

## Universality of Tricritical $^3\text{He}$ - $^4\text{He}$ Mixtures under Pressure

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The phase diagram of liquid  $^3\text{He}$ - $^4\text{He}$  mixtures in the vicinity of the tricritical point has been determined at saturated vapor pressure and at 22.6 bars, close to solidification. Neither the critical exponent  $\beta$  nor the slopes of the phase-separation curve change with pressure indicating universal behavior of the tricritical  $^3\text{He}$ - $^4\text{He}$  system.

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A fundamental aspect for the general treatment of phase transitions is given by the concept of universality.<sup>1</sup> According to it the character of a particular phase transition only depends on a few relevant quantities which determine the universality class of the system, such as the dimensionality or the degrees of freedom of the order parameter. All the other "irrelevant" variables may influence details like the absolute value of the critical temperature but leave essential quantities like the critical exponents unaffected.

For ordinary critical points these ideas have been checked, and verified, in a large number of systems. This applies not only to the exponents in the power laws describing the critical behavior, but also to the prefactors, called amplitudes, from which universal amplitude ratios can be constructed. Conversely, for higher-order critical points, information is very sparse and hence the situation is much less clear. Fisher and Sarbach have theoretically investigated a *tricritical* point of the so-called spherical model (where the number of components of the order parameter is infinite)<sup>2,3</sup> and have obtained the classical exponents characteristic of tricritical systems<sup>4</sup>; the dimensionless amplitude ratios, however, according to this model are *not* necessarily universal in magnitude.

In this work we report the first experimental results on the universality of tricritical behavior. For our measurements we have used  $^3\text{He}$ - $^4\text{He}$  mixtures which at saturated vapor pressure display a well-studied tricritical point (TCP) at a temperature  $T_t = 0.867$  K and a  $^3\text{He}$  concentration  $x_t = 0.675$ .<sup>5-9</sup> We have investigated the phase diagram of this system with the pressure  $p$  as an additional variable in order to study the effect of this presumably irrelevant parameter on the critical exponent  $\beta$  and also on the amplitudes given by the shape of the phase separation curve as defined below. Our main result is that  $\beta$  is indeed independent of  $p$  right up to the solidification pressure  $p_s$  of the mixture, indicating that the

nature of the TCP does not change. Moreover, we find that the slopes of the phase separation curves at saturated vapor pressure (SVP) and at elevated pressure closely coincide; using experimental evidence that the slope of the  $\lambda$  line at the TCP does not vary significantly with  $p$  either,<sup>10</sup> we obtain a pressure-independent amplitude ratio for the concentration, which signifies that, at least in  $^3\text{He}$ - $^4\text{He}$  mixtures, the suggested nonuniversality near the TCP does not occur.

The phase diagram of  $^3\text{He}$ - $^4\text{He}$  near  $T_t$ , characterized by a two-phase region terminating in an angular top, has been the first hint for the unique tricritical properties already in the very early experiments by Graf, Lee, and Reppy.<sup>6</sup> This shape, now well established at SVP, implies a critical exponent  $\beta = 1.0$ , following from the relation for the order parameter  $x_+ - x_- \propto t^\beta$ . (Here  $x_+$  and  $x_-$  are the  $^3\text{He}$  concentrations of the  $^3\text{He}$ - and the  $^4\text{He}$ -rich coexisting liquid phases and  $t = 1 - T/T_t$  is the reduced temperature.)

We have chosen the same quantity,  $x_+ - x_-$ , for testing tricritical properties under pressure because it is this difference in concentration which is experimentally accessible with high accuracy even for temperatures relatively close to  $T_t$ . Since the asymptotic tricritical region, as observed at SVP, is limited to  $T_t - t \lesssim 10$  mK,<sup>5,8</sup> one has to carry out measurements to within  $T_t - T \lesssim 1$  mK to cover at least one decade in reduced temperature—quite a small interval compared with the range available for other critical points. Even that very modest requirement, however, is usually hard to meet because measurements in  $^3\text{He}$ - $^4\text{He}$  mixtures close to the TCP in general suffer seriously from gravitationally induced concentration gradients.<sup>11,12</sup>

This latter problem can be circumvented by determining  $x_+ - x_-$  in the immediate neighborhood of the interface of the stratified mixture. In our measurement we have used a technique<sup>8</sup> where a laser beam enters an optical sample cell horizontally at the height of the interface. Because of

the discontinuity of the refractive index  $n$  at the interface, a small portion of the light is diffracted towards the medium with the higher refractive index, i.e., the lower phase, at an angle given by  $\gamma = \arccos(n_+/n_-)$  (where again the plus and minus sign refer to the  $^3\text{He}$ - and the  $^4\text{He}$ -rich phase, respectively). Since only light propagating within about a light wavelength from the interface<sup>13</sup> takes part in the diffraction process, the measurement samples  $\Delta n = n_- - n_+$  at distances less than  $1 \mu\text{m}$  away from the interface. Corrections arising from concentration gradients due to gravity are therefore negligible here. The quantity of interest,  $x_+ - x_-$ , is related to  $\Delta n$  via the molar volume  $V_m$  and the polarizability  $\alpha$  of helium<sup>9,14</sup>:

$$x_+ - x_- = \frac{n(V_m)^2}{2\pi\alpha(\partial V_m/\partial x)_{p,T}} \Delta n. \quad (1)$$

