

Phys. Fluids **17**, 1895 (1974).

<sup>5</sup>N. Hershkowitz, K. N. Leung, and T. Romesser, Phys. Rev. Lett. **35**, 277 (1975).

<sup>6</sup>S. Kogoshi, K. N. Sato, and T. J. Sekiguchi, J. Phys. D **11**, 1057 (1978).

<sup>7</sup>J. R. Greig and R. E. Pechacek, Appl. Phys. Lett. **29**, 798 (1976).

<sup>8</sup>W. D. Friedman, G. M. Halpern, and B. A. Brinker, Rev. Sci. Instrum. **45**, 1245 (1974).

<sup>9</sup>H. Baumhacker, H. Brinkschulte, R. S. Lang, W. Riedmuller, and M. Salvat, Appl. Phys. Lett. **30**, 461 (1977).

<sup>10</sup>A. F. Haught and D. H. Polk, Phys. Fluids **13**, 2825 (1970).

## Dynamical Scaling and Critical-Point Universality of Fluids

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The hypothesis of dynamical scaling asserts that, sufficiently close to the critical temperature, the decay rate  $\Gamma$  of the order-parameter fluctuations assumes the form  $\Gamma = q^z \Omega(q\xi)$ , where  $q$  is the wave number and  $\xi$  the correlation length. A new accurate experimental study of this decay rate in 3-methylpentane-nitroethane yields the value  $3.06 \pm 0.02$  for the dynamical scaling exponent  $z$ , in excellent agreement with the theoretical predictions for the dynamic universality class of fluid systems.

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It is generally believed that the asymptotic behavior of the static properties of fluids near the gas-liquid critical point and of binary liquids near the critical mixing point are determined by the universality class of the three-dimensional Landau-Ginzburg-Wilson model.<sup>1,2</sup> In recent years, experimental evidence in support of the validity of the hypothesis has been reported, provided that the critical point is approached sufficiently closely.<sup>3</sup>

In this Letter we report an experimental study of the dynamical scaling exponent that characterizes the asymptotic scaling behavior of the *time-dependent* order-parameter correlation function of fluids. To describe dynamic critical phenomena, systems can be grouped into *dynamic* universality classes.<sup>4</sup> In particular, fluids near the gas-liquid critical point and binary liquids near the critical mixing point are expected to belong to the same dynamic universality class. The hypothesis of dynamical scaling, originally proposed by Ferrell *et al.* and by Halperin and Hohenberg, and supported by the renormalization-group theory of dynamic critical phenomena, asserts that for sufficiently small wave numbers  $q$  and sufficiently close to the critical temperature  $T_c$ , the decay rate  $\Gamma$  of the order-parameter fluctuations should assume the form<sup>4</sup>

$$\Gamma = q^z \Omega(q\xi), \quad (1)$$

where  $\xi$  is the correlation length and  $z$  a universal dynamical scaling exponent. The dynamical scaling exponent of three-dimensional fluids satisfies the relation  $z = 3 + x_\eta$ , where  $x_\eta$  is the exponent of the power law  $\eta \propto \xi^{x_\eta}$  for the shear viscosity.

The scaling function  $\Omega(x)$  satisfies the boundary conditions

$$\lim_{x \rightarrow 0} \Omega(x) \propto x^{-(1+x_\eta)}; \quad \lim_{x \rightarrow \infty} \Omega(x) = \text{const.} \quad (2)$$

Thus in the hydrodynamic regime ( $x \rightarrow 0$ )  $\Gamma$  will vary as  $q^2$  at constant temperature, while in the "critical" regime ( $x \rightarrow \infty$ )  $\Gamma$  will vary as  $q^z$ . Using a perturbation expansion in  $\epsilon = 4 - d$  ( $d = \text{dimensionality}$ ) up to order  $\epsilon^2$ , one finds from the dynamic renormalization-group theory<sup>5</sup> and from the mode-coupling theory<sup>6</sup> the exponent value  $x_\eta = 0.065$ , so that  $z = 3.065$ .

