Heat Capacity and Thermal Relaxation of Bulk Helium very near the Lambda Point

J. A. Lipa, D. R. Swanson, and J. A. Nissen

Department of Physics, Stanford University, Stanford, California 94305

T. C. P. Chui and U. E. Israelsson

Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California 91109

(Received 18 September 1995)

We report new high-resolution measurements of the heat capacity of liquid helium to within 2 nK from the lambda transition, performed with a large sample in earth orbit. The optimum value for the critical exponent characterizing the divergence of the heat capacity below the transition was found to be -0.01285, giving improved support for the renormalization-group theory of phase transitions. Some information on the temperature dependence of the thermal conductivity just above the transition was also obtained.

PACS numbers: 67.40.Kh, 65.20.+w

One of the most basic quantities of interest near a critical phase transition is the temperature dependence of the heat capacity. Experimentally, this is an ideal property to study because of its accessibility and relative freedom from artifacts. From the theoretical perspective, it is well defined and calculable at least approximately for virtually any condensed-matter model containing a critical point. Since Onsager's exact solution [1] of the two-dimensional Ising model, which exhibited a symmetric logarithmic behavior of the heat capacity, there has been a great deal of interest in the detailed comparison of real system behavior with the models. For the lambda transition of helium a very nearly logarithmic singularity was measured [2], while an apparently similar behavior was seen [3] for the gas-liquid critical point. As the experimental work become more precise, the logarithmic asymptotic form was ruled out, first for xenon [4], for the gas-liquid singularity, and then for the lambda point [5], at least on the low temperature side. The heat capacity singularities now appear to be adequately represented by the simple asymptotic form $C_p = At^{-\alpha}(1 + Dt^{\Delta}) + B$, where $t = |1 - T/T_c|$ and T_c is the transition temperature [6]. Near the gas-liquid critical point the leading exponent α was found [7] to be close to 0.1, while at the lambda point a small negative value was obtained [8] indicating a finite heat capacity maximum. Among other things, these experiments showed that logarithmic singularities in nature must be the exception rather than the rule.

Our understanding of cooperative transitions was significantly advanced with the development [9] of the renormalization-group (RG) approach to the fluctuation problem. The RG theory has not only explained a wide range of experimental results in critical phenomena, but the concept has opened up the possibility of solving a significant number of problems that are among the most difficult in theoretical physics. Successful applications of the RG approach range from statistical physics to elementary particle physics. While these successes are considerable, it should be noted that the quantitative experimental sup-

port for the theory rests primarily on its predictions for critical phase transitions. Of all the results to date, there are only a few testable predictions in this field that can be considered exact. The most significant predictions are the relationships between groups of exponents, and the universality (or system independence) of certain key parameters. For example, the exponent α is expected to be the same for all systems falling within a given class, defined by the spatial dimensionality d of the system and the number of degrees of freedom n of its order parameter. Additional predictions of the theory are generally based on perturbation-expansion techniques or other approximation schemes. These methods have shown how the exponents depend on *n* and *d*, explaining the differences between the critical and lambda points. Also they have given rise to estimates [10,11] of the exponents characterizing the asymptotic singular behavior of various thermodynamic quantities in particular systems.

To test this theory, one clearly needs the most accurate exponent values possible, for comparison with both the detailed and broad predictions. Here the lambda transition is of great utility because of the weakness of the singularity in the compressibility of the fluid, the purity of the sample, and recent developments in thermometry. This situation allows the various exponents to be measured with exceptionally high accuracy. Key tests currently focus on the values obtained for the exponents ζ , characterizing the singularity of the superfluid density, and α . In this paper we report the results of a new heat capacity experiment performed in space to minimize gravity-induced broadening of the transition, and compare the observations with both predictions and earlier measurements. The experiment has led to a reduction by about a factor of 6 in the uncertainty of the exponent α at the lambda point of helium. We conclude that the new results lend further support to the RG theory, with the level of testing now limited by the accuracy of the theoretical calculations. We also briefly report on some associated thermal relaxation data which is of relevance to dynamic RG theory.

Our experiment was flown on the Space Shuttle in October 1992 as part of the USMP-1 payload. The primary objective of the mission was to measure the heat capacity below the transition with the smearing effect of gravity removed and derive an improved value for α . Secondary goals were to collect heat capacity and thermal relaxation data on the high-temperature side as time permitted. Because of noise problems due to cosmic-ray impacts, almost all the mission was devoted to the primary goal. The sample was a sphere of helium 3.5 cm in diameter contained within a copper calorimeter of very high thermal conductivity [12]. The calorimeter was attached to a pair of high-resolution paramagnetic-salt thermometers (HRTs) [13] with noise levels in the 10^{-10} K range, and suspended from a multistage high-stability thermal isolation system. The basic design of this system was similar in concept to that [14] used for earlier ground heat capacity measurements [15] of samples with small vertical height, but was ruggedized to survive launch. It was housed within the Low-Temperature Research Facility [16], a helium dewar and associated hardware developed for Shuttle experiments by the Jet Propulsion Laboratory (JPL). The heat capacity was measured by the heat-pulse method to within about 2 nK of the lambda point, and the thermal relaxation to within 3 nK. Particle radiation was found to inject clearly detectable thermal spikes into the sensing elements of the HRTs, resulting in a significantly higher noise figure in flight compared with ground-based experiments. This effect was the primary factor limiting the accuracy of the heat capacity results.

