Differential Osmotic Pressure Measurements of the Concentration Susceptibility of Liquid $^3He/^4He$ Mixtures near the Lambda Curve and Tricritical Point

C. A. Gearhart, Jr.* and W. Zimmermann, Jr.

School of Physics and Astronomy, University of Minnesota

Minneapolis, Minnesota 55455

February 1976

Prepared For

The U.S. Energy Research and Development Administration

Under Contract EY-76-S-02-1569

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ABSTRACT

Values of the concentration susceptibility $(\partial x/\partial\Delta)_{T,P}$ of liquid $^3He/^4He$ mixtures have been determined near the lambda line and tricritical point from measurements of the differential osmotic pressure as a function of temperature T at four values of the ^3He mole fraction, $x = 0.594$, $x = 0.644$, $x = 0.680$, and $x = 0.706$. Here $\Delta = \mu_3 - \mu_4$ is the difference between molar chemical potentials and P is the pressure. In contrast to other determinations, our results for the two values of x less than the tricritical value $x_t = 0.675$ show pronounced peaks at the lambda transition. For $3 \times 10^{-4} \lesssim |\varepsilon| = |T - T_\lambda(x)|/T_\lambda(x) \lesssim 10^{-2}$ these peaks may be characterized above and below the transition by the form $(A/\alpha)\,(|\varepsilon|^{-\alpha} - 1) + B$ with exponents α lying in the range from ~ 0.0 to ~ 0.2. Except perhaps for $x < x_t$ in the normal-fluid region away from the transition, our data appear to be consistent with a simple tricritical scaling relationship of the form

$$\left(\frac{\partial x}{\partial \Delta}\right)^{-1}_{T,P} = f(x)\,\Xi\left[\left(\frac{T-T_t}{T_t}\right) \Big/ \left|\frac{x-x_t}{x_t}\right|\right],$$

where f and Ξ are functions determined by experiment and $T_t = 0.867$ K is the tricritical value of T.

The current interest in tricritical points[1] has been stimulated by and has itself encouraged a number of experiments exploring the thermodynamic behavior of liquid $^3He/^4He$ mixtures in the tricritical region. For several reasons, including the ease of obtaining pure and homogeneous samples, these mixtures provide a very favorable system for the study of tricritical behavior. The experiments include capacitance[2] and optical[3] measurements of the density along the two branches of the coexistence curve, measurements of the molar specific heat $c_{x,P}$ at constant ^3He mole fraction x and pressure P,[4,5] second-sound measurements of the superfluid density ρ_S,[6] and determinations of the concentration susceptibility $(\partial x/\partial\Delta)_{T,P}$ by means of vapor pressure[7,8] and light-scattering[3,9] measurements. Here Δ is the difference $\mu_3-\mu_4$ between molar chemical potentials and T is the absolute temperature. The quantity $(\partial x/\partial\Delta)_{T,P}$, analogous in T,$\Delta$ space (at constant P) to the compressibility of a pure fluid in T,P space, is of particular importance because of the information it contains about the relation between T, x, and Δ. We report here determinations of $(\partial x/\partial\Delta)_{T,P}$ from measurements of the differential osmotic pressure.[10] We have been especially interested in the lambda transition, where, contrary to the vapor-pressure and light-scattering results, our data show a pronounced peak.

A schematic drawing of the experimental cell is shown in Fig. 1. The cell consists of two chambers which are maintained at the same temperature and which are completely filled with $^3He/^4He$ mixtures differing in composition by a small amount Δx. The chambers are connected by a Vycor glass superleak,[11] which, when a superfluid connection exists through its pores, permits the molar chemical potential μ_4 of the ^4He component to reach equilibrium between the two chambers. We measure the resulting osmotic pressure difference ΔP

between the chambers by means of a flexible stainless-steel diaphragm separating them. The deflection of this diaphragm is measured capacitively, the capacitor forming part of the tank circuit of a low-temperature back-diode oscillator.[12]

