

Universality and Finite-Size Scaling of the Specific Heat of ^3He - ^4He Mixtures.

Mark O. Kimball and Francis M. Gasparini*

University at Buffalo, The State University of New York, Buffalo, NY 14260-1500

(Dated: August 29, 2005)

We have measured the heat capacity of ^3He - ^4He mixtures confined in films of thickness 48.3 and 986.9 nm. The confinement is defined by direct bonding of two silicon wafers. The heat capacity is measured using an ac technique and then transformed to correct for exponent renormalization effects. The data address the expected universal critical behavior along the λ -line as function of ^3He concentration. We discuss the results of several analyses of the data, and we show that a universal collapse can be achieved for all the mixtures. However, this is on a locus which differs from that of the pure system. An alternative analysis is also presented which yields collapse of all the data under certain assumptions. We believe these data are the first to test universality of finite-size scaling for the specific heat along a locus of transitions.

PACS numbers: 64.70.-p, 67.60-g, 68.35.Rh

The superfluid transition of ^4He has been studied extensively as an example of a continuous phase transition. The behavior of the specific heat and superfluid density are well known in the thermodynamic limit. Critical exponents have been obtained experimentally and have been calculated to high precision [1–3]. Also, there have been a number of studies where helium is confined in a uniform geometry where the smallest spatial length L can become comparable to the temperature-dependent correlation length [4–6]. In this limit, according to correlation-length scaling, one expects that for various L 's one should obtain a collapse of data when plotted as a function of L/ξ where ξ is the bulk correlation length [7, 8]. Experiments have shown that for films, which crossover to two dimensions as the transition is approached, finite-size scaling works well in the region where the confined helium is normal but fails in the region where it becomes superfluid [4, 5]. The situation where the crossover dimension is not two, but one or zero has not been studied as extensively at this time.

With the addition of ^3He to ^4He , the bulk superfluid transition shifts to lower temperatures defining a locus at saturated vapor pressure referred to as the λ -line. The mixtures are expected to be in the same universality class as pure ^4He . Thus, the exponents are expected to be universal until one approaches the crossover region of the tricritical point at a ^3He concentration $x \cong 0.67$. Here the λ -line joins the first order phase separation lines. Also, studies along the analogous λ -line at $x = 0$, but as function of pressure, are expected to show a similar universal behavior. This universality implies for instance that the critical exponent of the specific heat, α , should remain unchanged. Interestingly, along either λ -line, one finds that the experimentally determined value of α becomes more negative as one moves away from saturated vapor pressure $P = P_{sat}$, and $x = 0$ [9–11].

The specific heat of confined mixtures reported here addresses the issue of universality in two different ways. The data at various concentrations are expected to obey

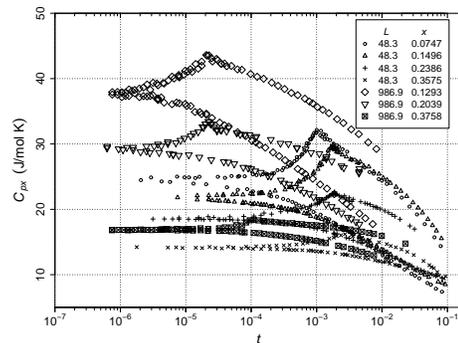


FIG. 1: C_{px} for the four mixtures measured in the 48.3 nm cell and the three mixtures confined to the 986.9 nm cell. These are plotted together to demonstrate the overall behavior of confined mixtures. The upper and lower branch for each mixture represents data below and above T_λ respectively.

finite-size scaling if one allows for the fact that the magnitude of the correlation length and the amplitude of the specific heat depend on concentration. In addition, the specific heat is sensitive to the universal value of α in a different way from that of the mixtures in the thermodynamic limit. We note that we have reported results for the superfluid density of confined mixtures at a single value of L which were also shown to scale on the same locus as for $x = 0$ [12]. However, the specific heat provides a much more sensitive way to test universality because α is very close to zero. For instance, a large change by as much as a factor of 2 in α would translate in a change of only $\sim 0.6\%$ in ν , the exponent governing the behavior of the superfluid density. This would have negligible effect in the finite-size scaling of the superfluid density, but, as we will see, it has a dramatic effect on the scaling of specific heat.