The time-dependent correlation function is most conveniently measured by light scattering. However, because of multiple-scattering and gravity effects, it is very difficult to determine an accurate value of the exponent  $z$  for fluids near the gas-liquid critical point.<sup>7</sup> The experimental values of the exponent  $z$  for binary liquids near the critical mixing point have tended to be somewhat smaller than the value predicted theoretically. The most recent value is  $z = 2.992 \pm 0.014$  reported by Sorensen *et al.* from light-

scattering measurements in 3-methylpentane-nitroethane.<sup>8</sup> This value is in good agreement with the value  $z = 2.99 \pm 0.05$  found earlier by Chang *et al.* for the same mixture,<sup>9</sup> but in definite disagreement with the theoretically predicted value  $z = 3.065$ .

Experience with the analysis of static properties of fluids near the critical point has indicated that the asymptotic power laws become valid provided the temperature is within about  $10^{-3}$  or  $10^{-4}$  from the critical temperature.<sup>3</sup> Because of the additional assumptions in deriving the critical behavior of the dynamic properties, the range of validity of the asymptotic power laws for the dynamic properties may be even smaller. For instance, the viscosity  $\eta$  is often separated into a critical viscosity enhancement  $\Delta\eta$  and a normal or background viscosity  $\bar{\eta}$ . In deriving the asymptotic equations it is assumed implicitly that the critical point is approached sufficiently closely so that  $\Delta\eta \gg \bar{\eta}$ ,<sup>4,10</sup> a condition never realized in practice. In the case of the viscosity this problem is approached by postulating that the viscosity anomaly is a multiplicative anomaly of the form<sup>11</sup>

$$\eta = \bar{\eta}(q, \xi)^x \eta. \quad (3)$$

When one fits the experimental viscosity data of the 3-methylpentane-nitroethane mixture with (3), taking into account the temperature dependence of  $\bar{\eta}$ , one finds  $x_{\eta} = 0.0635 \pm 0.0004$  in good agreement with the theoretical prediction.<sup>12</sup> In the case of the decay rate  $\Gamma$  it is suggested that for actual experimental data not asymptotically close to the critical point Eq. (1) should be replaced by<sup>13</sup>

$$\Gamma = q^z \Omega(q, \xi) R(q, \xi), \quad (4)$$

where  $R(q, \xi)$  is a function of both  $q$  and  $\xi$ , i.e., a function of both  $q$  and the temperature  $T$ . Clearly, in order to determine the dynamical exponent  $z$  accurately, experimental data very close to the critical temperature  $T_c$  are needed.

We have made a dedicated effort to measure the decay rate  $\Gamma$  of 3-methylpentane-nitroethane mixture close to the critical temperature with a higher accuracy than obtained heretofore. Using the same sample and optical cell previously used in determining the static correlation function for the mixture,<sup>14</sup> we measured the autocorrelation function of the scattered light intensity. Light of a He-Ne laser was focused at the center of the cell after passing an intensity stabilizer consisting of a variable attenuator with a servo loop.

The scattered light intensity was detected by a photomultiplier tube whose signal was analyzed by a clipped correlator with 128 channels connected to a minicomputer. The data were taken as a function of the incident light intensity and extrapolated to zero intensity, thus eliminating local heating effects.<sup>8,14</sup> The measurements were corrected for any after-pulsing effects in the photomultiplier tube as described elsewhere.<sup>15</sup> The data were taken at three different scattering angles,  $29.8^\circ$ ,  $89.5^\circ$ , and  $146.8^\circ$ , corresponding to  $q = 7.04 \times 10^4 \text{ cm}^{-1}$ ,  $1.924 \times 10^5 \text{ cm}^{-1}$ , and  $2.621 \times 10^5 \text{ cm}^{-1}$ , respectively. In principle, light will be scattered not only from the incident beam, but also from the beam reflected at the cell wall. This effect was negligibly small at the scattering angles of  $29.8^\circ$  and  $89.5^\circ$ , but a correction was required for the measurements obtained at  $146.8^\circ$ . Details will be presented in a future publication.

