Determination of the critical exponent of the specific heat from data on the refractive index along the coexistence curve of freon-113

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Results are presented of an experimental investigation of the temperature dependence of the refractive index along the coexistence curve of freon-113 at temperatures $6 \times 10^{-4} < |\tau| < 0.4$. The data near the critical points are obtained by measuring the gravitational effect. The behavior of the refractive index along the coexistence curve is described by an equation whose right-hand side is a sum of an asymptotic term, an asymmetric term, and correction terms for deviation from the asymptotic and symmetric behavior. There is no correction for the deviation from the asymptotic behavior in the equation for the square of the refractive index in the same temperature interval. The results are compared with the theoretically predicted behavior of the dielectric constant in the investigated region. A value $\beta = 0.3425 \pm 0.0010$ was obtained for the order-parameter critical exponent and two values $\alpha = 0.11 \pm 0.01$ and 0.16 ± 0.01 , depending on the method used to reduce the experimental data, were obtained for the critical exponent of the specific heat.

PACS numbers: 78.20.Dj, 64.60.Fr, 65.20. + w

Progress in modern theory of critical phenomena calls for an increasingly more accurate determination of the numerical parameters that describe the properties of matter near the critical point and in a large surrounding region. Particular interest attaches to the determination of the critical exponents, which are the most important quantities of the theory of critical phenomena. The present paper is devoted to a determination of the critical exponent α of the specific heat from data on the refractive index of trifluortrichlorethane (freon-113) in a large vicinity of the critical point.

The direct determination of the exponent α from data on the refractive index n (or the dielectric constant $\varepsilon = n^2$) was made possible by the development of a theory of the dielectric constant and of the refractive index near the critical point. Thus, a singularity of the derivative of the dielectric constant with respect to temperature along the coexistence curve (CC) and the critical isochore was predicted theoretically in Ref. 2. It was found that the series for the dielectric constant along the CC should include terms proportional to $|\tau|^{1-\alpha}$ and $|\tau|^{2\beta}$, where $\tau = (T - T_c)/T_c$ is the relative temperature and β is the exponent that describes the behavior, along the CC, of the principal singular order parameter, whose role in an individual substance is assumed by the density. In Ref. 3 are considered the contributions made to the dielectric constant by the principal singular order parameter, the relative density, and by the less singular order parameter, the relative internal energy. The formulas of the expanded theory of scale transformations for these quantities yielded theoretically for these quantities the functional dependences of the dielectric constant ε in a large vicinity of the critical point for various limiting directions along which this point is approached. In particular, the temperature dependence of the dielectric constant ε along the coexistence curve of an individual liquid the temperature dependence is of the form

$$(\varepsilon_{\pm}-\varepsilon_c)/\varepsilon_c=\pm A_0|\tau|^{\beta_0}+A_1|\tau|^{1-\alpha}\pm A_2|\tau|^{\beta+\Delta}+A_3|\tau|+\ldots, (1)$$

where ε_c is the dielectric constant at the critical point, $\Delta \approx 0.5$ is Wegner's critical exponent, and A_i (i = 0, 1, 2, 3)are numerical coefficients. The upper and lower signs pertain respectively to the liquid and gas phases of the CC. The results of Ref. 3 differ from those of Ref. 2 in that the series for the dielectric constant does not contain a term $\sim |\tau|^{2\beta}$. In the opinion of the authors of Ref. 3 this is due to a different choice of the regular part of the dielectric constant.

We have investigated in the present study the temperature dependence of the refractive index in a large vicinity of the critical point along thess of freon-113 at a wavelength $\lambda = 5791$ Å. The results are listed in Table I. The data for temperatures far from the critical one (points 1-49) were obtained by a method⁴ in which prisms are placed at the bottom and top of the chamber. The error of the refractive index determined by the prism method does not exceed 0.01%. Near the critical point, a gravitational effect takes place in the substance, i. e., a substantial density drop along the height of the vessel. To allow for the drop in the determination of the refractive index of the coexisting phases at the meniscus itself, the prism method was supplemented by the Toepler shadow method. Points 50-61 are the results of integrating the refractive-index gradient with respect to height. The numerical integration was by the trapezoidal rule, using as the initial values the refractive-index absolute values obtained by the prism method. The error in this region is somewhat larger, but does not exceed 0.02%. The temperature of the investigated substance was maintained constant by a double-thermostating method⁵ accurate to 0.001 °C and was measured by a calibrating platinum resistance thermometer.