In this work we have used two experimental cells to study the specific heat of mixtures of ^3He and ^4He confined in a planar geometry between two wafers of sili-

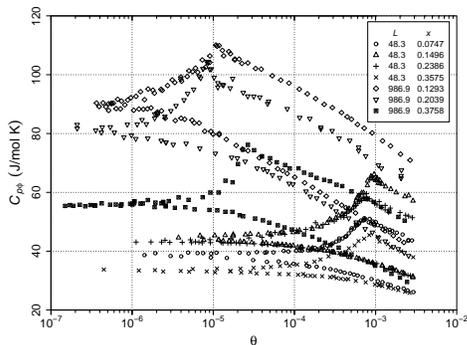


FIG. 2: $C_{p\phi}$ converted from the C_{px} data seen in Fig. 1 for the four mixtures measured in the 48.3 nm cell and the three mixtures confined within the 986.9 nm cell. Here, $\phi = \mu_3 - \mu_4$, the difference in the chemical potentials of ^3He and ^4He .

con. These cells are formed by first patterning the silicon dioxide grown on one of the wafers into a series of posts about 0.5 mm apart, and then using direct bonding of a second wafer to form a cell. The thickness of the oxide determines the small confinement L . The homogeneity of L can be determined for $L > \sim 500$ nm by infrared interference measurements. One finds that the 986.9 nm cell is uniform to better than 1% across the 5 cm wafers. The oxide pattern has a continuous 0.4 cm border which, upon bonding, forms a leak-tight seal. Further details about these cells can be found in previous publications [4, 13]. When the cell is filled with helium a small amount of liquid, $\sim 1 \text{ mm}^3$, collects in the filling line above the cell. This is used as a marker for T_λ of bulk helium. The cell has two doped-germanium thermometers epoxied directly on the silicon. One of these is used to regulate the average temperature while the other is used simultaneously to determine the amplitude of temperature oscillations. These oscillations are imposed by a uniform film heater which is driven at a fixed frequency, typically in the range of 25 to 45 Hz. The cell is connected to two temperature-regulated stages: one stage is where the filling line is attached, and where a valve is located; the other is a stage which maintains the cell colder than the filling line. The equations which govern the temperature response of the cell in this arrangement and the procedure for extracting the specific heat of the confined helium have been published [4, 13].

In Fig. 1 are shown the specific heat data, C_{px} , for 7 different ^3He concentrations in cells of two different L 's [14]. These are plotted as function of $t \equiv |1 - T/T_\lambda(x)|$ where $T_\lambda(x)$ is the transition of the bulk helium. The overall behavior of these data shows two effects. One is the decrease in specific heat as one goes from the large to the small L at fixed x . The other is the decrease in specific heat at fixed L as one increases x . To test the universality of correlation-length scaling, and collapse these data, one must recognize first that the true critical behav-

ior is not obtained in C_{px} but in $C_{p\phi}$, where $\phi = \mu_3 - \mu_4$, the difference in the chemical potentials [15, 16]. This requires a thermodynamic conversion of C_{px} , and a conversion of the distance to criticality from a path of constant x , where $t = |1 - T/T_\lambda(x)|$, to a path of constant ϕ , where $\theta \equiv |1 - T/T_\lambda(\phi)|$ is the relevant variable [9]. These conversions can be done by using thermodynamic derivatives along the λ -line $\left(\frac{\partial x}{\partial T}\right)_\lambda, \left(\frac{\partial \phi}{\partial T}\right)_\lambda, \left(\frac{\partial s}{\partial T}\right)_\lambda$, where s is the entropy per mole [9, 17]. We have used values tabulated in Ref. [9] and fit these to a function so that one can extract the values which correspond to the concentrations of our data. Ideally, one should use values constructed for fixed L , however, no such data are available. $C_{p\phi}$ can be related to C_{px} as follows: [9, 17]

