Crossover Behavior in the Susceptibility near the ^{3}He Critical Point

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We present high-resolution measurements of the isothermal susceptibility of pure ^{3}He near the liquid-gas critical point. The PVT Measurements were performed in the single phase region along the critical isochore over the reduced temperature range $3\times10^{-5} < T/T_c - 1 < 1.5\times10^{-1}$. The crossover behavior of the susceptibility was analyzed using a field theoretical renormalization group calculation based on the ϕ^4 model. A similar crossover analysis was performed on previously obtained Xe susceptibility measurements. Comparison of the effective susceptibility exponent for those two fluids shows theoretically predicted universal crossover behavior.

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It is well known that thermodynamic quantities exhibit singularities asymptotically close to the critical point. The power law behavior of these singularities, characterized by critical exponents and the concept of universality and scaling, have been successfully described by renormalization-group (RG) theory [1]. Earlier experimental studies of critical phenomena were mostly dedicated to the quest of *true* asymptotic behavior. Recently there has been a renewed interest in understanding critical crossover phenomena from asymptotic to classical critical behavior [2]. Away from the asymptotic region, thermodynamic quantities of real physical systems deviate from simple power law behavior. However, RG theory can still provide insight in understanding correction-to-scaling behavior as long as the correlation length is comparable or larger than the characteristic length scale of a system.

In a fluid system, the isothermal susceptibility is defined as $\chi_T \equiv \rho\,(\partial\rho/\partial P)_T$, where ρ is the fluid density and P is the pressure. In the single-phase region, the normalized susceptibility, $\chi_T^* = \frac{P_c}{\rho_c^2}\chi_T$ is conveniently characterized by the Wegner series expansion [3]

$$\chi_T^* = \Gamma_0^+ t^{-\gamma}(1 + \Gamma_1^+ t^{\Delta} + \Gamma_2^+ t^{2\Delta} + a_1 t + \ldots), \qquad (1)$$

where $t = (T - T_c)/T_c$ is the reduced temperature and $\Gamma_0^+, \Gamma_1^+, \Gamma_2^+$, and a_1 are non-universal system dependent amplitudes. Theoretical values for the critical exponents [4] are $\gamma = 1.239 \pm 0.002$ and $\Delta = 0.504 \pm 0.008$. Experimental data in the crossover region can be characterized by an effective susceptibility exponent $\gamma_{\rm eff} = -d\ln\chi_T/d\ln|t|$. In a recent paper Anisimov et al. [5] investigated the critical crossover behavior of several fluid systems by using a phenomenological crossover model of the susceptibility based upon a RG matching technique [6]. They considered the effective exponent $\gamma_{\rm eff}$ and found that for $T \to T_c$, $\gamma_{\rm eff}$ can approach the asymptotic value γ either from below or from above. This implies that the Wegner amplitude Γ_1^+, which describes the leading correction, is either positive or negative depending on the particular fluid system.

In this letter, we present high resolution measurements of the ^{3}He susceptibility near the liquid-gas critical point. The data are compared with the RG theory for the ϕ^4 model of Schloms and Dohm [7]. The Wegner corrections are included to all orders in the crossover region by numerically integrating the RG equations to determine the susceptibility and the effective exponent. We have also compared the crossover behavior of ^{3}He to a similar analysis of susceptibility measurements in Xe [8] which is a room temperature classical fluid.

The experimental cell used to measure the susceptibility was made of Oxygen Free High Conductivity Copper. The sample was contained in a flat pancake cell 0.05 cm high and 11 cm in diameter. The small cell height was chosen to minimize the effect of gravity. The cell was mounted on a thermal stage that was surrounded by a radiation shield stage. High purity ^{3}He ($<$ 0.2 ppm ^{4}He) was used for the experiment. A fill line was connected to an in-situ charcoal adsorption pump. A Straty-Adams type capacitive pressure gauge was mounted in the middle of the cell to directly measure the pressure of the fluid. A density sensor consisting of a capacitor with a 50 μm gap was also located in the middle of the cell. The density of the fluid was determined from the measured dielectric constant using the Clausius-Mossotti relation. Both a conventional Germanium resistance thermometer and a $GdCl_3$ high resolution paramagnetic susceptibility thermometer [9] measured the temperature of the cell wall. An advantage of this cell design is the ability to perform other thermodynamic measurements like the specific heat as well as *PVT* measurement.

The cell was also equipped with three equally-spaced leveling capacitor sensors. Before performing the *PVT* measurements, the cell was leveled by monitoring the average density of the fluid at the leveling capacitors. During the experiment the leveling was maintained to better than 0.01 degree. After the leveling was completed, the susceptibility of the sample was measured along constant temperature isotherms using a conventional *PVT* measurement technique [10]. For these measurements, a low temperature valve was opened and the density of the sample was decreased linearly by controlling the temperature of the in-situ charcoal adsorption pump.