Taking into account the functional character of the theoretically predicted¹ dependence, we have reduced our experimental data by using the formula

$$(n_{\pm}^{2} - n_{c}^{2})/n_{c}^{2} = \pm N_{0} |\tau|^{\beta_{0}} + N_{i} |\tau|^{\beta_{1}} \pm N_{2} |\tau|^{\beta_{2}} + N_{3} |\tau|^{\beta_{3}}, \qquad (2)$$

where n_c is the refractive index at the critical point, and determined the powers β_i and the coefficients N_i directly

Number of point	T]	<i>n</i> +	n	Number of point	t	n+	n-
$\begin{array}{c} 1\\ 2\\ 3\\ 4\\ 5\\ 6\\ 7\\ 8\\ 9\\ 10\\ 11\\ 12\\ 13\\ 14\\ 15\\ 16\\ 17\\ 18\\ 19\\ 20\\ 21\\ 22\\ 23\\ 24\\ 25\\ 26\\ 27\\ 28\\ 29\\ 30\\ 31\\ \end{array}$	$\begin{array}{c} 0,39575\\ 0,27250\\ 0,27250\\ 0,24993\\ 2,26\cdot10^{-1}\\ 1,95\cdot10^{-1}\\ 1,95\cdot10^{-1}\\ 1,77\cdot10^{-1}\\ 1,52\cdot10^{-1}\\ 1,26\cdot10^{-1}\\ 1,23\cdot10^{-1}\\ 1,20\cdot10^{-1}\\ 1,20\cdot10^{-1}\\ 1,20\cdot10^{-1}\\ 9,72\cdot10^{-2}\\ 8,80\cdot10^{-2}\\ 8,80\cdot10^{-2}\\ 8,80\cdot10^{-2}\\ 8,80\cdot10^{-2}\\ 8,80\cdot10^{-2}\\ 6,98\cdot10^{-2}\\ 6,98\cdot10^{-2}\\ 6,98\cdot10^{-2}\\ 6,98\cdot10^{-2}\\ 4,31\cdot10^{-2}\\ 6,98\cdot10^{-2}\\ 4,31\cdot10^{-2}\\ 4,31\cdot10^{-2}\\ 2,89\cdot10^{-2}\\ 2,89\cdot10^{-2}\\ 2,89\cdot10^{-2}\\ 2,57\cdot10^{-2}\\ 2,02\cdot10^{-2}\\ 1,92\cdot10^{-2}\\ 1,92\cdot10^{-2}\\ \end{array}$	$\begin{array}{c} 1.3573\\ 1.3214\\ 1.3147\\ 1.3068\\ 1.2960\\ 1.2889\\ 1.2850\\ 1.2751\\ 1.2653\\ 1.2667\\ 1.2653\\ 1.2667\\ 1.2653\\ 1.2610\\ 1.2532\\ 1.2489\\ 1.2460\\ 1.2421\\ 1.2407\\ 1.2389\\ 1.238\\ 1.2225\\ 1.2263\\ 1.2263\\ 1.2263\\ 1.2263\\ 1.2263\\ 1.2263\\ 1.2263\\ 1.22070\\ 1.2057\\ 1.2059\\ 1.2059\\ 1.2059\\ 1.2023\\ 1.1962\\ 1.1947\\ \end{array}$	$\begin{array}{c} 1.0004\\ 1.0037\\ 1.0047\\ 1.0057\\ 1.0086\\ 1.0112\\ 1.0123\\ 1.0142\\ 1.0123\\ 1.0144\\ 1.0158\\ 