$$C_{p\phi} = \frac{C_{px} \left(T \left. \frac{\partial x}{\partial T} \right|_{p,t} \left. \frac{\partial \phi}{\partial T} \right|_{p,t} - T \left. \frac{\partial s}{\partial T} \right|_{p,t} \right) + \left(T \left. \frac{\partial s}{\partial T} \right|_{p,t} \right)^2}{T \left. \frac{\partial s}{\partial T} \right|_{p,t} + T \left. \frac{\partial x}{\partial T} \right|_{p,t} \left. \frac{\partial \phi}{\partial T} \right|_{p,t} - C_{px}}, \quad (1)$$

where in practice the derivatives can be taken at $t = 0$, i.e. along the λ -line. The calculation of θ involves an iterative procedure which converges rapidly [9]. The result of our calculation is shown in Fig. 2 where $C_{p\phi}$ is plotted as function of θ . Notice that relative to C_{px} , these data have a larger magnitude and have a smaller θ than the equivalent t . Note that the amplitude of $C_{p\phi}$ is not a monotonic function of concentration unlike C_{px} . There is no reason *a priori* to expect it should be monotonic and this has also been seen in previous analyses of bulk mixtures [9, 18]. To test correlation-length scaling one can use an equation which is a modification of the equation used for $x=0$ [19],

$$\Delta C \theta^{\alpha_\phi} \left| \frac{\alpha_\phi}{A_\phi(x)} \right| = g_2 \left(\left[\frac{L}{\xi(x,t)} \right]^{1/\nu} \right), \quad (2)$$

Here, ΔC is the difference between the bulk and confined specific heats defined as $[C_{p\phi}(\theta, \infty) - C_{p\phi}(\theta, L)]$ and A_ϕ and α_ϕ are the specific heat amplitude and critical exponent of $C_{p\phi}(\theta, \infty)$. This equation differs in several respects from that used for pure ^4He . In the latter case one does not need the factor α/A , since it is always the same at $x = 0$ for any L . For the mixtures, however, A_ϕ is not universal and one must take this into account. The exponent α_ϕ , on the other hand, is expected to be universal along the λ -line; however, we leave this explicitly in Eq. 2. On the right hand side we indicate that for each concentration $\xi(x,t)$ will differ [20]. Thus the scaling variable, which for pure ^4He with $\xi_0 = \xi_0(0)$ can be taken as $t(L/\xi_0)^{1/\nu}$, must now allow for $\xi_0 = \xi_0(x)$. To calculate $\xi_0(x)$ we note that the correlation length below T_λ is related to the superfluid density [21, 22] and one may write for ξ_0

$$\xi_0(x) = \xi_0(0) \frac{T_\lambda(x)}{T_\lambda(0)} \frac{\rho(0)}{\rho(x)} \frac{k(0)}{k(x)}, \quad (3)$$

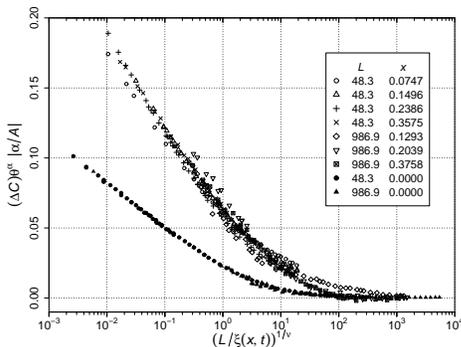


FIG. 3: Data for $T > T_{\lambda}$ scaled according to equation 2. The data for pure ^4He (solid symbols) are plotted using $\alpha = -0.0115$ while the mixture data (open symbols) use an α close to -0.025 : see text.

with ρ and k being respectively the density and the amplitude of the leading temperature dependence of the superfluid fraction. We use $\xi_0(0) = 1.42$ nm [23] and vary the value for $x \neq 0$ according to Eq. 3. We have used the data from Ref. [24] for the behavior of $k(x)$.

