c$ and $AT_c = 4.3$ J/mole-deg for $T > T_c$. The results of the measurements of $C_{P,x}$ and $C_{V,x}$ have enabled us to clarify the behavior of other thermodynamical quantities near the critical line of this mixture. The major difficulty associated with measurements of C_V of a liquid is the marked increase of the pressure associated with heating in a closed volume ($(\partial P/\partial T)_V \sim 10$ atm/deg). Therefore, in order to carry out such measurements we prepared a high-pressure calorimeter, which is a cylindrical bomb made out of cold-worked steel having a volume of approximately 40 cm³ and with 0.1 cm thick walls, which can be sealed off by a globe valve. The intrinsic heat capacity of the calorimeter amounts to ~40% of the heat capacity of the sample. Tests have shown that the calorimeter retains the ability to be deformed elastically up to pressures of 300 to 350 atm. The increase of the volume associated with the increase of the pressure as a consequence of elastic expansion of the calorimeter does not exceed 5% of the thermal expansion of the liquid.

Measurements of the specific heat C_V of water in the interval from 315 to 350°K showed that the contribution to the specific heat arising from the deformation of the calorimeter is a monotonic function of the tem-

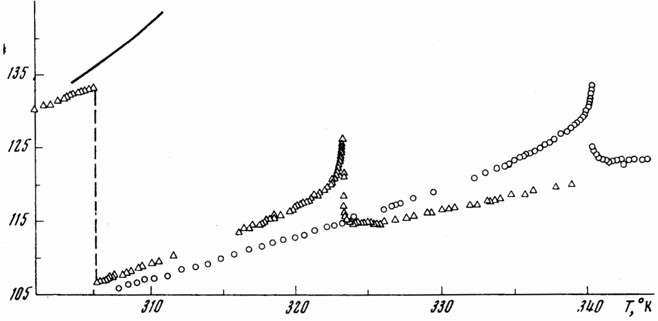

FIG. 1. Dependence of the specific heat at constant volume on the temperature near the critical point of demixing of a methanol—cyclohexane mixture. The triangles pertain to a mixture without any water impurities, the open circles pertain to a mixture containing 0.35% water by weight, and the solid line indicates the dependence of $C_{p,x}$ on T in the same mixture.

Table I. Experimental values of the specific heat $C_{V,x}$ of a methanol-cyclohexane mixture (29.2% by weight of methanol)