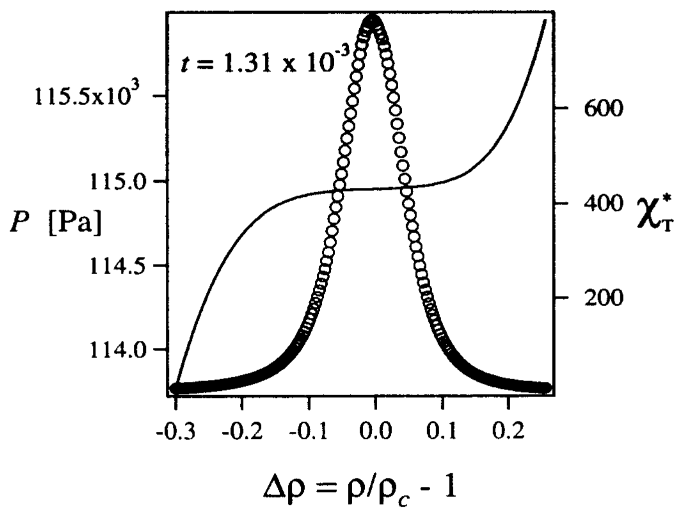

FIG. 1. A typical *PVT* run at $t = 1.31 \times 10^{-3}$. The solid line shows the pressure P as a function of the density difference $\Delta\rho$. The susceptibility χ_T^*, obtained from the slope of the $P-\rho$ curve, is shown as open circles. Data were taken while reducing the density of the sample.

Figure 1 shows a typical isotherm run at $t = 1.31 \times 10^{-3}$ that covered the density range $-0.3 < \Delta\rho \equiv \rho/\rho_c - 1 < 0.3$. The solid line shows the pressure P as a function of the density difference $\Delta\rho$. Relatively slow sweep rates of 5-10 hours per isotherm were chosen to minimize the density inhomogeniety in the sample. Temperature stability of the sample during the ramping was typically better than 3 μK (rms). The susceptibility was determined from the slope of the measured isotherm (solid line) and is represented in Fig. 1 by the open circles. Although the susceptibility was measured throughout the critical region, this letter will only report on measurements along the critical isochore. Gravity induced a vertical density inhomogeniety in the cell due to the strong divergence of the susceptibility. We have estimated the gravity effect within the cell and density sensor using a cubic model equation of state [11]. These calculations predict a 1% correction in the susceptibility due to Earth's gravity at $t = 5.5 \times 10^{-5}$ for the density capacitor gap and $t = 2.5 \times 10^{-4}$ for the cell height. Gravity corrections close to the transition introduce an uncertainty in the determination of the critical temperature. In this data analysis, the different choices of the critical temperature within the experimental uncertainty resulted in the exponent γ being between 1.18 to 1.24. The data were analyzed with current theoretical values of the exponents, γ and Δ [4], in order to test *universal* crossover behavior near the liquid-gas critical point.

Isothermal susceptibility data along the critical isochore are plotted as a function of reduced temperature in Fig. 2. The leading power law dependence was eliminated by multiplying the susceptibility by t^γ. The experimental data are shown as open circles. The asymptotic critical amplitude, Γ_0^+ is obtained in the limit of $t \to 0$ (dashed line). The curvature in the data indicates the crossover behavior.

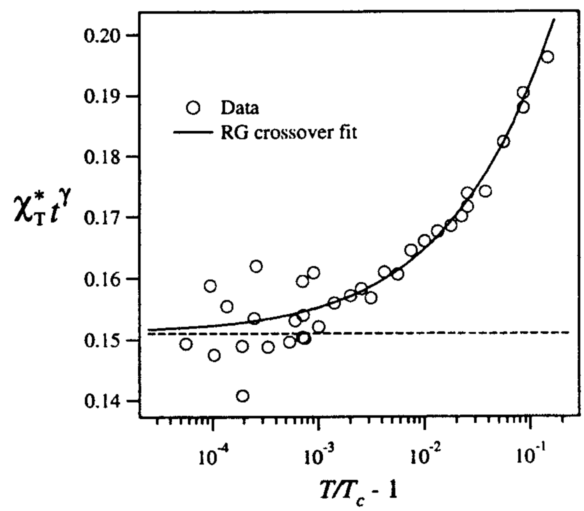

FIG. 2. The susceptibility multiplied by t^γ, plotted against the logarithm (to base 10) of the reduced temperature. The solid curve is the best RG fit, and the open circles are the data points. The details of RG fit is discussed in the text.