To implement Eq. 2 one needs $C_{p\phi}(\theta, \infty)$. One could proceed in two ways. One could take existing calculations of $C_{p\phi}$ [9, 18] and interpolate these to the concentrations at which we have measured $C_{px}(t, L)$. Alternatively, one could go to the primary data C_{px} and, from a suitable representation of these, interpolate to the concentrations of interest. Then one can construct $C_{p\phi}(\theta, \infty)$ and θ as outlined above. We have chosen the second route since it gives us a direct check on our calculation of $C_{p\phi}(\theta, L)$. This also allows us to explore the role of λ -line parameters simultaneously for $C_{p\phi}(\theta, \infty)$ and $C_{p\phi}(\theta, L)$. Another advantage of this procedure is that possible systematic errors in the calculation, such as arising from the λ -line parameters, become second order when one calculates the difference ΔC which is needed for scaling. When the constructed $C_{p\phi}(\theta, \infty)$ is fitted to the function

$$C_{p,\phi}(\theta, \infty) = \frac{A_{\phi}}{\alpha_{\phi}} \theta^{-\alpha_{\phi}} (1 + D_{\phi} \theta^{0.5}) + B_{\phi}, \quad (4)$$

the results for the various parameters, A_{ϕ} , α_{ϕ} , D_{ϕ} , B_{ϕ} , fit smoothly as a function of x within the parameters obtained with the original experimental data. This effectively checks on the correctness of our calculation of $C_{p\phi}(\theta, \infty)$. In particular, one finds, as previously, that the best exponent that describes this specific heat is $\alpha \cong -0.025$ with minor variations as function of concentration. This contrasts with $\alpha \cong -0.012$ for the data at $x = 0$.

In this analysis we have limited ourselves to data for which $\theta < 3 \times 10^{-3}$ to avoid higher order corrections in Eq. 4. With these results one can now calculate the left hand side of Eq. 2 and see if one achieves universal collapse. This is shown in Fig. 3 for $T > T_{\lambda}$. For this plot

we have used for each mixture whatever set of α_{ϕ} , A_{ϕ} were obtained from the analysis of $C_{p\phi}(\theta, \infty)$ and the corresponding ν_{ϕ} which can be obtained from the hyper-scaling relation, $\alpha_{\phi} = 2 - 3\nu_{\phi}$. It is clear from this plot that the seven mixtures for both values of confinement L do indeed fall on a universal locus, while the data for $x = 0$ are at a quite distinct locus. The same is true for the data for $T < T_{\lambda}$.

From the analysis leading to Fig. 3 it is easy to ascertain that variations in the value of ν_{ϕ} coming from α_{ϕ} are not important. A very similar plot would be obtained were one to take a fixed $\nu_{\phi} = 0.6705$ as has been used to scale data for $x = 0$ [5]. Further, the effect on the scaling plot of the term $\theta^{\alpha_{\phi}}$ is also not very important (although not negligible), i.e. α_{ϕ} is very close to zero in all cases. The major determinant in the lack of universal collapse between the mixtures and the pure system is in the factor α_{ϕ}/A_{ϕ} . In this respect, finite-size scaling along the λ -line, and the expected universality, is more demanding and hence more sensitive to the data than the equivalent expectation for the mixtures in the thermodynamic limit. In the latter case one wants universality in α_{ϕ} and amplitude ratios. With the confined system, the expected universal scaling function has both the exponent and the non-universal amplitude A_{ϕ} as multiplicative factors.