C, J/mole-deg	T_1, °K	T_2, °K	C, J/mole-deg	T_1, °K	T_2, °K
108,4	310,8666	312.3550	116.3	321.7885	321,9218
110,1	313,8569	315.3283	116.9	321.9229	322,0559
111,4	315,3376	316,7982	116.7	321,8116	321.9294
112,4	316.8049	318.2565	117.1	321,9306	322,0482
112.1	316.6278	316.9735	116.6	321.4271	321.7649
112,4	316,9724	317.3173	117.7	322.0974	322.4333
112.4	317.3160	317,6609	118.3	322.4301	322,7649
112.6	317.6593	318,0385	117.2	322.1420	322.3009
112.4	317.4443	317,7825	117.3	322.0498	322.1673
112,9	317,7809	318,2872	117.4	322.1684	322.2858
113,6	318,2615	319,7028	117.3	322.2870	322.4045
113,2	318.0360	318.3798	117.8	322.4054	322.5284
113,2	318.3777	318.7215	118.0	322.5290	322.6695
113,5	318,7195	319.0629	118.5	322.7984	322.6700
114.2	319,3994	319.7415	119.0	322.7990	322.9272
114.4	319,7382	320.0799	119.4	322.9275	323.0437
114.9	320.0762	320.4227	119.3	322.8734	322.9895
115.3	320.4194	320,7596	119.2	322.9384	323.0600
115.6	320,7561	321.0957	118.9	322.7423	322.8344
113.4	318.2856	318,7907	119.4	322.9296	322.9755
113.7	318.7885	319.2932	117.7	322,2535	322.5465
114.2	319.2918	319,7949	117.3	322,0186	322.2715
114.8	319.7937	320.2954	117.4	322.2570	322.5101
115,2	320.2941	320.7947	118,3	322,5091	322.7611
115,6	320.7831	321.0781	117.0	322.0116	322.2308
115.0	319.7073	321.1374	117.7	322.2308	322.4498
116.2	321.0920	321.4306	118.0	322.4498	322.6687
115.9	321,0779	321,3721	118.2	322.6717	322.7388
116.3	321,3723	321.6665	118.6	322.7417	322.8088
116.4	321,5166	321.7706	117.3	322.3078	322.4000
116.9	321.7685	322.0219	118.7	322.8065	322.8674
116.8	321,1402	322.5565	119.2	322,8671	322,9281
117,2	321.7630	322.0998	120.2	323.8460	323.1592
115.6	320,9887	321,0814	120.8	323.0762	323.1912
116.0	321.0809	321.1733	121.9	323.2379	323.2494
116.1	321.1726	321.2651	122.4	323.2560	323.2617
116.5	321,6225	321.7823	112.3	324.0144	324.2751
124.3	323.2613	323.2670	121.6	323.2036	323.2191
119.7	322.9949	323.0292	121.9	323.2196	323.2351
119.6	322.9885	323.0888	112.2	323,9196	324.2103
114.4	323.2775	323,3464	112.2	324.5002	324.9354
113.2	323.3820	323,5022	112.4	323.8893	324.1784
112.4	323.8870	324.0077	112.3	324.1778	324,6110
118.0	322.4856	322,5255	112.1	324.2502	324.5293
118,1	322.5680	322.6079	112.2	324.5271	324.8061
122.9	323.2573	323.2670	112.2	324.8042	325.0830
119.0	323.2768	323.2866	112.4	324.2775	324.5380
116.0	323.2868	323,2968	112.4	324.5401	324.8006
115,0	323.2970	323.3071	112.4	324.0066	324.1274
113,2	323.3071	323.3172	112.1	324.1252	324.2462
112.5	323.6001	323.8893	112.4	324.4459	324.7918
112.6	323.5203	323.7984	112.3	324.7873	325.1334
112.4	323,7943	324.0727	112.4	325.1278	325.4737
112.8	322.4809	323.5504	112,5	325.4675	325.8133
119.3	322,9051	322.9727	112.1	325.0754	325.4938
119.6	323.9720	323.0395	112.3	325.4912	325.9092
119.8	323.0386	323.1061	112.2	324.9346	325.2830
121,4	323.1629	323.2433	112.4	325.2820	325.8210
122,9	323.2426	323.2674	112.5	325.8196	326.3406
114.5	323,2910	323.3169	112.4	325,8992	326.4295
113,7	323.3307	323.3568	112,9	326.4269	327.2201
113.3	323.3519	323.3781	113.4	326,8970	328.3372
113.1	323.3754	323.4016	113,2	327.2166	328.0080
112,4	323,6284	323.9196	113.7	328.8296	329.6319
			113.7	328.3402	329.7772

Note. T_C = (323.275 ± 0.005) °K and $T_2 - T_1 = \Delta T$ is the calorimetric step of heating. We are compelled to present the values of the temperature to within 10^{-4} degree, characteristic only of the sensitivity of our apparatus, in order not to foist on the reader our own procedure for smoothing the experimental points, which is a matter of opinion. Thus, having assumed the formula (2), we must determine the averages of the points according to the formula $\tau_{av}^{-\alpha} = (\tau_1^{-\alpha} + \tau_2^{-\alpha})/2$, but for some other approximation this formula would be different. The actual accuracy of the temperature on the thermodynamic scale is approximately 10^{-2} degrees, and it is only with such accuracy that our data is to be compared with the data obtained by other authors.

Table II. Experimental values of the specific heat by weight of methanol, T_C = 319.302 ± 0.003°K)

C, J/mole-deg	T_1, °K	T_2, °K	C, J/mole-deg	T_1, °K	T_2, °K
134.2	305.1471	306.3063	140.2	319.6959	319.7869
135.2	306.4063	307.5445	140.0	319.7855	319.8766
136.1	307.5570	308.6879	152.2	318.9991	319.0772
137.0	308.7011	309.8246	153.0	319.0782	319.1190
137.2	309.3683	310.0199	153.7	319.1200	319.1606
137.9	309.8355	310.9519	154.2	319.1617	319.2023
138.9	310.9638	312.0729	155.9	319.2027	319.2429
140.0	312.0831	313.1839	157.8	319.2431	319.2829
139.9	312.2506	312.8955	143.7	319.3235	319.3668
140.5	312.9014	313.5457	142.4	319.3667	319.4103
141.0	313.1928	314.2860	156.5	319.2391	319.2502
141.1	313.5508	314.1940	157.7	319.2498	319.2664
141.9	314.1986	314.8395	159.1	319.2659	319.2824
141.1	314.8446	315.4829	160.9	319.2819	319.2982
143.4	315.4875	316.1237	144.4	319.3144	319.3324
144.3	316.1273	316.7603	158.5	319.2613	319.2750
145.4	316.7645	317.3932	159.7	319.2755	319.2850
145.0	317.3966	318.0206	160.5	319.2853	319.2966
146.2	317.0349	318.0883	139.3	320.1195	320.7737
149.5	318.0948	319.1267	139.1	320.2165	321.3164
148.4	318.0261	318.6440	139.3	321.3204	322.4184
150.9	318.6468	319.1538	139.2	320.7769	321.4322
140.1	319.4654	320.1160	139.1	321.4376	322.0936
139.5	319.6629	320.2114	139.2	322.0965	322.7524
149.9	318.6749	318.7760	139.3	322.4229	323.5198
150.6	318.7767	318.9108	139.4	323.5618	324.6398
151.6	318.9117	319.0451	139.4	322.7553	323.4109
153.2	319.0455	319.1779	139.9	324.6429	325.7278
156.3	319.2027	319.2604	140.6	325.7357	326.8180
142.6	319.3422	319.4319	140.8	326.8195	327.8990
141.5	319.4300	319.5203	141.3	327.8994	328.9761
140.6	319.5174	319.6082	141.7	328.9767	330.0505
140.6	319.6062	319.6975	142.2	330.0305	331.1207