A remarkable feature of the experiment is that, despite the small size of the pores, at values of $x \gtrsim 0.55$ the superleak functions not only in the superfluid region of the phase diagram for the bulk fluid but also in a small portion of the normal-fluid region adjacent to the lambda and coexistence curves, thus permitting us to make measurements on both the superfluid and normal-fluid sides of the tricritical point. We believe this phenomenon to be due to the formation of a superfluid ^{4}He-rich film covering the walls of the pores with which the ^{3}He/^{4}He mixture is in contact; the effects of such films have been seen in a number of other experiments.[13] The occurrence of this phenomenon in the vicinity of the tricritical point and the extent of the region of the phase diagram in which we observe it were reported by us in an earlier publication.[14]

Measurements were made at four different values of the average ^{3}He mole fraction in the chambers, $x = 0.594$, $x = 0.644$, $x = 0.680$, and $x = 0.706$. At each value, ΔP was measured as a function of T for several different values of Δx ranging in magnitude from 0 to 4×10^{-4}, a value of 1×10^{-4} being representative of those with which our best results were obtained. Data were recorded with a cell temperature stability of several microkelvins at intervals down to 20 μK. The resolution of our pressure measurements, limited by the long-term stability of the pressure-measuring system, was $\sim \pm 0.02$ Pa.

The concentration susceptibility as a function of T was obtained from P(T) by means of the relationship

$$\left(\frac{\partial x}{\partial \Delta}\right)_{T,P} = \frac{x}{v_4}\left(\frac{\partial x}{\partial P}\right)_{T,\mu_4} \cong \frac{x}{v_4}\,\frac{\Delta x}{\Delta P} \quad , \qquad (1)$$

where $v_4 = v - x\,(\partial v/\partial x)_{T,P}$ and where v, in turn, is the molar volume. Because we were not able to make direct determinations of Δx, it was necessary to normalize our results for $(\partial x/\partial \Delta)_{T,P}$ to outside data at one value of T for each value of x. For this purpose we used the results of the vapor pressure measurements of Goellner, Behringer, and Meyer (GBM).[7]

Our results for $(\partial x/\partial \Delta)_{T,P}$ are shown in Fig. 2. Of particular interest is the prominent peak which occurs at the lambda transition for the two lowest values of x. Such a peak is entirely missing at these concentrations in the susceptibilities derived from vapor pressure measurements,[7] and only a minor maximum in susceptibility at T_λ was observed at x = 0.632 in the light-scattering results.[9]

We have attempted to fit the T dependence of $(\partial x/\partial \Delta)_{T,P}$ at constant x near the lambda transition to the simple power-law form

$$(\partial x/\partial \Delta)_{T,P} = (A/\alpha)(|\varepsilon|^{-\alpha} - 1) + B \quad , \qquad (2)$$

where $\varepsilon \equiv [T - T_\lambda(x)]/T_\lambda(x)$ and where, in turn, $T_\lambda(x)$ is the lambda temperature at the particular value of x in question. Successful fits were obtained for data in the range $2 \times 10^{-4} \leq |\varepsilon| \leq 10^{-2}$ for x = 0.594 and $4 \times 10^{-4} \lesssim |\varepsilon| \lesssim 10^{-2}$ for x = 0.644; when data at smaller values of $|\varepsilon|$ were used, the quality of fit decreased considerably. We believe that the data at smaller values of $|\varepsilon|$ were subject to serious distortion as the result of gravitational inhomogeneities in x within each chamber and of finite Δx between chambers. Considerable latitude was present in the fits, with values for α both above and below the transition ranging from ~ 0.0 to ~ 0.2. Except perhaps for $T > T_\lambda$ at $x = 0.644$, where $\alpha > 0$ seemed to be favored, it was possible to obtain acceptable fits with $\alpha = 0$ both above and below the transition.