Except for  $\Delta n$  the factors on the right-hand side of Eq. (1) vary only slowly with temperature near  $T_t$ ; therefore, the critical exponent  $\beta$  already follows directly from the temperature dependence of the refractive index difference.

Values for  $\Delta n$  are shown in Fig. 1 for two representative runs at saturated vapor pressure and at 22.6 bars, 0.1 bar below solidification.<sup>15</sup> The tricritical temperature was evaluated from a fit

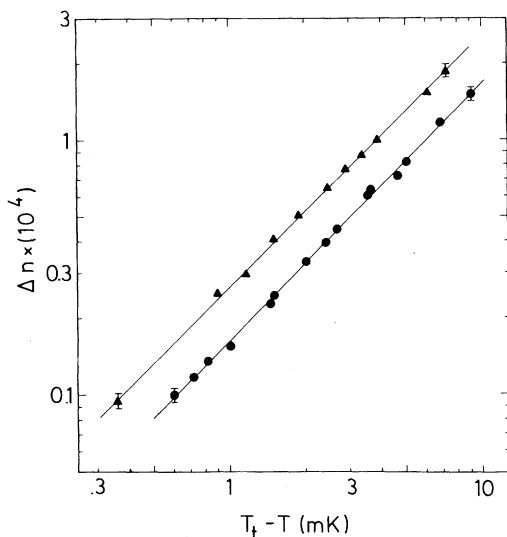


FIG. 1. Refractive index difference  $\Delta n$  between the  $^3\text{He}$ - and the  $^4\text{He}$ -rich phase along the phase separation curve. Solid triangles, at saturated vapor pressure; solid circles, at 22.6 bars. The lines represent best fits with  $\beta = 0.99$  and  $1.02$ , respectively. Although the accuracy in the absolute value of  $T_t$ ,  $0.867 \text{ K}$  at SVP and  $0.745 \text{ K}$  at 22.6 bars, is limited to  $\pm 1 \text{ mK}$  given by the temperature calibration, the fits allowed determination of  $T_t - T$  to within  $0.05 \text{ mK}$ .

of the data with  $\Delta n = A(1 - T/T_t)^\beta$ , where  $A$ ,  $T_t$ , and  $\beta$  are fit parameters, which yielded  $T_t = 0.867 \text{ K}$  at SVP and  $0.745 \text{ K}$  at 22.6 bars. The critical exponent  $\beta$  derived from these data is

$$\beta = \begin{cases} 0.99 \pm 0.02 & \text{at SVP,} \\ 1.02 \pm 0.03 & \text{at 22.6 bars;} \end{cases} \quad (2)$$

similar values were also found at intermediate pressures. All the data are consistent with an exponent  $\beta = 1$ . (Logarithmic factors, expected as a correction to the pure critical power laws near the TCP,<sup>4,16</sup> are beyond the precision of this experiment, although here the TCP has been approached more closely than in any other experiment so far.)

A value of  $\beta$  independent of pressure is in accord with universality. Amplitude ratios, however, as shown by Fisher and Sarbach,<sup>3</sup> might well vary with pressure for tricritical systems. In order to gain information about this possibly nonuniversal behavior in  $^3\text{He}$ - $^4\text{He}$  mixtures, a measurement of  $x_+ - x_-$  alone is not sufficient. The amplitude ratio given by  $\overline{OP}/\overline{PQ}$  as sketched in Fig. 2 requires  $x_+$  and  $x_-$  to be known separately.

Using a Mach-Zehnder interferometer we have therefore in addition measured the refractive index of the superfluid and, independently, of the normal phase with respect to the tricritical value

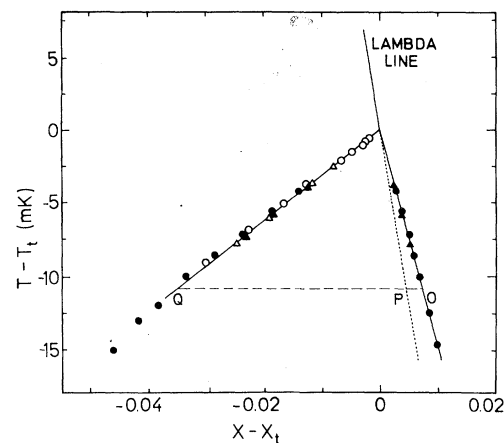


FIG. 2. Phase diagram of  $^3\text{He}$ - $^4\text{He}$  mixtures near the tricritical point. The full triangles and circles refer to interferometric measurements at SVP and 22.6 bars, respectively. The solid lines represent results at SVP from Refs. 5, 8, and 9. The extrapolation of the  $\lambda$  line (dotted) is used to define the amplitudes  $\overline{OP}$  and  $\overline{PQ}$ . Data from Fig. 1 (open symbols) are included for comparison, with the  $^3\text{He}$ -rich branch serving as reference concentration.