1.0192\\ 1.0192\\ 1.0198\\ 1.0203\\ 1.0220\\ 1.0256\\ 1.0259\\ 1.0259\\ 1.0259\\ 1.0229\\ 1.0229\\ 1.0229\\ 1.0229\\ 1.0229\\ 1.0321\\ 1.0322\\ 1.0339\\ 1.0367\\ 1.0367\\ 1.0376\\ 1.0413\\ 1.0423\\ 1.0456\\ 1.0537\\ 1.0546\\ 1.0545\\ 1.0572\\ 1.0546\\ 1.0545\\ 1.0572\\ 1.0617\\ 1.0627\\ \end{array}$	$\begin{array}{c} 32\\ 33\\ 34\\ 35\\ 36\\ 37\\ 38\\ 39\\ 40\\ 41\\ 42\\ 43\\ 44\\ 45\\ 46\\ 47\\ 48\\ 49\\ 50\\ 51\\ 52\\ 53\\ 54\\ 55\\ 56\\ 57\\ 58\\ 59\\ 60\\ 61\\ \end{array}$	$\begin{array}{c} 1.79\cdot 10^{-2}\\ 1.70\cdot 10^{-2}\\ 1.70\cdot 10^{-2}\\ 1.58\cdot 10^{-2}\\ 1.58\cdot 10^{-2}\\ 1.49\cdot 10^{-2}\\ 1.49\cdot 10^{-2}\\ 1.28\cdot 10^{-2}\\ 1.18\cdot 10^{-2}\\ 1.18\cdot 10^{-2}\\ 1.18\cdot 10^{-2}\\ 1.18\cdot 10^{-2}\\ 1.06\cdot 10^{-2}\\ 9.90\cdot 10^{-3}\\ 8.85\cdot 10^{-3}\\ 7.54\cdot 10^{-3}\\ 8.85\cdot 10^{-3}\\ 6.59\cdot 10^{-3}\\ 6.59\cdot 10^{-3}\\ 6.59\cdot 10^{-3}\\ 4.30\cdot 10^{-3}\\ 3.80\cdot 10^{-3}\\ 3.23\cdot 10^{-3}\\ 2.29\cdot 10^{-3}\\ 2.29\cdot 10^{-3}\\ 2.29\cdot 10^{-3}\\ 1.53\cdot 10^{-3}\\ 1.53\cdot 10^{-3}\\ 1.53\cdot 10^{-3}\\ 1.59\cdot 10^{-4}\\ 5.90\cdot 10^{-4}\\ \end{array}$	$\begin{array}{c} 1.1931\\ 1.1916\\ 1.1913\\ 1.1900\\ 1.1890\\ 1.1887\\ 1.1886\\ 1.1852\\ 1.1835\\ 1.1835\\ 1.1811\\ 1.1800\\ 1.1759\\ 1.1747\\ 1.1728\\ 1.1716\\ 1.1696\\ 1.1678\\ 1.1661\\ 1.1663\\ 1.1663\\ 1.1663\\ 1.1558\\ 1.1558\\ 1.1558\\ 1.1558\\ 1.1558\\ 1.1558\\ 1.1558\\ 1.1514\\ 1.1458\\ 1.1514\\ 1.1458\\ 1.1458\\ 1.1558\\$	$\begin{array}{c} 1.0640\\ 1.0652\\ 1.0652\\ 1.0652\\ 1.0652\\ 1.0675\\ 1.0675\\ 1.0691\\ 1.0702\\ 1.0715\\ 1.0717\\ 1.0737\\ 1.0737\\ 1.0737\\ 1.0748\\ 1.0765\\ 1.0792\\ 1.0809\\ 1.0820\\ 1.0839\\ 1.0855\\ 1.0872\\ 1.0887\\ 1.0905\\ 1.0927\\ 1.0944\\ 1.0956\\ 1.0982\\ 1.1004\\ 1.1025\\ 1.0057\\ 1.0125\\ 1.0057\\ 1.0055\\$
	1	1	1	1	1	1	1