The behavior of $(\partial x/\partial \Delta)_{T,P}$ at the lambda curve is closely related to that of the specific heat $c_{\Delta,P}$. Near-zero values have been obtained for the exponents describing the temperature behavior of c_P for pure ^{4}He and of $c_{\Delta,P}$ for mixtures in the range $0 < x \leq 0.53$ along paths of constant Δ.[15,16] Although a proper comparison should involve the behavior of $(\partial x/\partial \Delta)_{T,P}$ along paths of constant Δ rather than constant x, peaks in $(\partial x/\partial \Delta)_{T,P}$ with near-zero exponents would be consistent with these specific-heat results when combined with the hypothesis that the exponents are universal along the lambda curve. An exponent for $(\partial x/\partial \Delta)_{T,P}$ significantly greater than zero for $T > T_\lambda$ at $x = 0.644$ might reflect the presence of crossover effects in our data there, rather than a departure from the above consistency.[8,17]

Riedel, Meyer, and Behringer (RMB) have proposed a simple tricritical scaling relationship for $(\partial x/\partial \Delta)_{T,P}^{-1}$ and find that the results from vapor pressure measurements appear to satisfy this relationship within a certain "scaling region" around the tricritical point.[8] Their relationship is of the form

$$\left(\frac{\partial x}{\partial \Delta}\right)^{-1}_{T,P} = f(x)\,\Xi\left[\left(\frac{T-T_t}{T_t}\right)\Big/\left|\frac{x-x_t}{x_t}\right|^{1/\omega_u}\right] , \qquad (3)$$

where x_t and T_t are the tricritical values of x and T and Ξ is a function with two branches, one for $x < x_t$ and one for $x > x_t$, determined by experiment. The exponent ω_u is found experimentally to be 1.00. Their function f(x) equals

$|(x-x_t)/x_t|^{\delta_{n,t}-1}$, where $\delta_{n,t}$ is found experimentally to be 2.05. We have tested our results against this relationship by plotting $\Xi = (\partial x/\partial\Delta)_{T,P}^{-1}/|(x-x_t)/x_t|^{1.05}$ versus $z = [(T-T_t)/T_t]/|(x-x_t)/x_t|$. A major portion of our results for $x < x_t$ are shown in Fig. 3 together with the vapor pressure results of GBM and RMB that lie in the RMB scaling region[7,8,18] and the light-scattering results of Watts and Webb (WW) at $x = 0.632$,[9] all assuming $x_t = 0.675$ and $T_t = 0.867$ K.[4,6,9]

It is interesting to note that in the superfluid region and in the normal-fluid region immediately adjacent to the lambda curve, our data seem to obey this scaling relation, even though the form of $\Xi(z)$ that we obtain near the lambda transition is quite different from that of RMB.[8] It should be emphasized that, because of the normalization of our data to those of GBM, our results do not provide an independent test of the form of f(x). Indeed, for the purpose of enhancing the coincidence of our data for different x near the lambda transition, the normalization of the susceptibilities for $x = 0.594$ used in Fig. 3 was increased by a few percent relative to that used in Fig. 2. Figure 3 shows that in the normal-fluid region away from the transition our data for $x < x_t$ deviate from scaling, although our data for $x = 0.594$ at the highest temperatures are open to some question. Our results in the normal-fluid region for $x > x_t$ appear to be roughly consistent with the RMB scaling relationship. In this connection, attention is called to the discovery that the RMB scaling relationship implies a scaling form for the specific heat $c_{x,P}$ which appears to be satisfied in the superfluid but not in the normal-fluid region.[19]

We wish to acknowledge the support given this work by the U.S. Energy Research and Development Administration.[20]

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* Present address: Department of Physics, Concordia College, Moorhead, Minnesota 56560.

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FIGURE CAPTIONS

Fig. 1. The experimental cell.

Fig. 2. The concentration susceptibility versus temperature at four values of ^{3}He mole fraction. The double-circles mark the points at which the results are normalized to those of GBM.[7]

Fig. 3. A plot of our results at x = 0.594 and x = 0.644, those of GBM and RMB which lie in their scaling region for $x < x_t$,[7,8,18] and those of WW at x = 0.632,[9] in the scaling form of RMB. We have assumed T_t = 0.867 K and x_t = 0.675.