$n_i$ . Because of the finite width of the test light beam, the signal in this case represents an average over a sample height of 1 mm, instead of 1  $\mu\text{m}$ , leading to a much larger gravity effect. Hence the TCP could not be approached as closely as before without applying significant (and somewhat uncertain) corrections. Fortunately, for the present purposes such data very close to  $T_t$  are not really essential: With  $\beta = 1$  being established only one point on each branch is sufficient, in principle at least, to determine the slopes of the phase diagram.

Our results for  $x_{\pm} - x_t$ , obtained from the interferometric measurement and already corrected for the gravity effect, are plotted in Fig. 2. (The absolute values of  $x_t$  at SVP and 22.6 bars almost coincide.<sup>10</sup>) The solid lines represent earlier data at SVP,<sup>5,8,9</sup> with a slope  $(\partial T/\partial x)_p^+ = -1.47$  K/mole for the <sup>3</sup>He-rich branch and  $(\partial T/\partial x)_p^- = 0.31$  for the <sup>4</sup>He-rich branch. The slopes of the phase diagram at 22.6 bars differ from these values by less than 5%. For defining the amplitudes  $\overline{OP}$  and  $\overline{PQ}$  one, in addition, needs the slope of the  $\lambda$  line, which at SVP is  $(\partial T_\lambda/\partial x)_p = -2.47$  K/mole.<sup>5</sup> Unfortunately, at elevated pressures an accurate value for  $(\partial T_\lambda/\partial x)_p$  near the tricritical point is not available, but an extrapolation of data taken from Beal, Hatton, and Harrison<sup>17</sup> as well as the work of Cueto *et al.*<sup>10</sup> suggest that  $(\partial T_\lambda/\partial x)_p$  also is nearly independent of pressure. The amplitude ratio thus appears to remain constant at a value of  $\overline{OP}/\overline{PQ} = 0.075 \pm 0.02$ <sup>18</sup>; evidence for nonuniversal behavior is not observed.

Clearly, from the results presented here one cannot strictly exclude that some other quantity, like the concentration susceptibility  $\chi$ , might indeed exhibit nonuniversal behavior. Yet for the particular case of  $\chi$  being measured along the phase separation curve the amplitude ratio is predicted to be the same as for the concentration  $x$ ,<sup>3</sup> and hence again universality should apply.

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<sup>1</sup>See, for instance, L. P. Kadanoff, in *Critical Phenomena, Proceedings of the International School of Physics "Enrico Fermi" Course LI*, edited by M. S. Green (Academic, New York, 1972), p. 100; R. B. Griffiths, *Phys. Rev. Lett.* **24**, 1479 (1970).

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<sup>9</sup>H. A. Kierstead, *J. Low Temp. Phys.* **35**, 25 (1979).

<sup>10</sup>J. del Cueto, R. L. Johnson, T. Rohde, F. H. Wirth, and E. H. Graf, to be published. This work also contains information about the phase separation curves at elevated pressures.

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<sup>12</sup>P. C. Hohenberg and M. Barmatz, *Phys. Rev. A* **6**, 289 (1972).

<sup>13</sup>The finite width of the interface itself is expected to be of the order of the correlation length for concentration fluctuations in the bulk, which was always smaller than the wavelength of the laser light, 632.8 nm. See Ref. 8.

<sup>14</sup>I. V. Bogoyavlenskii and S. I. Yurchenko, *Fiz. Nizk. Temp.* **2**, 1379 (1976) [*Sov. J. Low Temp. Phys.* **2**, 672 (1976)].

<sup>15</sup>Measurements very close to the solidification pressure  $p_s$  were motivated by our observation that for the locus of the tricritical point  $(dp/dT)_t$  appears to diverge for  $p \rightarrow p_s$ . Therefore one might have speculated for some peculiarity, if not a change, in the tricritical properties to occur near  $p_s$ .

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<sup>18</sup>In order to improve on the uncertainty in the amplitude ratio it appears important to measure more accurately the slope of the  $\lambda$  line under pressure, because it is mainly this quantity which leads to the large estimated error.