TABLE I. Experimental data on the temperature dependence of the refractive index along the coexistence curve of freon-113.

from the experimental data. The zeroth term of (2) describes the asymptotic behavior of the CC branches near the critical point, the first term describes this asymmetry, and the last two terms are corrections for the first two.

Since the refractive indices of the coexisting liquid and gas were determined experimentally at the same temperature, we used in the analysis, for the half-difference and halfsum ("diameter") of the n^2 -CC branches, the expressions

$$(n_{+}^{2}-n_{-}^{2})/2n_{c}^{2}=N_{0}|\tau|^{\beta_{0}}+N_{2}|\tau|^{\beta_{2}}, \qquad (3)$$

$$(n_{+}^{2}+n_{-}^{2}-2n_{c}^{2})/2n_{c}^{2}=N_{1}|\tau|^{3}+N_{3}|\tau|^{\beta}, \qquad (4)$$

in which the symmetric and asymmetric terms are located in different equations. This allows us to determine the parameters of the symmetric and asymmetric terms independently of one another, i.e., their correlation and the correlation of their confidence intervals are zero.

Comparison of the experimentally determined values of the exponents β_i with the powers of Eq. (1) leads to conclusions concerning the numerical values of the exponents β , α , and Δ .

All the equations employed contain the refractive index n_c of the material at the critical point and the critical temperature T_c , quantities whose correct determination govern the sought exponents and coefficients. The value $T_c = 486.958 \pm 0.003$ was obtained by us in Ref. 6. The value of n_c was obtained from an analysis of the behavior of the "*n*-CC diameter", which has an inflection near the critical point. Extrapolation of the experimental data for the *n*-CC diameter $(n^+ + n^-)/2$ in the immediate vicinity of the critical point prior to intersection with the vertical line $T_c = 486.958$ yields a value 1.1255 ± 0.0005 , which agrees

with an analysis⁶ of the critical density at a specific refraction r = 0.1436 cm³/g. A more accurate value $n_c = 1.1255 \pm 0.0001$ was obtained from an analysis of the behavior of the expressions for the refractive index

$$(n_{+}+n_{-})/2n_{c}-2$$

and the square of the refractive index

$$(n_{+}^{2}+n_{-}^{2})/2n_{c}^{2}-1$$

in a doubly logarithmic sale upon variation of the experimentally obtained value of n_c within the indicated error limits. Namely, at $n_c = 1.1255$ all the points closest to the critical temperature and having the maximum error were located, within the limits of this error, on the straight line made up of points 5 to 40. It can be seen from Fig. 1 that the same regularity is observed both for the diameter of the refractive index and for the diameter of the squared refractive index.

The calculations for the determination of N_i and β_i were performed with a computer by the FUMILI program, which effects linearization when finding the minimum of the functional.⁷ The use of this program has shown that if the experimental data are directly approximated by the eightparameter formula (2) the exponents and the coefficients obtained from the calculation are not single-valued and depend strongly on the initial values of the parameters that serve as the starting point for the approximation. The reason is that the functional of this problem has several minima. To obtain physically meaningful parameter values we have therefore used the following procedure.

The zeroth term $N_0 |\tau|^{\beta_0}$ in the right-hand side of (2) is the leading one and as the temperature comes close to criti-



FIG. 1. Temperature dependences of the diameters 1) n^2 -CC $(x_1 = (n_+^2 + n_-^2)/2n_c^2 - 1)$ and 2) n-CC $(x_2 = (n_+ + n_-)/2n_c - 1)$ in a doubly-logarithmic scale.

cal its relative contribution increases, i.e., it exceeds substantially the remaining terms. A correct determination of the succeeding terms is possible if the leading term is correctly calculated. To determine β_0 and N_0 numerically we obtained their values as the limits in the temperature dependence of the effective values β^{eff} of the exponents and N^{eff} of the coefficients with the experimental values of $(n_{\pm}^2 - n_c^2)/n_c^2$ and $(n_{+}^2 - n_{-}^2)/2n_c^2$ approximated at different temperatures by the single-term expressions

$$(n_{+}^{2} - n_{c}^{2})/n_{c}^{2} = \pm N_{+}^{\text{eff}} |\tau|^{\beta_{\pm}^{\text{eff}}}, \qquad (5)$$

$$(n_{+}^{2}-n_{-}^{2})/2n_{c}^{2}=N^{\text{eff}}|\tau|^{\beta^{\text{eff}}}$$
 (6)

To obtain the temperature dependences of the effective values of the exponent and of the coefficient, we narrowed down the investigated temperature interval by successively excluding from each five low-temperature points. Figure 2 shows the dependence of the effective values of the exponents and coefficients of Eqs. (5) and (6) on the logarithm of the average temperature of each of the 12 obtained temperature intervals. It can be seen from Fig. 2a, the effective values of the exponents come closer together, the values of β_{\pm}^{eff} and β_{-}^{eff} are symmetrical about the values of β_{-}^{eff} , the latter remaining practically unchanged with changed of the temperature interval. A perfectly analogous situation is observed also for the coefficients N_{\pm}^{eff} and N^{eff} . The limiting values of β_0 and N_0 were chosen to be the averages of all the effective values β^{eff} and N^{eff} for the half-difference (6): $\beta_0 = 0.3425 \pm 0.0010, N_0 = 0.456 \pm 0.001$. At these values of β_0 and N_0 , the single term formula

$$(n_{\pm}^{2}-n_{c}^{2})/n_{c}^{2}=\pm N_{0}|\tau|^{\beta}$$

describes well the experimental results for the liquid and gas phases in the temperature region $6 \times 10^{-4} < |\tau| < 0.01$, which is in fact the asymptotic one.