perature. The measurement procedure is analogous to the one described in[9]. The random error in the determination of $C_{V,x}$ varied from 0.1–0.2% far away from the critical point to 0.5–1% in the immediate vicinity of the critical point (when the calorimetric measurements were made in steps of $\lesssim 0.01°$). The results of measurements of $C_{V,x}$ for a mixture containing 29.2% of methanol by weight (the critical composition is x_K = 29 ± 1% by weight) and for the same mixture with 0.35% by water weight are presented on Fig. 1 and in Table I (the results of measurements of $C_{P,x}$ are given in Table II).

The calorimeter was filled with the liquid mixture at $T \approx 306°K$ so that no vapor space was left; therefore upon lowering the temperature, when the pressure in the calorimeter becomes equal to the pressure of the saturated vapors of the mixture, a vapor phase appears (the discontinuity in the specific heat is ~27 J/mole-deg). Upon further reduction of the temperature, the pressure changes slightly—the measured value is close to $C_{P,x}$ of the liquid. One can attribute the ~1% discrepancy with the results obtained in[9] to differences in the extent to which the calorimeter was filled and, in part, to differences in the compositions of the samples (0.2% by weight). For $T > 306°K$ the measured value is close to $C_{V,x}$. The transition temperature, T_C = 323.27°K, determined according to the maximum of the specific heat, was found to be ~4° above the value of T_C at the saturated vapor pressure.[9] Because $dT_C/dP \approx 0.034$ deg/atm in this mixture,[10] for $T = T_C$ the value of $P \approx 120$ atm. The transition temperature in a three-component mixture (0.35% by weight of water) is shifted both due to the increase of the pressure and as a consequence of the addition of the third component (water) (see[9]). Here the value of $P \approx 250$ to 300 atm for $T = T_C'$ = 340.236°K. The change in the critical composition has been neglected, since estimates indicate that it lies within the limits of error of the determination of x_K.

The results of the measurements are presented on Fig. 2 in a semi-logarithmic scale (each plot is treated with respect to its "own" transition temperature). For clear representation the curves in the

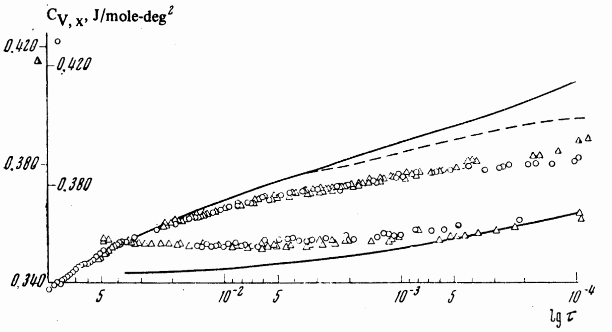

FIG. 2. Dependence of the specific heat on log τ in a mixture of methanol and cyclohexane (the continuous line represents the dependence of $C_{P,x}$ on log $\tau(P)$, the dashed line represents the dependence of $C_{P,x}$ on log $\tau(P)$ in a mixture containing an admixture of 0.35% by weight of water, the triangles represent the dependence of $C_{V,x}$ on log $\tau(V)$, the open circles represent the dependence of $C_{V,x}$ on log $\tau(V)$ in a mixture containing 0.35% by weight of water, the upper curves pertain to the heterogeneous region, and the lower curves pertain to the homogeneous region).