It is noted that the dynamical scaling prediction (1) does not presuppose exponential decay of the time-dependent correlation function,<sup>4</sup> but applies to any suitably defined decay rate  $\Gamma$ . Small, but detectable, deviations from exponential decay have been found very close to the critical point.<sup>16</sup> For the purpose of the analysis we define  $\Gamma$  as the effective decay rate of the time-dependent correlation function  $g^{(1)}(\tau)$ , when fitted by an exponential decay law  $\exp(-\Gamma\tau)$  over a fixed time interval  $0 < \tau < p\Gamma^{-1}$ . In our experiment  $p$  was equal to  $1.0 \pm 0.2$  for all runs, in spite of the fact that the decay rate  $\Gamma$  varied by two orders of magnitude in the range of the experimental temperatures and wave numbers. The decay rate data obtained experimentally are shown in Fig. 1

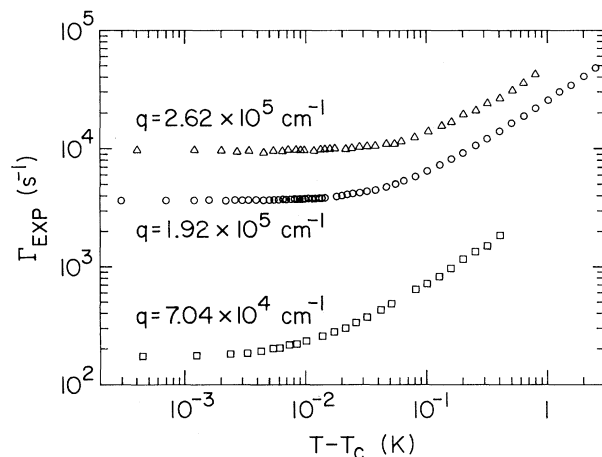


FIG. 1. Experimental values of the decay rate  $\Gamma$  as a function of  $T - T_c$ .

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We try to determine the dynamical scaling exponent  $z$  without assuming an explicit approximation for the functions  $\Omega(q\xi)$  and  $R(q, \xi)$ , which are theoretically known with limited accuracy only. We do this by fitting the decay rate data  $\Gamma(T)$  at each temperature by a power law of the form

$$\Gamma(T) \propto q^{z_{\text{eff}}(T)}. \quad (5)$$

This procedure defines an effective exponent  $z_{\text{eff}}$  as a function of temperature which away from  $T_c$  should approach the value 2 as predicted by the laws of hydrodynamics, while  $z_{\text{eff}}$  should approach  $z$  in the limit  $T \rightarrow T_c$ . The values obtained for  $z_{\text{eff}}$  as a function of temperature are shown in Fig. 2. In determining  $z_{\text{eff}}$ , the decay rate data were corrected for a small background contribution determined by a procedure proposed by Oxtoby and Gelbart<sup>13</sup> and discussed elsewhere.<sup>12</sup> However, this correction only affects the data far away from  $T_c$  and has, therefore, no effect on the determination of the dynamical scaling exponent  $z$ . In principle, the data should also be corrected for multiple-scattering effects.<sup>17</sup> Because of the small cross section for light scattering of the 3-methylpentane-nitroethane mixture,<sup>14</sup> this effect on the decay rate  $\Gamma$  was found to be negligibly small at all temperatures.