FIG. 2. Dependences of the effective exponents β^{eff} and β^{eff}_{\pm} (a) and of the coefficients N^{eff} and N^{eff}_{\pm} (b) on the logarithm of the average temperature of the integral in the approximation of the experimental data by Eqs. (5) and (6).

In Eq. (4), which describes the behavior of the CC diameter for the square of the refractive index, the term $N_1 |\tau|^{\beta_1}$ plays the leading role. We have determined it by excluding from the data reduction the experimental points of the temperature interval in which the zeroth term is the principal "working" one, and the contribution of the remaining terms is comparable with the experimental error. In the temperature range $0.01 < |\tau| < 0.4$ the data were reduced by using the equations

$$(n_{\pm}^{2} - n_{c}^{2})/n_{c}^{2} \mp N_{0} |\tau|^{\beta_{0}} = N_{1,\pm} |\tau|^{\beta_{1,\pm}},$$
(7)

$$(n_{+}^{2}+n_{-}^{2})/2n_{c}^{2}-1=N_{i}^{\text{eff}}|\tau|^{\beta_{i}} e^{\text{eff}}.$$
(8)

As a result of the analysis of the computer-calculation data, the following limiting values were chosen: $\beta_1 = 0.84 \pm 0.01$, $N_1 = 0.252 \pm 0.005$. The fact that the exponent differs from unity is evidence that the n^2 -CC diameter is singular.

To estimate the quality of the approximation of the experimental data by the terms $N_0 |\tau|^{\beta_0}$ and $N_1 |\tau|^{\beta_1}$, we investigated the remainders Δ_0 and Δ_1 :

$$\Delta_0 = (n_+^2 - n_-^2) / 2n_c^2 - N_0 |\tau|^{\beta_0}, \qquad (9)$$

$$\Delta_{1} = (n_{+}^{2} + n_{-}^{2})/2n_{c}^{2} - 1 - N_{1} |\tau|^{\beta_{1}}, \qquad (10)$$

whose temperature dependences are shown in Figs. 3a and 3b.

It can be seen from Fig. 3a that the difference Δ_0 is equal to zero within the limits of the experimental error σ_{Δ_0} $= 6 \times 10^{-4}$ in the entire investigated temperature interval. This is evidence that the contribution of the term $N_2 |\tau|^{\beta_2}$ in Eq. (2) does not exceed the experimental-data errors, i.e., it is insignificant. For the n^2 -CC diameter the remainders Δ_1 (Eq. (10) shown in Fig. 3b demonstrate that its description by the single term 0.252 $|\tau|^{0.84}$ in the entire temperature interval is inadequate. It can be seen from Fig. 3b that on moving away from the critical temperature the positive deviations due to the contribution of the term $N_3 |\tau|^{\beta_3}$ increase systematically. In the temperature interval $0.20 < |\tau| < 0.40$ the term $N_3 |\tau|^{\beta_3}$ is significant. Since this term tends to appear some-



FIG. 3. Temperature dependences of the remainders: a) Δ_0 [Eq. (9)], b) Δ_1 [Eq. (1)], and c) $\Delta_2 = \Delta_1 - N_3 |\tau|^{\beta_3}$ at $\beta_0 = 0.3425$, $N_0 = 0.456$, $\beta_1 = 0.84$, $N_1 = 0.252$, $\beta_3 = 4.5$, $N_3 = 0.69$.

what earlier, we have determined it in the temperature interval $0.15 < |\tau| < 0.40$. This interval contains only eight experimental points, so that we were unable to determine the fourth term with high accuracy, and we obtained $\beta_3 = 4.5 \pm 1.0$ and $N_3 = 0.69 \pm 0.10$. The remainders $\Delta_2 = \Delta_1 - N_3 |\tau|^{\beta_3}$ shown in Fig. 3c indicate that the n^2 -CC diameter is well described by Eq. (4), i.e., that this description is adequate.