To explore further this problem between $x = 0$ and $x \neq 0$, we have tried a variety of other analyses. We have fixed the exponent in the range of -0.0115 to -0.025 and fitted all bulk data to extract the amplitudes A_{ϕ} 's which correspond to each fixed power law. This, of course compromises the goodness of the fits, but nevertheless yields a set of amplitudes which correspond to a universal exponent. The result of this analysis is scaling plots which in one extreme, $\alpha = -0.0115$, neither the mixtures nor the pure system fall on a universal locus. At the other extreme, $\alpha = -0.025$, the mixtures collapse on a separate locus from $x = 0$. In this latter case a universal collapse can be achieved if one introduces a numerical factor for $\xi_0(0)$ which is not justified on the basis of Eq. 3.

Yet another possible approach is to assume that α_{ϕ} for the mixtures is not the true asymptotic exponent. Thus, one takes α_{ϕ} as universal at the value obtained by the pure system. However, the amplitudes A_{ϕ} are retained as the "best fit" values. This yields the scaling plot shown in Fig. 4. This represents a reasonable collapse of all the data on a universal curve with the same universal exponent. Note that this plot tests two ideas: finite-size scaling with the two different confinements; and universality with the 7 mixtures. The equivalent plot for $T < T_{\lambda}$ is shown in Fig. 5. Here, the collapse is not as good as for $T > T_{\lambda}$, but probably satisfactory. Note that in the region of the specific heat maximum (shown as a minimum in Fig. 5) there is a lack of scaling which is also seen in the pure system [5].

These last two figures might be taken as evidence of universality of finite-size scaling along the λ -line with

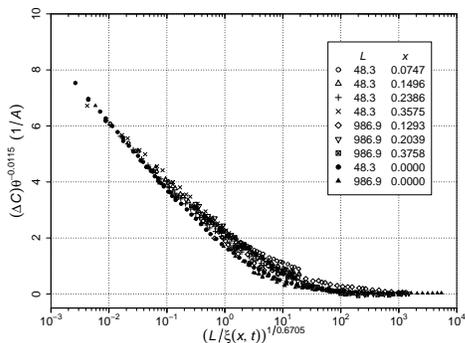


FIG. 4: Data for $T > T_\lambda$ plotted according to equation 2. Contrary to figure 3, all data are plotted using $\alpha = -0.0115$ but with A_ϕ retained at its “best fit” value: see text.

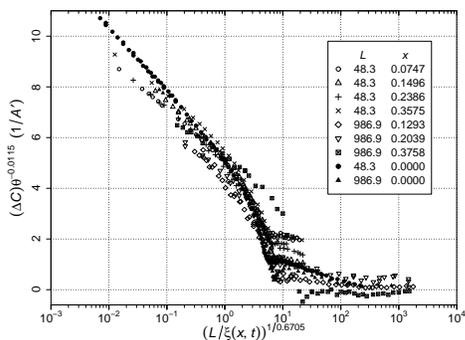


FIG. 5: Data for $T < T_\lambda$ plotted according to equation 2. All data are plotted using $\alpha = -0.0115$.

mixtures. However, one should be aware that the use of amplitudes A_ϕ 's which do not correspond to the exponent $\alpha = -0.0115$ that is used in this plot is not a self-consistent analysis. The alternative to this is Fig. 3 with the less palatable conclusion that the point at $x = 0$ is somehow special on the phase diagram. This is not supported on any theoretical basis [25].