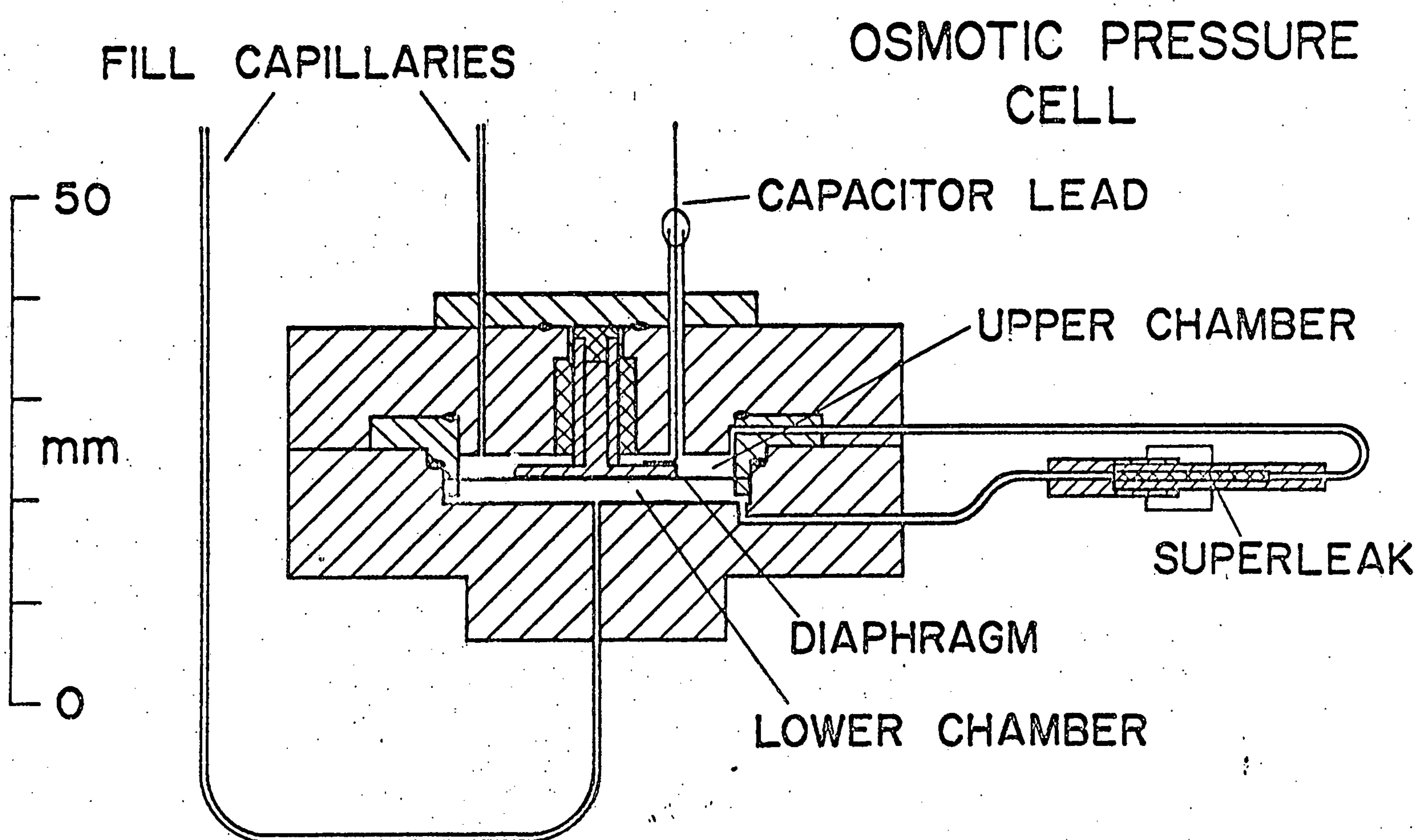
OSMOTIC PRESSURE
CELL
FILL CAPILLARIES
CAPACITOR LEAD
UPPER CHAMBER
SUPERLEAK
DIAPHRAGM
LOWER CHAMBER
50
mm
0

Figure 1.

