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Crossover behavior in the isothermal 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. *PVT* measurements were performed in the single-phase region over the reduced temperature range $3\times10^{-5}<T/T_c-1<1.5\times10^{-1}$. The crossover behavior of the susceptibility along the critical isochore 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. A comparison of the rescaled susceptibility for ^{3}He and Xe 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 critical crossover behavior as long as the order parameter correlation length is larger than the characteristic microscopic length scale of a system. In this Rapid Communication, we present measurements of the isothermal susceptibility, $\chi_T=\rho(\partial\rho/\partial P)_T$, of pure ^{3}He near the liquid-gas critical point. The experimental data are analyzed using the RG-based ϕ^4 model with the minimal-subtraction renormalization scheme developed by Dohm and co-workers [3,4]. We have also compared the crossover behavior of the isothermal susceptibility in ^{3}He and Xe [5], and demonstrated universality in the crossover from the asymptotic to the mean-field regimes.

The isothermal susceptibility was obtained from isotherm *PVT* measurements that were performed in a copper cell. High purity ^{3}He (<0.2 ppm ^{4}He) was contained in a flat pancake volume with internal dimensions 0.05 cm height and 11 cm diameter. A Straty-Adams type capacitive pressure gauge with 0.2 ppm resolution was mounted in the middle of the cell. A capacitor with a 50 μm gap was also located in the middle of the cell. The density of the sample was determined with 2 ppm resolution from the measured dielectric constant using the Clausius-Mossotti relation. Both a Germanium resistance thermometer with resolution of 1.6 ppm and a $GdCl_3$ high-resolution paramagnetic susceptibility thermometer [6] with resolution of 0.3 ppb measured the cell wall temperature. Typical uncertainty in measuring temperature during a *PVT* run was ~5 μK. The details of the experimental setup and the resolution of the sensors are given in Ref. [7]. The cell was also equipped with three equally spaced leveling capacitor sensors. Before performing the *PVT* measurements, the cell was leveled *in situ* by monitoring the average density of the sample in the two-phase region at the leveling capacitors. During the experiment the leveling angle of the cell was maintained to better than $\pm0.03°$ peak to peak. By placing both the density and pressure sensors at the mid-plane of the leveled cell, the gravity induced, vertical density inhomogeneity was only relevant for the density sensor gap. Our calculation, using the restricted cubic model for the equation-of-state [8], predicts a 1% correction in the susceptibility due to gravity at $t=5.5\times10^{-5}$ for the density capacitor gap.

The sample density in the cell was lowered by decreasing the temperature of an *in situ* charcoal adsorption pump. Relatively slow ramping rates of $\delta\rho/\rho_c\sim1-2\%$ per hour were chosen to minimize the density inhomogeneity in the sample. Figure 1 shows a typical P-ρ curve at $t=1.31\times10^{-3}$ that covered the reduced density range $-0.25<\Delta\rho\equiv\rho/\rho_c-1<0.25$. The solid curve shows the measured pressure P as a function of the reduced density $\Delta\rho$. The open circles show the derived dimensionless susceptibility, $\chi_T^*=(P_c/\rho_c^2)\chi_T$ with an estimated 2% uncertainty, where $P_c=114.6$ kPa and $\rho_c=0.0137$ mole/cm^3 for ^{3}He. The present measurements agree to within 5% [7] with those of Pittman, Doiron, and Meyer [9]. Although the susceptibility was measured throughout the entire critical region, this paper will only report on measurements along the critical isochore.