Thus, we obtained for the description of the temperature dependence of the squared refractive index of the liquid and gaseous freon-113, in the entire investigated coexistence region $6 \times 10^{-4} \le |\tau| \le 0.40$ the formula

$$\frac{(n_{\pm}^2 - n_{c}^2)/n_{c}^2 = \pm (0.456 \pm 0.001) |\tau|^{0.3125 \pm 0.0010}}{+ (0.252 \pm 0.005) |\tau|^{0.84 \pm 0.01} + (0.69 \pm 0.10) |\tau|^{4.5 \pm 1.0}}.$$
 (11)

Figure 4 shows the temperature dependences of the functions y_i , which represent in succession the contributions made to the liquid and gaseous CC branches of Eq. 11 by the first term $y_1 = \pm 0.456 |\tau|^{0.3425}$ (curve 1–1), of the second term $y_2 = 0.252 |\tau|^{0.84}$ (curve 2), of the third term $y_3 = 0.69 |\tau|^{4.5}$ (curve 3), and the total contribution of all three terms:

$$y_4 = \pm 0.456 |\tau|^{0.3425} + 0.252 |\tau|^{0.84} + 0.69 |\tau|^{4.5}$$

(curve 4-4). The same figure shows the experimental n_{\pm}^2 points that lie, within the error limits, on the curve 4-4. This demonstrates the good description, by Eq. (11), of the experimental values of the squared refractive index for the liquid and gaseous branches of the CC.

A comparison of Eqs. (11) and (1) yields the following values of the critical exponents; $\beta = \beta_0 = 0.3425 \pm 0.0010$ and $\alpha = 1 - \beta_1 = 0.16 \pm 0.01$. The obtained value of the exponent agrees within the error limit with the exponent β_0 calculated in the analysis of the density along the CC of freon-113 (Ref. 6), and with most experimental and theoretical results.¹ Our value $\alpha = 0.16 \pm 0.01$ does not agree with the results of the most accurate direct experimental specificheat investigations, which yielded $\alpha = 0.11 \pm 0.01$ (Ref. 8).



FIG. 4. Contributions of the asymptotic term (curve 1–1), of the asymmetric term (curve 2), and of its correction (curve 3) to n^2 -CC (curve 4–4) of freon-113.

The deviation of the exponent β_0 from unity contradicts the theoretical conclusion that the correction to the principal asymmetric term in the right-hand side of (1) is linear.

To compare our results with the theoretical conclusions of Ref. 2 we have also analyzed the temperature dependence of the refractive index directly on the CC of freon-113. The data on the refractive index were approximated by the formula

$$(n_{\pm} - n_{c})/n_{c} = \pm N_{0}' |\tau|^{\beta_{0}'} + N_{1}' |\tau|^{\beta_{1}'} \pm N_{2}' |\tau|^{\beta_{2}'} + N_{3} |\tau|^{\beta_{3}'}, \quad (12)$$

which was written down in analogy with Eq. (2). The exponents β'_i and the coefficients N'_i were calculated in the same manner as the exponents β_i and the coefficients N_i . The results were $\beta'_0 = 0.341 \pm 0.001$; $\beta'_1 = 0.89 \pm 0.01$; $\beta'_2 = 2.25 \pm 0.50$; $\beta'_3 = 4.4 \pm 1.0$; $N'_0 = 0.225 \pm 0.001$; $N'_1 = 0.101 \pm 0.001$; $N'_2 = -0.055 \pm 0.010$; $N'_3 = 0.288 \pm 0.070$. Just as in the case of Eq. (2), the parameters of the second and fourth terms of (12) have low accuracy because of the small number of experimental points in the region where the contribution of these terms exceeds the experimental error. In (12), however, in contrast to (2), the term $N'_2 |\tau|^{\beta'_2}$ is not equal to zero and its contribution to the right-hand side of (12) reaches 3% at $|\tau| = 0.4$.