heterophase region are superposed for $\tau = 10^{-2}$. The analysis of the results of the measurements of $C_{V,x}$ in the heterophase region ($T < T_c$) indicates that, for $10^{-4} < \tau < 5 \times 10^{-3}$ one can approximate the dependence of $C_{V,x}$ on τ by the expression

$$C_{V,x}/T = a\tau(V)^{-\alpha} + b, \tag{2}$$

where $\tau(V) = |T - T_c(V)|/T_c(V)$, $a/A = 0.55 \pm 0.03$ (for the given value of α), and the values of the critical index α for $C_{V,x}$ and $C_{P,x}$ coincide within the limits of error of the experiment. A quantitative interpretation for $T > T_c$ is difficult because here the singular part of $C_{V,x}$ is too small (2 to 3% of b for $\tau = 10^{-3}$); however, qualitatively the picture is similar. In the present case the influence of the impurities of the third component (water) for $T < T_c$ is weaker (the corresponding value of τ is an order of magnitude smaller) than it is in the case of the measurements of $C_{P,x}$, where it leads to an appreciable renormalization of the critical index (the exponent of the power of τ becomes positive) already for $\tau \lesssim 10^{-3}$. This is associated with the fact that $a < A$ (see the estimates of the effect of impurities given in[9]).

Now let us analyze the results from the point of view of the isomorphism hypothesis[3] and attempt to understand the behavior of the other thermodynamical quantities near the critical line on this basis. According to[3] near the critical line of a binary mixture the quantities $(\partial V/\partial T)_{P,x}$ and $(\partial V/\partial P)_{T,x}$ are singular just like the specific heat $C_{P,x}$ (see formula (1)):

$$\left(\frac{\partial V}{\partial T}\right)_{P,x} = A\frac{dT_c}{dP}\tau(P)^{-\alpha} + B_2; \tag{3}$$

$$\left(\frac{\partial V}{\partial P}\right)_{T,x} = -A\left(\frac{dT_c}{dP}\right)^2\tau(P)^{-\alpha} + B_3, \tag{4}$$

where B_2 and B_3 are the regular parts of $(\partial V/\partial T)_{P,x}$ and $(\partial V/\partial P)_{T,x}$ (let us take $B_2 = 10^{-1}$ cm^3/mole-deg and $B_3 = -10^{-2}$ cm^3/mole-atm;[11] the accuracy of the determination of B_2 and B_3 amounts to ~10%). As to the quantities $C_{V,x}$ and $(\partial P/\partial T)_{V,x}$, then only in the region

$$\tau^{\alpha} \ll \xi = -A\frac{dT_c}{dP}\frac{dT_c}{dV}\Big/\left(1 + B_2\frac{dT_c}{dV}\right)$$

is the quantity $C_{V,x} \sim \text{const} - \tau(V)^{\alpha/(1-\alpha)}$ (the renormalization of the critical index leads to a peak of finite magnitude) and $(\partial P/\partial T)_{V,x} \approx dP/dT_c = \text{const}$.

However, in the region $T^{\alpha} \gg \xi$ the quantities $C_{V,x}$ and $(\partial P/\partial T)_{V,x}$ increase in proportion to $\tau(V)^{-\alpha}$. Assuming $B_2 dT_c/dV \approx (B_2/B_3)dT_K/dP \approx -0.3$, for $T < T_c$ we obtain the value $T^{\alpha} = \xi$ for $\tau \approx 10^{-10}$, that is, the entire region which has been experimentally investigated satisfies the condition $\tau^{\alpha} \gg \xi$ (analogously in helium $\ln\tau \gg \xi$).

Let us calculate the coefficients for the increasing parts of $C_{V,x}$ and $(\partial P/\partial T)_{V,x}$.

1. Substituting (2)--(4) into the thermodynamical equation

$$\frac{C_{V,x}}{T} = \frac{C_{P,x}}{T} + \left(\frac{\partial V}{\partial T}\right)^2_{P,x}\Big/\left(\frac{\partial V}{\partial P}\right)_{T,x} \tag{5}$$

and taking into consideration that for $\tau^{\alpha} \gg \xi$ we have $\tau(P) = (1 + B_2 dT_c/dV)\tau(V)$ (see[3]), we obtain the following result in the region where the singular parts of $(\partial V/\partial T)_{P,x}$, $C_{P,x}$, and $(\partial V/\partial P)_{T,x}$ are small:

$$a \approx A\left(1 + B_2\frac{dT_c}{dV}\right)^{-\alpha}\left\{1 + 2\frac{B_2}{B_3}\frac{dT_c}{dP} + \left(\frac{B_2}{B_3}\frac{dT_c}{dP}\right)^2\right\} \approx (0.6 \pm 0.06)A,$$

$$b \approx B_1 + (B_2^2/B_3).$$

For $T \approx 300$°K the regular part of $C_{V,x}$ amounts to $bT \approx 100$ J/mole-deg and, finally, the jump $C_{P,x} - C_{V,x}$ far away from the critical point amounts to ~30 J/mole-deg. The obtained values are in quite satisfactory agreement (within an accuracy of 10% which is primarily determined by errors in the calculation) with the experimentally determined values.