The theoretical expressions for the susceptibility in the $O(1)$ universality class in three dimensions [3,4] were used to analyze our experimental measurements. These expressions were derived from the minimal-subtraction renormalization scheme within the ϕ^4 model. A different massive-renormalization scheme within the ϕ^4 model, developed by Bagnuls and Bervillier [10], was previously used to analyze Xe data [11]. The difference between the two schemes is

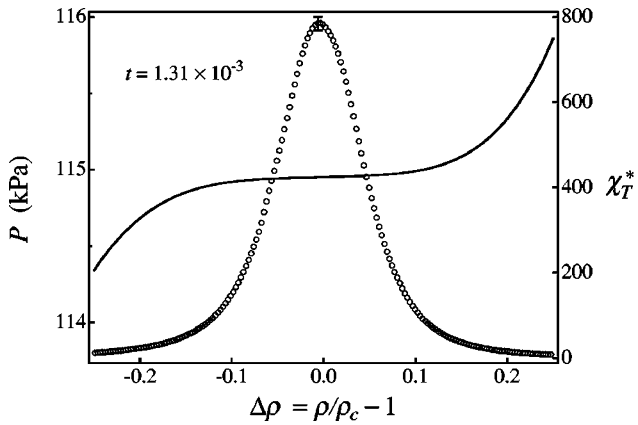

FIG. 1. A typical *PVT* run at $t=1.31\times10^{-3}$. The solid curve shows the measured pressure P as a function of the reduced density $\Delta\rho$ of ^{3}He. The susceptibility χ_T^*, obtained from the slope of the P-ρ curve, is shown as open circles with a 2% error bar near ρ_c.

discussed in detail in Ref. [3].

The Hamiltonian of the ϕ^4 model can be written as

$$H=\int d^3r\left\{\frac{1}{2}r_0\phi^2+\frac{1}{2}(\nabla\phi)^2+u_0\phi^4\right\}. \tag{1}$$

Here ϕ is the order parameter, u_0 is a bare coupling constant, and r_0 is related to the reduced temperature $t\equiv(T-T_c)/T_c$ by

$$r_0=a_0t, \tag{2}$$

where a_0 is a nonuniversal constant. Within this approach, the susceptibility and reduced temperature can be expressed in terms of a RG flow parameter l as

$$\chi_T^*=\chi_0 l^{-\gamma/\nu}\frac{\exp[-F_\phi(\overline{u}(l))]}{f_+(\overline{u}(l))}, \tag{3}$$

$$t=t_0Q(\overline{u}(l))l^{1/\nu}\exp[-F_r(\overline{u}(l))]. \tag{4}$$

The functions $F_r(\overline{u}(l))$ and $F_\phi(\overline{u}(l))$ are defined as

$$F_r(\overline{u}(l))=\int_{u*}^{\overline{u}(l)}\frac{d\overline{u}'}{\beta(\overline{u}')}[\zeta_r(\overline{u}')-\zeta_r(u^*)], \tag{5}$$

$$F_\phi(\overline{u}(l))=\int_{u*}^{\overline{u}(l)}\frac{d\overline{u}'}{\beta(\overline{u}')}[\zeta_\phi(\overline{u}')-\zeta_\phi(u^*)]. \tag{6}$$

Here $\overline{u}(l)$ is the effective coupling parameter of the ϕ^4 model that satisfies the differential equation

$$l\frac{d}{dl}\overline{u}(l)=\beta(\overline{u}(l)), \tag{7}$$

with the initial condition of $\overline{u}(l=1)=u$, where u is the renormalized coupling parameter of u_0 in Eq. (1).

The RG functions $\beta(\overline{u})$, $\zeta_r(\overline{u})$, and $\zeta_\phi(\overline{u})$ are expanded to two-loop order with extrapolation to the calculations of high-order Borel resummations at the fixed point [3]. For a system of dimension $d=3$ and single component order-parameter $n=1$, they become

$$\beta(\overline{u})=-\overline{u}+36\overline{u}^2(1+a_4\overline{u})/(1+a_5\overline{u}), \tag{8}$$

$$\zeta_r(\overline{u})=12\overline{u}-120\overline{u}^2+a_1\overline{u}^3-a_2\overline{u}^4, \tag{9}$$

$$\zeta_\phi(\overline{u})=-24\overline{u}^2+a_3\overline{u}^3, \tag{10}$$

where $a_1=3075$, $a_2=30390$, $a_3=37.5$, $a_4=14.10$, and $a_5=31.85$. The amplitude functions $f_+(\overline{u})$ in Eq. (3) and $Q(\overline{u})$ in Eq. (4) were calculated by Krause *et al.* [4] using a Borel resummation technique to give