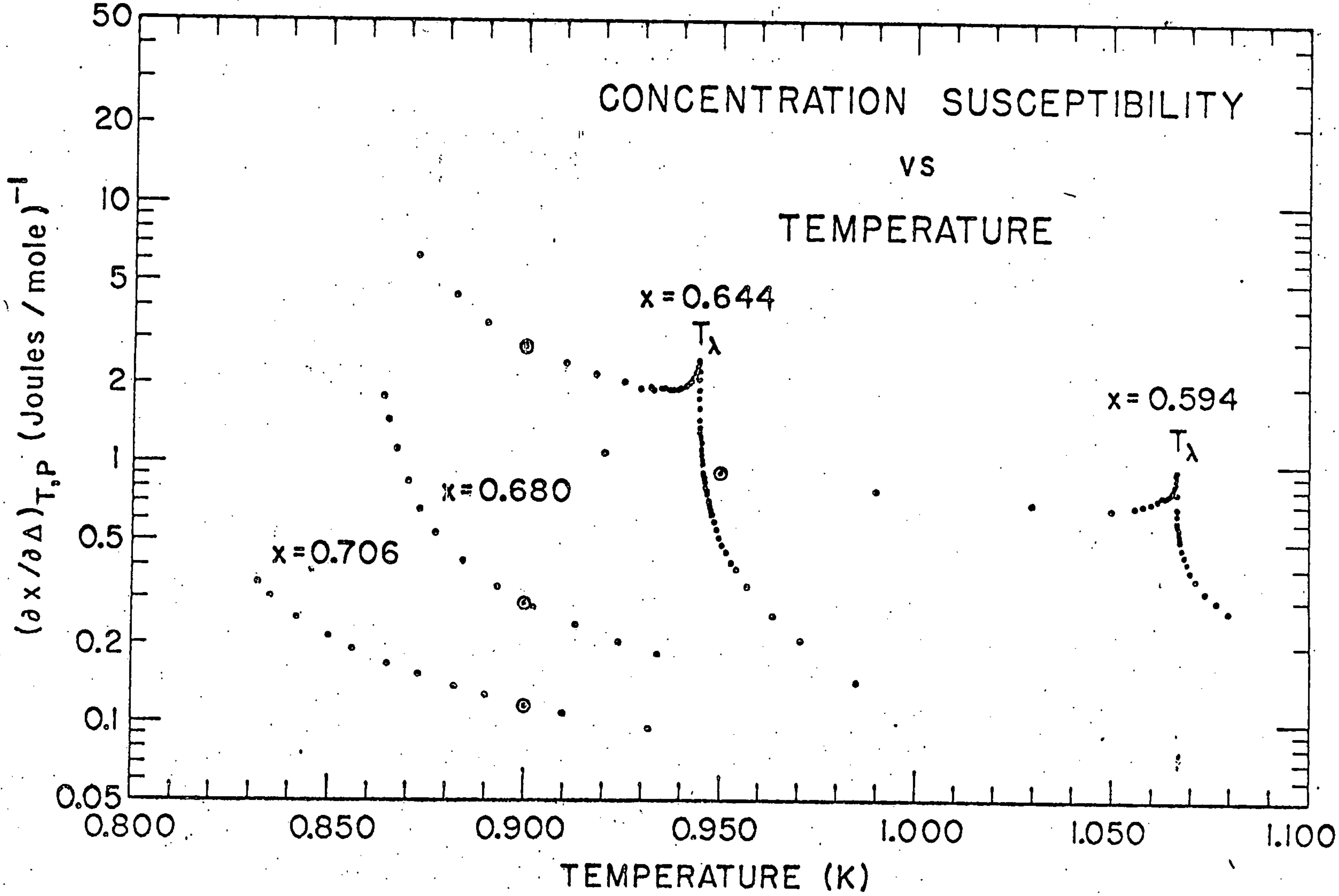
CONCENTRATION SUSCEPTIBILITY
vs
TEMPERATURE
x = 0.644
T_λ
x = 0.594
T_λ
x = 0.680
x = 0.706
$(\partial x/\partial \Delta)_{T,P}$ (Joules / mole)$^{-1}$
50
20
10
5
2
1
0.5
0.2
0.1
0.05
0.800
0.850
0.900
0.950
1.000
1.050
1.100
TEMPERATURE (K)

Figure 2.