In conclusion, we find that for $x \neq 0$, we obtain a collapse of all the data on a universal locus. This involves seven different mixtures in two different confinements differing by a factor of ~ 20 . This must be viewed as a significant result supporting universality and finite-size scaling along the λ -line. However, our analysis also shows that the data at $x = 0$ cannot be made to collapse on the mixtures in a self-consistent way. This is due to the fact that the mixtures and the pure system do not yield the same critical exponent in the bulk. This is an issue which possibly will be resolved by new bulk measurements.

edu; This work is supported by the NSF, DMR-9972285, and DMR-0242246; and by the Cornell NanoScale Science & Technology Facility, 526-94

- [1] L. S. Goldner, N. Mulders, and G. Ahlers, *J. Low Temp. Phys.* **93**, 125 (1993).
- [2] M. Campostrini, A. Pelissetto, P. Rossi, and E. Vicari, *Phys. Rev. B* **61**, 5905 (2000).
- [3] J. A. Lipa, J. A. Nissen, D. A. Stricker, D. R. Swanson, and T. C. P. Chui, *Phys. Rev. B* **68**, 174518 (2003).
- [4] S. Mehta, M. O. Kimball, and F. M. Gasparini, *J. Low Temp. Phys.* **114**, 467 (1999).
- [5] M. O. Kimball, S. Mehta, and F. M. Gasparini, *J. Low Temp. Phys.* **121**, 29 (2000).
- [6] J. A. Lipa, D. R. Swanson, J. A. Nissen, Z. K. Geng, P. R. Williamson, D. A. Stricker, T. C. P. Chui, U. E. Israelsson, and M. Larson, *Phys. Rev. Lett.* **84**, 4894 (2000).
- [7] M. E. Fisher, in *Critical Phenomenon, Proc. 51st “Enrico Fermi” Summer School, Varenna, Italy*, edited by M. Green (Academic Press, NY, 1971).
- [8] M. E. Fisher and M. N. Barber, *Phys. Rev. Lett.* **28**, 1516 (1972).
- [9] F. M. Gasparini and M. Moldover, *Phys. Rev. B* **12**, 93 (1975).
- [10] K. H. Mueller, F. Pobell, and G. Ahlers, *Phys. Rev. Lett.* **34**, 513 (1975).
- [11] F. M. Gasparini and A. A. Gaeta, *Phys. Rev. B* **17**, 1466 (1978).
- [12] M. O. Kimball and F. M. Gasparini, *Phys. Rev. Lett.* **86**, 1558 (2001).
- [13] S. Mehta and F. M. Gasparini, *J. Low Temp. Phys.* **110**, 287 (1998).
- [14] Strictly speaking we measure the heat capacity along saturated vapor pressure, not constant pressure. However, for our purposes, this distinction is negligible. Data for $x = 0$ can be found in Refs. [4, 5].
- [15] M. E. Fisher, *Phys. Rev.* **176**, 257 (1968).
- [16] B. J. Lipa and M. J. Buckingham, *Phys. Lett. A* **26**, 643 (1968).
- [17] G. Ahlers, *Phys. Rev. A* **10**, 1670 (1974).
- [18] T. Takada and T. Watanabe, *J. Low Temp. Phys.* **41**, 221 (1980).
- [19] One can derive this relationship by starting with a free energy in the form $F(\theta, L) = t^{2-\alpha} f(\theta(L/\xi)^{1/\nu}) + f_0(\theta)$. Differentiating twice one obtains $C(\theta, L)$. Matching with a power law form for $C(\theta, \infty)$ leads to Eq. 1.
- [20] One could also write $\xi(x, \theta)$ instead of $\xi(x, t)$. However, the latter is more convenient since it can be directly related to the superfluid density which is measured as a function of t .
- [21] R. A. Ferrell, N. Menyhard, H. Schmidt, F. Schwabl, and P. Szépfalusy, *Ann. Phys. (N.Y.)* **47**, 565 (1968).
- [22] B. I. Halperin and P. C. Hohenberg, *Phys. Rev.* **177**, 952 (1969).
- [23] R. Haussmann and V. Dohm, *Phys. Rev. B* **46**, 6361 (1992).
- [24] P. C. Schubert and W. Zimmermann, Jr., *J. Low Temp. Phys.* **41**, 177 (1981).
- [25] M. E. Fisher, private communication (2002).

* fmg@buffalo.edu; <http://enthalpy.physics.buffalo>.