$$f_+(\overline{u})=1-\frac{92}{9}\overline{u}^2(1+b_\chi\overline{u}), \tag{11}$$

$$Q(\overline{u})=1+b_Q\overline{u}^2\ln(c_Q\overline{u}), \tag{12}$$

where $b_\chi=9.68$, $b_Q=28.2$, and $c_Q=7.66$. The constant nonuniversal amplitudes χ_0 in Eq. (3) and t_0 in Eq. (4) can also be expressed in terms of two more fundamental parameters, μ and a as

$$\chi_0=\mu^{-2}Z_\phi(u)\exp[F_\phi(u)], \tag{13}$$

$$t_0=\frac{\mu^2}{a}\exp[F_r(u)]. \tag{14}$$

Here μ^{-1} is a reference length that links the flow parameter l to the correlation length ξ, $l=(\mu\xi)^{-1}$, and a is the renormalized temperature coefficient of a_0 defined in Eq. (2). The minimal-renormalization factor, Z_ϕ, in Eq. (13), is given by

$$Z_\phi(u)^{-1}=\exp\left(\int_0^u\frac{du'}{\beta(u')}\zeta_\phi(u')\right). \tag{15}$$

The theoretical susceptibility $\chi_T^*(t)$ contains three nonuniversal parameters, μ, a, and u. Once they are determined, we can calculate the correlation length as a function of reduced temperature in the crossover regime. Most importantly, the three parameters explicitly appear only in χ_0 and t_0. Thus, χ_T^*/χ_0 and t/t_0 are functions of the fluid-independent flow parameter l. Since l can be solved as a function of t/t_0 from Eq. (4), χ_T^*/χ_0 is a universal scaling function of t/t_0.

The theoretical susceptibility $\chi_T^*(t)$ was calculated by evaluating Eqs. (3)–(15) numerically and eliminating l in Eq. (3) using Eq. (4). The complete expression of $\chi_T^*(t)$ can also be expanded in a series around the fixed point, $\overline{u}(l=0)=u^*$, to generate the Wegner expansion for the pure ϕ^4 model [10,12],

$$\chi_T^*(t)=\Gamma_0^+t^{-\gamma}(1+\Gamma_1^+t^\Delta+\Gamma_2^+t^{2\Delta}+\cdots), \tag{16}$$

with

$$\Gamma_0^+ = \frac{\chi_0 [Q(u^*) t_0]^\gamma}{f_+(u^*)}, \tag{17}$$

and

$$\Gamma_1^+ = \left[\gamma \left(\frac{\zeta_r'}{\omega} - \frac{Q'}{Q} \right) + \frac{\zeta_\phi'}{\omega} + \frac{f_+'}{f_+} \right]_{u=u^*} \frac{u^* - u}{[Q(u^*) t_0]^\Delta}. \tag{18}$$

Here $\Delta = \nu\omega$, $\omega = d\beta/du|_{u^*}$, and ν is the critical exponent for the correlation length.

Among the three nonuniversal parameters, two are independent in the renormalization scheme. We chose to fix $u = 0.999u^*$ and let μ and a be determined from the fit of the theoretical susceptibility $\chi_T^*(t)$ to the experimental data. The choice of u/u^* value comes from the requirement of $u \sim u^*$ in the Γ_1^+ calculation using Eq. (18). In addition to μ and a, T_c was also adjusted in the fit. $\gamma = 1.2396$ and $\Delta = 0.504$ [13] were used in the calculation. The stated uncertainty of 2% in the susceptibility measurements leads to a weighting of $1/(0.02\chi_T^*)$ for each data point. No uncertainty in temperature was introduced in the fit. The nonlinear least-square fit using the Levenberg-Marquardt algorithm yielded $\mu = (2.08 \pm 0.17) \times 10^{-4}$, $a = 0.1363 \pm 0.0037$, and $T_c = 3.315534\ \text{K} \pm 3\ \mu\text{K}$. All the reported statistical uncertainties in the fitting parameters were from the covariance matrix of standard errors in the algorithm. The calculated correlation is 0.99 between μ and a and -0.5 between T_c and μ. Once u, μ, and a are known, one can calculate $t_0 = (3.17 \pm 0.52) \times 10^{-7}$, $\Gamma_0^+ = 0.1498 \pm 0.0077$, and $\Gamma_1^+ = 1.01 \pm 0.08$ from Eqs. (14), (17), and (18).