Comparing the results of the experimental-data reduction in terms of $n^2 = \varepsilon$ and *n*, we have found that the exponents β_0 and β'_0 are equal to each other and also, within the error limits, to the exponent determined from the behavior of the density.⁶ This confirms the feasibility of determining the critical exponent β by reducing direct experimental data on the refractive index *n* or on its square n^2 . The sensitivity of the method of determining β_0 from the data on n^2 is higher than for the density data. At the same time, the exponents β_1 and β'_1 of the leading asymmetric term are not equal to each other in the two considered cases. This is directly seen from Fig. 1, where the diameters for *n* and n^2 have in doubly logarithmic scale different slopes, with the exponent β obtained by reducing the data for *n* agreeing with the analogous exponent for the density.⁶

The difference between β_1 and β'_1 can be easily explained by starting from Eq. (2). The refractive index can be obtained in this cause by using an expansion in a Taylor series:

$$n_{\pm}(\tau) = (n_{\pm}^{2}(\tau))^{\frac{1}{2}}.$$

Confining ourselves to a quadratic approximation, we have

$$n_{\pm} = n_{c} \left[1 \pm \frac{1}{2} N_{0} |\tau|^{\beta_{0}} + \frac{1}{2} N_{1} |\tau|^{\beta_{1}} \pm \frac{1}{2} N_{2} |\tau|^{\beta_{2}} \right]$$

$$+ \frac{1}{2}N_{3} |\tau|^{\beta_{3}} - \frac{1}{8} (N_{0}^{2} |\tau|^{2\beta_{0}} + N_{1}^{2} |\tau|^{2\beta_{1}} \pm 2N_{1}N_{0} |\tau|^{\beta_{0} + \beta_{1}})].$$
(13)

This relation $n = n(\tau)$ differs from (12) that its right-hand side contains additional symmetric and asymmetric terms. Let us separate the asymmetric terms of (13):

$$(n_{+}+n_{-})/2n_{c}-1={}^{i}/{}_{2}N_{i}|\tau|^{\beta_{1}}+{}^{i}/{}_{2}N_{3}|\tau|^{\beta_{3}}-{}^{i}/{}_{8}N_{0}{}^{2}|\tau|^{2\beta_{0}}+\dots$$
(14)

The last term in the right-hand side of (14) is of the same order as the first, and their exponents β_1 and $2\beta_0$ are close to each other. To separate the term $1/2N_1|\tau|^{\beta_1}$ we therefore substracted from both halves of (14) the term $-1/8N_0^2|\tau|^{2\beta_0}$ whose parameters were determined by us above. The difference obtained was approximated by the single-term power function $N_1''|\tau|^{\beta_1''}$ by the method described above, obtaining as a result for the sought parameters the values $\beta_1'' = 0.835 \pm 0.010$ and $N_1'' = 0.125 \pm 0.005$. The exponent β_1'' obtained by reducing the refractive-index data in accord with (14) coincided thus, within the error limits,

with the value of β_1 obtained by reducing the square of the refractive index with the aid of (2).

We have carried out a similar calculation by deriving from (12) for n^2 an expression that agrees qualitatively with the results of Ref. 2, since it contains the term $|\tau|^{2\beta}$ in the right-hand side. The exponent β''_{1} obtained by this approach was found to equal 0.90 ± 0.01 which agreed with the value of β'_{1} obtained by reducing the refractive index in accord with Eq. (12).

Consequently, the difference between the exponents β_1 and β'_1 obtained by reducing the experimental data with the aid of Eqs. (2) and (12) is legitimate and can be easily explained.

Taking into account the theoretical predictions of Ref. 2 we obtain the critical exponent $\alpha_1 = 1 - \beta_1''' = 1 - \beta_1'' = 0.11 \pm 0.01$, which agrees, as indicated above, with most experimental and theoretical results for the specific-heat exponent.^{1,8}

Thus, two methods of reducing the same experimental data in terms of the refractive index and of its square give two different values of the specific-heat exponent, $\alpha = 0.16 \pm 0.01$ and $\alpha = 0.11 \pm 0.01$. Both exponents were obtained from experiments on light refraction on the basis of the theoretical premises of Refs. 2 and 3. Our investigation allows us to conclude that the theoretical model, which was

proposed in Ref. 2 and whose use to describe the experimental data on the refractive index of freon-113 yielded an exponent $\alpha = 0.11 \pm 0.01$, is preferable to the model proposed in Ref. 3, whose use yielded for the exponent α a value different from that obtained from specific-heat experiments. We hope that our results will stimulate further theoretical research into the refractive index and the dielectric constant near the critical point.

The authors thank A. V. Chalyĭ for constant interest in the work and for a discussion of the results.

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Translated by J. G. Adashko