The data in Fig. 2 show the crossover from the hydrodynamic regime to the critical regime. Because of the low light-scattering intensity, we were unable to obtain accurate data in the hydrodynamic regime, i.e., at temperatures  $T - T_c \geq 0.5^\circ\text{C}$ . However, here we focus our attention on the approach to the extreme critical regime. For this purpose we show in Fig. 3 the values obtained for  $z_{\text{eff}}$ , together with their standard de-

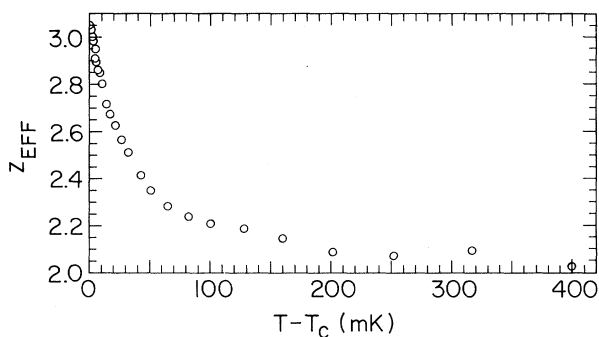


FIG. 2. Values of the effective exponent  $z_{\text{eff}}$  as a function of  $T - T_c$ .

viations, over the last 10 mdeg from the critical temperature. Our results are in satisfactory agreement with the less precise value  $z_{\text{eff}} = 2.98 \pm 0.05$  at  $\Delta T = 3$  mK earlier obtained by Chang *et al.* for the same mixture.<sup>9</sup> However, our new more accurate data indicate that the asymptotic limit has not yet been reached at that temperature. Within the experimental accuracy,  $z_{\text{eff}}$  can be represented by a linear function of  $\Delta T$  for  $\Delta T \leq 10$  mK and we obtain

$$z = \lim_{T \rightarrow T_c} z_{\text{eff}}(T) = 3.063 \pm 0.024, \quad (6)$$

where the quoted error represents *two* standard deviations. This result is in excellent agreement with the theoretical prediction, as well as with the exponent value  $x_\eta = 0.0635 \pm 0.0004$  deduced from the viscosity, thus removing the apparent disagreement with the theory recently reported by Sorensen *et al.*<sup>8</sup>

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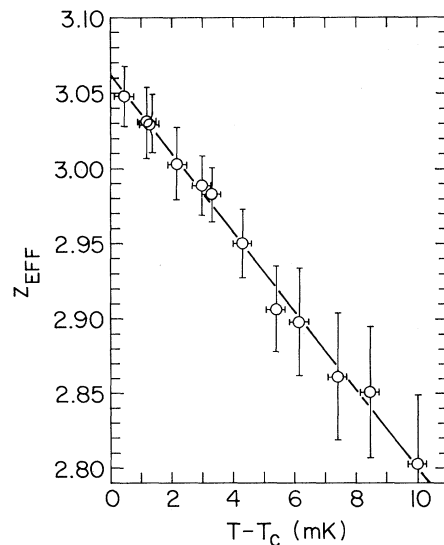


FIG. 3. Values of the effective exponent  $z_{\text{eff}}$  as a function of  $T - T_c$  close to the critical temperature.

ence Center of the University of Maryland.

<sup>1</sup>J. Hubbard and P. Schofield, Phys. Lett. **40A**, 225 (1972).

<sup>2</sup>J. F. Nicoll, Phys. Lett. **76A**, 112 (1980).

<sup>3</sup>For a review see A. Levelt Sengers, R. J. Hocken, and J. V. Sengers, Phys. Today **30**, No. 12, 42 (1977).

<sup>4</sup>For a review see P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977).

<sup>5</sup>E. D. Siggia, B. I. Halperin, and P. C. Hohenberg, Phys. Rev. B **13**, 2110 (1976).

<sup>6</sup>J. D. Gunton, in *Dynamical Critical Phenomena and Related Topics*, edited by C. P. Enz (Springer-Verlag, New York, 1979), p. 1.

<sup>7</sup>H. Güttinger and D. S. Cannell, Phys. Rev. A (to be published).