Güttinger and Cannell [5] measured the susceptibility near the liquid-gas critical point of Xe using a light scattering method. They analyzed their data using Eq. (16) up to $t^{3\Delta}$ with $\gamma = 1.241$, $\Delta = 0.496$. They obtained $T_c = 289.65$ K, $\Gamma_0^+ = 0.0577 \pm 0.0001$, and $\Gamma_1^+ = 1.29 \pm 0.03$ using five adjustable parameters. We refit their data with Eqs. (3)–(15) assuming a 2% uncertainty in the experimental data. With $u/u^* = 0.999$ fixed, the fitting parameters were $\mu = (2.97 \pm 0.33) \times 10^{-4}$, $a = 0.333 \pm 0.012$, and $T_c = 289.65002\ \text{K} \pm 27\ \mu\text{K}$. The rms difference between the data and fit is 0.14%. These parameters led to $t_0 = (2.65 \pm 0.59) \times 10^{-7}$, $\Gamma_0^+ = 0.0587 \pm 0.0040$, and $\Gamma_1^+ = 1.11 \pm 0.13$. This Γ_1^+ value compares well with $\Gamma_1^+ = 1.08$ obtained by Anisimov *et al.* [14].

To demonstrate universality in crossover behavior, the susceptibility and reduced temperature were scaled by their corresponding fluid-dependent parameters, χ_0 and t_0. The susceptibility χ_T^*/χ_0 was further scaled by the leading singularity $(t/t_0)^{-\gamma}$ to provide a more sensitive representation of crossover behavior. Based on Eq. (17), $\chi_0/t_0^{-\gamma}$ is proportional to Γ_0^+, therefore $\chi_T^*/(\Gamma_0^+ t^{-\gamma})$ is a universal function of t/t_0. Figure 2 shows the experimental data for both ^{3}He and Xe and the weighted, three parameter, least-squares fitted theoretical curve from the ϕ^4 analysis. The experimental data collapse by scaling the reduced temperature with t_0. The universal ϕ^4 theoretical solid curve agrees very well with the experimental data for both ^{3}He and Xe within the uncertainties of the measurements. The error bars in Fig. 2 represent a combined uncertainty in the scaled susceptibility from the measured χ_T^* and t. The error bar size of Xe is comparable to that of ^{3}He. The horizontal dot-dashed line indicates a pure asymptotic power law for $\chi_T^*(t)$. The difference between the best fits and dot-dashed line is due to correction-to-scaling effects. This universal crossover behavior, implicit in the original RG theory and predicted by the ϕ^4 model, has also recently been demonstrated by numerical simulations in spin systems [15].

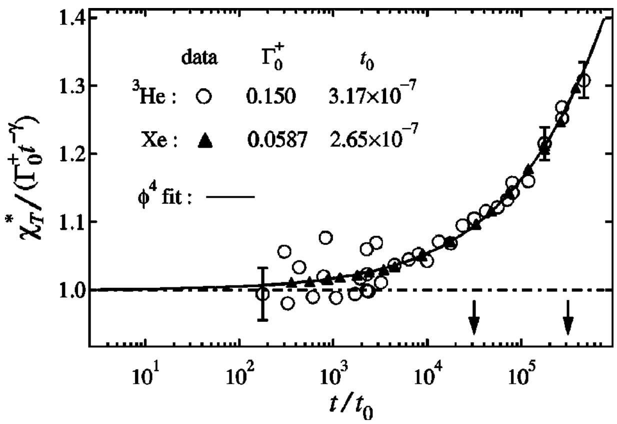

FIG. 2. Susceptibility χ_T^* scaled by $(\Gamma_0^+ t^{-\gamma})$ vs t scaled by t_0. The solid curve is calculated from the ϕ^4 model, Eqs. (3)–(15), and the symbols are data points. The dot-dashed line corresponds to a pure power law for $\chi_T^*(t)$. The error bar represents combined uncertainties from measured χ_T^* and t. Arrows indicate where $t = 10^{-1}$ and 10^{-2} would be for ^{3}He.