For comparison we numerically calculated the susceptibility by applying the RG theory for the ϕ^4 model developed by Schloms and Dohm [7] to the $O(1)$ universality class. We find the susceptibility

$$\chi_T^* = \chi_0 \, l^{-\gamma/\nu} \exp[-F_\phi(l)] \, / \, f_+(u(l)) \tag{2}$$

depends on the RG flow parameter l, that is related to the reduced temperature t by

$$t = t_0 \, l^{1/\nu} \exp[-F_r(l)] \, . \tag{3}$$

The renormalization factors on the right hand side of Eqs. (2) and (3) are represented by the exponential functions with the integrals

$$F_r(l) = \int_0^l \frac{dl'}{l'}[\zeta_r(u(l')) - \zeta_r(u^*)] \ , \tag{4}$$

$$F_\phi(l) = \int_0^l \frac{dl'}{l'}[\zeta_\phi(u(l')) - \zeta_\phi(u^*)]. \tag{5}$$

Here $u(l)$ is the renormalized coupling parameter of the ϕ^4 model that satisfies the differential equation

$$l\frac{d}{dl}u(l) = \beta(u(l)) \ . \tag{6}$$

The RG functions $\beta(u)$, $\zeta_r(u)$, and $\zeta_\phi(u)$ have been determined by Schloms and Dohm [7] via Borel resummation of high-order perturbation series. For the dimension $d = 3$ and a $n = 1$ component order-parameter field $\phi(\mathbf{r})$ they become

$$\beta(u) = -u + 36u^2(1 + a_4u)/(1 + a_5u) \ , \tag{7}$$

$$\zeta_r(u) = 12u - 120u^2 + a_1u^3 - a_2u^4 \ , \tag{8}$$

$$\zeta_\phi(u) = -24u^2 + a_3u^3 \ , \tag{9}$$

where $a_1 = 3075$, $a_2 = 30390$, $a_3 = 37.5$, $a_4 = 14.10$, and $a_5 = 31.85$. The amplitude function $f_+(u)$ in (2) was calculated by Krause et al. [12] using a Borel resummation technique. The result is

$$f_+(u) = 1 - \frac{92}{9}u^2(1 + b_\chi u) \ , \tag{10}$$

where $b_\chi = 9.68$. The susceptibility $\chi^*_T(t)$ was calculated by evaluating Eqs. (2)-(10) numerically. First, Eq. (6) was solved together with Eq. (7) and the initial condition $u(l = 1) = u$ to obtain the renormalized coupling $u(l)$ as a function of the RG flow parameter l. The renormalized coupling approaches the fixed point value $u^* = 0.0405$ [7] in the limit $l \to 0$. The functions $F_r(l)$ and $F_\phi(l)$ were then obtained by evaluating the integrals in Eqs. (4) and (5) together with Eqs. (8) and (9). $\chi^*_T(l)$ and $t(l)$ are determined from (2), (3), and (10). The susceptibility $\chi^*_T = \chi^*_T(t)$ was then obtained by eliminating l.

The theoretical susceptibility $\chi^*_T(t)$ contains three nonuniversal parameters, χ_0, t_0, and the initial value u. Only two of them, say χ_0 and t_0, are independent, while the third one u is irrelevant because the RG flow parameter l can be eliminated. The theoretical susceptibility $\chi^*_T(t)$ was fitted to the experimental data by adjusting the two nonuniversal amplitudes χ_0 and u while t_0 is kept constant. The best result for $\chi_0 = 0.254$, and u/u^* = 0.34 is shown by the solid line in Fig. 2. The theoretical curve agrees very well with the experimental data. We find that the Wegner corrections to the asymptotic power law, which are included in the theory, are essential for the explanation of the experimental data. The difference between the solid and dashed lines is due to the Wegner corrections. The theoretical susceptibility $\chi^*_T(t)$ can be expanded in the series given in Eq. (1), where the amplitudes Γ_0^+, Γ_1^+, etc. are expressed in terms of the two nonuniversal fit parameters t_0 and χ_0. From the fit in Fig. 2 we find $\Gamma_0^+ = 0.152$ and $\Gamma_1^+ = 0.93$ for ^{3}He.

The effective exponent $\gamma_{\rm eff}$ obtained from the ^{3}He susceptibility measurements is shown by the solid line in Fig. 3. The effective exponent $\gamma_{\rm eff}$ is monotonically decreasing in the crossover region, which implies that the Wegner amplitude Γ_1^+ is positive, or equivalently that $u/u^* < 1$. This analysis indicates that the effective susceptibility exponent, $\gamma_{\rm eff}$, changes monotonically from the asymptotic critical value (γ=1.239) close to the transition to the classical value farther away. The crossover behavior is not completed before leaving the critical region.