<sup>8</sup>C. M. Sorensen, R. C. Mockler, and W. J. O'Sullivan, Phys. Rev. Lett. **40**, 777 (1978).

<sup>9</sup>R. F. Chang, P. H. Keyes, J. V. Sengers, and C. O. Alley, Phys. Rev. Lett. **27**, 1706 (1971), and Ber. Bunsenges. Phys. Chem. **76**, 260 (1972).

<sup>10</sup>F. Garisto and R. Kapral, Phys. Rev. A **14**, 884 (1976).

<sup>11</sup>T. Ohta, J. Phys. C **10**, 791 (1977).

<sup>12</sup>H. C. Burstyn, J. V. Sengers, and P. Esfandiari, Phys. Rev. A (to be published).

<sup>13</sup>D. W. Oxtoby and W. M. Gelbart, J. Chem. Phys. **61**, 2957 (1974).

<sup>14</sup>R. F. Chang, H. Burstyn, and J. V. Sengers, Phys. Rev. A **19**, 866 (1979).

<sup>15</sup>H. C. Burstyn, to be published.

<sup>16</sup>H. C. Burstyn, R. F. Chang, and J. V. Sengers, Phys. Rev. Lett. **44**, 410 (1980).

<sup>17</sup>R. A. Ferrell and J. K. Bhattacharjee, Phys. Rev. A **19**, 348 (1979).

## Observation of a New Sound-Attenuation Peak in Superfluid <sup>3</sup>He-B

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Results of zero-sound attenuation measurements in <sup>3</sup>He-B, at frequencies up to 60 MHz and pressures between 0 and 20 bars, are reported. At frequencies of 30 MHz and above, a new attenuation feature is observed which bears the signature of a collective mode of the superfluid.

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Zero sound has proven to be an important probe of the quantum properties of liquid <sup>3</sup>He. The first observations of zero sound by Keen, Matthews, and Wilks<sup>1</sup> and Abel, Anderson, and Wheatley<sup>2</sup> provided a striking confirmation of the Landau theory of a normal Fermi liquid. Soon after the discovery of the superfluid phases of <sup>3</sup>He, measurements by Lawson *et al.*<sup>3</sup> and Paulson, Johnson, and Wheatley<sup>4</sup> revealed a large peak in the attenuation of zero sound located just below the superfluid transition temperature ( $T_c$ ). Such peaks, occurring in both <sup>3</sup>He-A and <sup>3</sup>He-B, have since been attributed to the resonant excitation, by the sound wave, of a collective oscillation of the superfluid, as well as to sound absorption through direct breaking of Cooper pairs.<sup>5-7</sup> For <sup>3</sup>He-B, the resonant excitation of the collective mode is predicted to occur at a temperature satisfying the relation

$$\hbar\omega = \left(\frac{12}{5}\right)^{1/2} \Delta_B(T), \quad (1)$$

where  $\Delta_B(T)$  is the energy gap in <sup>3</sup>He-B and  $\omega$  is the angular frequency of the sound wave. Pair breaking takes place in the temperature interval  $\hbar\omega \geq 2\Delta_B(T)$ . Measurements<sup>4</sup> up to 25 MHz show a single attenuation peak near  $T_c$ , which is presumably caused by an overlap of these two peaks due to quasiparticle broadening effects.<sup>8</sup> In this Letter we report results of zero-sound attenuation measurements in <sup>3</sup>He-B at a variety of pressures between 0 and 20 bars and frequencies between 10 and 60 MHz.<sup>9</sup> Our most striking result is the observation of an unexpected attenuation peak, in addition to the one usually observed,<sup>4,10</sup> which bears the clear signature of a collective mode of the liquid.

The measurements reported here were carried out in two separate cells, each mounted in a nuclear demagnetization cryostat capable of operation down to 0.7 mK. The first cell contained two 10-MHz fundamental X-cut quartz transducers separated by 0.769 cm. The second cell contained