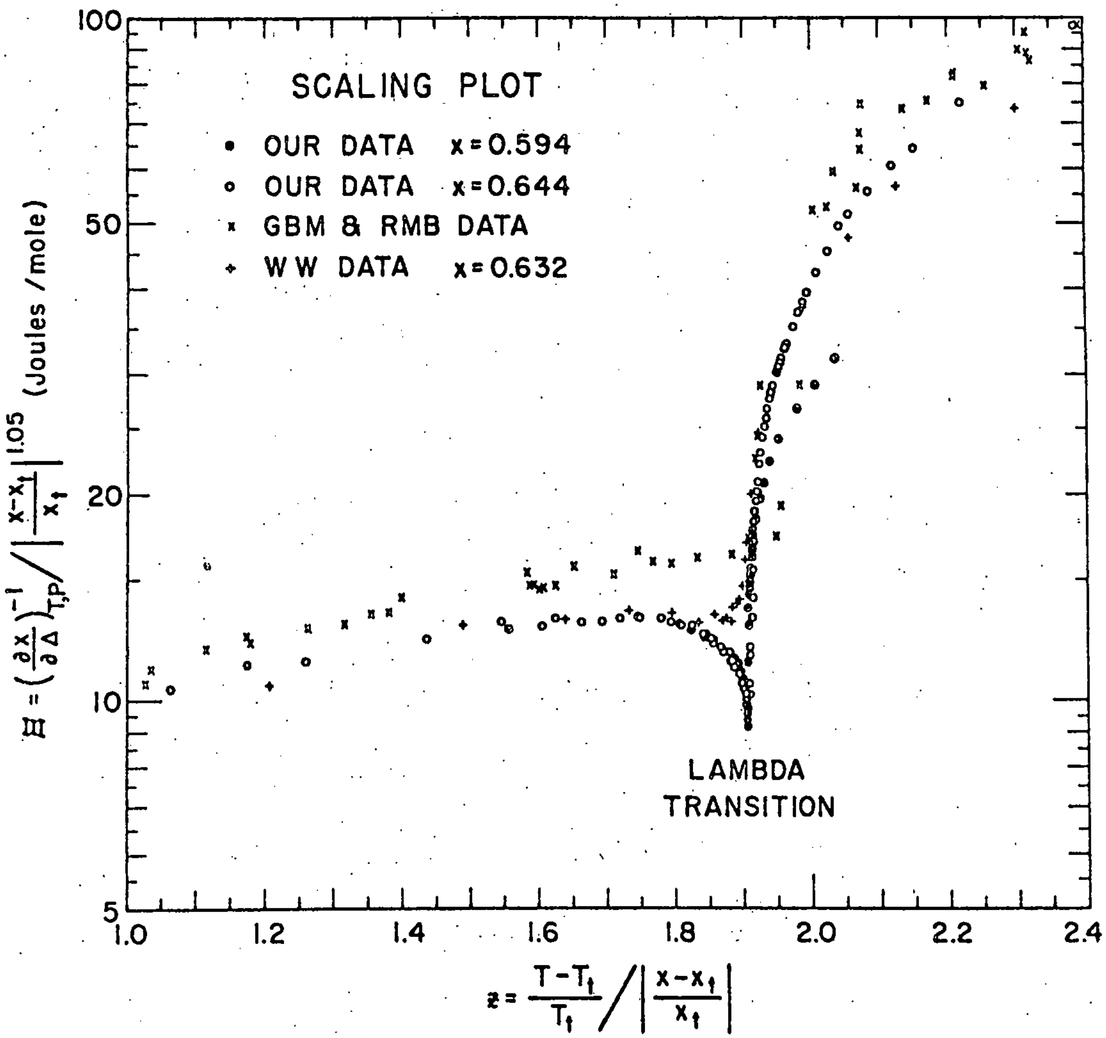
SCALING PLOT
OUR DATA x=0.594
OUR DATA x=0.644
GBM & RMB DATA
WW DATA x=0.632
LAMBDA TRANSITION
100
50
20
10
5
1.0
1.2
1.4
1.6
1.8
2.0
2.2
2.4
(Joules /mole)

Figure 3.