In characterizing the crossover regime, the Ginzburg number, G, is often used as an indicator for separating the region far away from the transition where mean-field theory is valid and the region close to the transition where fluctuations renormalize the critical behavior of the system. In Ref. [16], the Ginzburg number is linked to the first Wegner correction amplitude in the case of infinite cut-off wave number as

$$\Gamma_1^+ = \frac{1}{9} G^{-\Delta}. \tag{19}$$

The comparison of Eq. (19) to Eq. (18) leads to a linear relation between G and t_0 and yields $G(^3\text{He}) = 0.012 \pm 0.001$ and $G(\text{Xe}) = 0.010 \pm 0.002$. Even though t_0 is a function of u, fitting ^{3}He and Xe susceptibility data to the ϕ^4 model shows that the ratio of $t_0(\text{Xe})/t_0(^3\text{He})$ remains constant for any given $0 < u < u^*$. This implies that $G(\text{Xe})/G(^3\text{He}) = t_0(\text{Xe})/t_0(^3\text{He}) = 0.84 \pm 0.23$.

The RG-based crossover model with a minimal set of *three* adjustable parameters provides an excellent fit to the ^{3}He and Xe susceptibility data over more than three decades in reduced temperature outside the asymptotic regime. The applicable range of the RG-based crossover model was discussed by Bagnuls and Bervillier [10]. They defined a preasymptotic temperature region in which the ϕ^4 model is strictly valid. The upper bound of this region is defined by the validity of the Wegner expansion up to the first correction-to-scaling term, t^{Δ}. Beyond this range other corrections neglected in the ϕ^4 model could influence theoretical predictions. Using the values of Γ_0^+ and Γ_1^+ calculated from Eqs. (17) and (18), we obtain Wegner-expansion curves for ^{3}He and Xe that agree within 0.6% with the full ϕ^4 model calculation out to $t=10^{-2}$. Deviations between this Wegner expansion and the ϕ^4 model calculation increase for temperatures $t>10^{-2}$. Based on the arguments by Bagnuls and Bervillier given above, the preasymptotic regimes for ^{3}He and Xe are identified to be $t\leqslant 10^{-2}$, even though a good fit based on RG model is obtained out to $t\sim 10^{-1}$, as seen in Fig. 2. In the case of ^{3}He, additional corrections neglected in the ϕ^4 model could include quantum effects, which are not taken into account in the Hamiltonian, Eq. (1). Quantum effects are expected to become important when the correlation length is comparable to, or smaller than the de Broglie wavelength λ_T. In the case of ^{3}He, our calculation of the correlation length yields $\xi_0=2.75$ Å, which is consistent with the published value of 2.57 Å [17]. The calculated ξ is equal to λ_T near $t\sim 0.9$, thus, quantum effects are not expected to significantly affect the present measurements. In contrast, a recent comparison of earlier ^{3}He compressibility data to Monte Carlo calculations [18] suggests that quantum effects may play an important role in understanding crossover behavior of ^{3}He.

It should be noted that the RG-based expressions in the minimal-subtraction and massive-renormalization schemes were derived using an infinite cut-off wave number. The limitation of an infinite cut-off wave number has been discussed in Refs. [3,10]. In more general complex fluid systems, crossover models that include a finite cut-off wave number, in addition to the Ginzburg number, are necessary to describe crossover behavior [14,19,20]. We have also applied the ϕ^4 model to analyze other experimental measurements, such as the specific heat at constant volume [21] and correlation length.

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