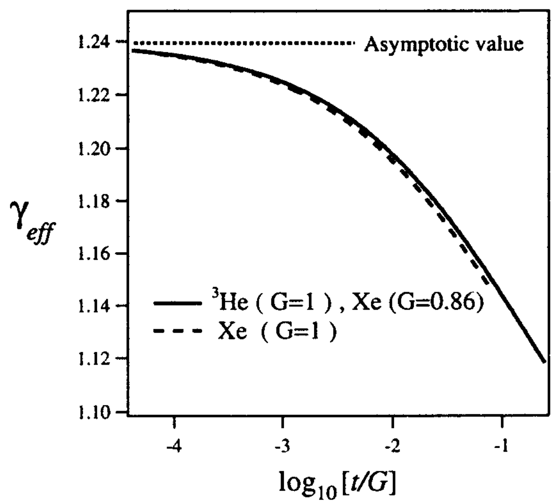

FIG. 3. $\gamma_{\rm eff}$ vs normalized reduced temperature. $\gamma_{\rm eff}$ is the slope of $\log(\chi_T)$ vs $\log(t)$ plot. The reduced temperature is scaled by a arbitrary factor G.

Güttinger and Cannell [8] have measured the susceptibility near the liquid gas critical point in Xe. Their data can also be fitted with the RG theoretical susceptibility. For comparison we have determined the Xe effective exponent $\gamma_{\rm eff}$ which is shown in Fig. 3 by the dashed line. In this case a slightly larger Wegner amplitude $\Gamma_1^+ = 1.0$ is obtained. In Fig. 3 the reduced temperature is scaled by a factor G. By adjusting $G = 0.86$, the $\gamma_{\rm eff}$ for Xe was collapsed onto the ^{3}He curve. The theoretically expected scale factor G between Xe and ^{3}He can be determined from the expression

$$G = (\Gamma^+_{1,\rm Xe}/\Gamma^+_{1,^3\rm He})^{-1/\Delta} \tag{11}$$

using the leading Wegner amplitudes. By inserting the above Wegner amplitudes we obtain $G = 0.86$ in agreement with experiment. The experimental data are actually consistent with this theoretical scale factor well beyond the reduced temperature region associated with the leading Wegner amplitude. This universal crossover behavior is predicted by the ϕ^4 crossover model and has also been recently demonstrated by numerical simulation of spin systems [13]. We are unaware of any previous experimental demonstration of the existence of a universal crossover curve in simple fluid systems.

There should be a relationship between scale factor G and the Ginzberg number N_{Gi} that provides a quantitative means of locating the boundary between the region in which mean field theory is valid and the regime in which fluctuations renormalize the critical behavior of the system. The smaller the Ginzburg number, the closer one must approach to the critical point before fluctuations significantly modify the thermodynamics of the system. The Ginzburg number itself is a nonuniversal amplitude that can not be calculated by the RG theory.

We interpret the scale factor G as the ratio of the Ginzberg numbers for Xe and ^{3}He, $G = N_{Gi,\mathrm{Xe}}/N_{Gi,^3\mathrm{He}}$. In Ref. [13], it is argued that the Ginzburg number, N_{Gi}, depends on the effective range R of the interaction via the relationship $N_{Gi} \sim R^{-6}$ in $d = 3$ dimensions. This implies $N_{Gi,Xe}/N_{Gi,^3\mathrm{He}} = R^6(^3\mathrm{He})/R^6(\mathrm{Xe}) \sim \xi_0^6(^3\mathrm{He})/\xi_0^6(\mathrm{Xe}) = 6.57$, where ξ_0 is the correlation length amplitude. The difference between this ratio and the result for the scale factor G obtained in this analysis cannot be accounted for by either random or systematic experimental error. This suggests that more than one property of the system controls the factor N_{Gi}. We believe that other microscopic details need to be included in defining the Ginsburg number. This conclusion is consistent with the latest phenomenological crossover model and numerical studies of three dimensional Ising model [14].

In the case of ^{3}He, the microscopic detail is governed by quantum effects, which are not taken into account in the standard Ginzburg-Landau Wilson formulation of the effective Hamiltonian. We expect quantum effects to become important when the correlation length is comparable to or smaller than the de Broglie wavelength. The estimated correlation length of ^{3}He is approximately equal to the de Broglie wavelength near $t \sim 0.4$. Thus, we do not expect quantum effects will significantly affect measurements close to the transition. At this time, calculations of the precise influence of quantum effects on crossover behavior have not been performed for this system.

The quality of the fit of the RG-based crossover form to the present data over more than four decades in reduced temperature is encouraging. However, it should be noted that fits of comparable quality are obtained using both an alternative crossover form based on work of Nicoll and Albright [15,16] and the asymptotic power-law behavior modified by the first Wegner correction, i.e. Eq. (1) with no coefficients beyond Γ_1^+ retained. Data capable of distinguishing between the various candidates for the crossover to asymptotic critical behavior should be available once measurements have been performed closer to the transition in a microgravity environment [17].

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