In the region close to the critical line, where the singular parts of $(\partial V/\partial T)_{P,x}$ and $(\partial V/\partial P)_{T,x}$ are large, the specific heat $C_{V,x}$ tends to the finite limit $C_{V,x} \approx B_1 T_c - (dP/dT_c)^2 B_3 T_c \approx 450$ J/mole-deg, as is evident from Eq. (5).

2. For $T^{\alpha} \gg \xi$ we have

$$\left(\frac{\partial P}{\partial T}\right)_{V,x} = -\left(\frac{\partial V}{\partial T}\right)_{P,x}\Big/\left(\frac{\partial V}{\partial P}\right)_{T,x} \approx -\frac{B_2}{B_3} + K\tau(V)^{-\alpha}, \tag{6}$$

where

$$K = -\frac{A}{B_3}\frac{dT_c}{dP}\left(1 + \frac{B_2}{B_3}\frac{dT_c}{dP}\right)\left(1 + B_3\frac{dT_c}{dV}\right)^{-\alpha}$$
$$\approx 0.7 \text{ atm/deg (for } T < T_c).$$

(In the homogeneous region ($T > T_c$) all of the coefficients containing A are half as large.) We note that in the quantities $(\partial V/\partial T)_{P,x}$ and $(\partial P/\partial T)_{V,x}$ the coefficients of the singular parts even for $T > T_c$ are smaller than the coefficients of the regular parts by only a factor on the order of 1.5. Therefore, just like the corresponding anomalies near the λ-transition in helium,[6] it should be comparatively easy to observe their increase in a direct experiment. The singularity of $(\partial V/\partial P)_{T,x}$ is more difficult to observe (for $T > T_c$ we have $-AB_3^{-1}(dT_c/dP)^2 \approx 10^{-2}$). In our opinion the agreement between the experimental results and the calculations is sufficiently convincing to confirm the conclusion[3] that the quantities $C_{P,x}$, $(\partial V/\partial T)_{P,x}$ and $(\partial V/\partial P)_{T,x}$ have a singular behavior along the entire critical line of a binary mixture. The presence of the small parameter $\rho_c R\, dT_c/dP$ which, just as in helium, determines the magnitude of the singular parts of $(\partial V/\partial T)_{P,x}$ and $(\partial V/\partial P)_{T,x}$ and the discontinuities in the behavior of $C_{V,x}$ and $(\partial P/\partial T)_{V,x}$, is related to the large value of the binding energy—that is, the "incompressibility" of the liquid.

In conclusion let us point out certain differences in the behavior of the thermodynamical quantities in the investigated mixture and in liquid helium. Since $R\rho_\lambda dT_\lambda/dP < 0$ in helium, then $(\partial V/\partial T)_P$ and $(\partial P/\partial T)_V$ decrease near the λ-transition, but the singular part of C_P and the increasing part of C_V practically coincide (in our case the coefficient for the increasing part of $C_{V,x}$ is half as large as for $C_{P,x}$). Therefore, in particular, the effect of the decrease in the thermodynamical speed of sound $u = \{(C_P/C_V)(\partial P/\partial\rho)_T\}^{1/2}$ which is well-known near the λ-transition,[5,8] will be very small in our case.

In fact, in the region where the singular parts of $C_{P,x}$, $(\partial V/\partial T)_{P,x}$, and $(\partial V/\partial P)_{T,x}$ are small, we have

$$\rho^2u^2 = -\frac{C_{P,x}}{C_{V,x}}\left(\frac{\partial P}{\partial V}\right)_{T,x} \approx \rho^2u_0^2 - \varkappa\tau^{-\alpha},$$

$$\rho^2u_0^2 = -\frac{B_1}{bB}, \quad \varkappa = \frac{A}{bB_3}\left\{1 - \frac{aB_1}{Ab} + \frac{B_1}{B_3}\left(\frac{dT_c}{dP}\right)^2\right\}$$

$$\approx 0.4 \text{ atm-mole/cm}^3 \quad (\text{for } T > T_c).$$

Then the decrease in the speed of sound relative to the regular part will amount to ~0.3% for $\tau = 10^{-2}$, and it will amount to ~0.5% for $\tau = 10^{-4}$. In this interval of temperatures, the regular part (u_0) increases by ~1%.

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