The useful resolution of measurements at the lambda point on Earth is limited in principle by the competition between finite-size effects and the pressure dependence of the transition temperature [17]. The rounding expected from finite-size effects is not known exactly, but an estimate can be made by using simple scaling and correlation length arguments. Experiments [18] show that the heat capacity curve exhibits a finite maximum slightly below the bulk transition temperature when the correlation length becomes a significant fraction of the characteristic sample dimension, at least for length scales of up to a few microns. From the power law behavior of the correlation length we can easily extrapolate this rounding to larger sample sizes. For our case we expect a heat capacity maximum of about 133 J/mole K occurring about 10^{-11} K below the bulk transition. Although transition broadening due to pressure gradients can be calculated [19] with a great deal of confidence, in a flight experiment the residual acceleration is quite variable, and this broadening effect can be bounded only roughly. The low-frequency residual acceleration in the Shuttle is generally no more than $2 \times 10^{-6}g$ below 0.03 Hz, the frequency range of interest here. From this number and the slope of the lambda line we obtain a pressure broadening of about 10^{-11} K. Transient acceleration amplitudes on the order of $10^{-3}g$ exist, generally in the form of high-frequency spikes, but since we are

concerned only with the energy of the sample, they have little direct effect on the results. As one moves further from the transition, both effects can be characterized by rapidly decreasing deviations of the heat capacity from its ideal singular form. These were estimated to be <0.1% at temperatures >2 nK from the transition, and are negligible in the data analysis. The gas supply from which the sample was drawn was found to contain about 0.45 ppb ³He impurity, which would lead to a negligible renormalization of the exponent [20] over the range of the measurements.

The heat capacity results over the whole range measured are shown on a logarithmic temperature scale in Fig. 1. It can be seen that there is no sign of transition broadening within the resolution of the experiment. To compare the observations with theory, we fit the data using functions of the type

$$C_p = At^{-\alpha}(1 + Dt^{\Delta} + Et) + B,$$
 (1)

where the parameters A, B, D, E, α , and Δ can be independently adjusted on each side of the transition. By convention, the exponents and constants are primed below the transition to distinguish them from those above. In Eq. (1), the first and last terms represent the asymptotic singular form, while the other two terms represent the confluent singularity and a slowly varying background, which is carried to allow for higher-order effects in the region far from the transition. In addition to the predictions for the exponents, a number of universal relationships are expected [21] to exist between the parameter values on the two sides of the transition, giving us a wide selection of possible constraints or tests. In general, we need some constraints in the data analysis to decrease the effects of correlations between the parameters.

Figure 2(a) shows a deviation plot of the heat capacity measurements below the transition from the best-fit function [22] with all parameters except Δ (set [23] at 0.5)



FIG. 1. Flight heat capacity data on a log-linear scale. Curves show the optimal fits of Eq. (1) to the two branches of the data as described in the text.

optimized. It can be seen that the fit is excellent, indicating that Eq. (1) is an adequate asymptotic representation of the data. The lower parts of the figure show for comparison the deviations of selected ground measurements from the same function. Figure 2(b) shows the data of Lipa and Chui [15] as given in their Fig. 2, obtained with a cell 3 mm high. Also shown by the broken curve is the expected heat capacity in the Earth's gravity field [24]. Data obtained with a 0.3 mm high cell are also available [25], but these results are noticeably distorted by finitesize effects. Figure 2(c) shows the data of Ahlers [26] obtained with a cell 1.59 cm high, plotted in a similar fashion. Given the scale factor uncertainties in these latter two experiments, 1.5% and $\sim 1\%$, respectively, the overall level of agreement between the data sets is encouraging. Careful attention was paid to all aspects of calibration of the flight experiment, and the heat capacity values are expected to be reliable to within 0.2% using the EPT-76 temperature scale. It is clear that the new results give us the opportunity for extending the testing of the theoretical predictions much closer to the transition than was possible previously, avoiding the severe limitations of gravity in the ground experiments.

A direct test of the RG theory is to compare the value of the exponent α with predictions based on approximate expansion techniques. The optimum value for α' obtained from the curve fitting was -0.01285 ± 0.00038 . This result is in agreement with two theoretical estimates [10,11], but the experimental uncertainty is much lower. We hope that our measurement stimulates work in this



FIG. 2. Deviation plots of heat capacity data from the function which best fits the flight results below the transition: (a) flight data; (b) Lipa and Chui (Ref. [15]); (c) Ahlers (Ref. [24]). Dashed curves: estimated behavior allowing for gravity.

946

area. The various experimental and theoretical values for α' are collected in Table I. It can be seen that the uncertainty in the new result is about a factor of 6 smaller than that obtained in the ground experiments. Another important check on the RG theory can be obtained by including ground results for ζ and testing the exact relation [30], $\alpha' = 2 - 3\zeta$. Recent measurements [31,32] give $\zeta = 0.6705 \pm 0.0006$, and 0.6708 ± 0.0004 . The corresponding values for α' are -0.0115 ± 0.0018 and -0.0124 ± 0.0012 , respectively. Both of these values, with their uncertainties, nicely overlap the flight result. The value we obtain for the magnitude of the confluent singularity term, $D' = -0.0228 \pm 0.003$, is in good agreement with the previous experimental estimate [33], -0.020.

Substantially less heat capacity data was collected on the high temperature side of the transition due to mission time constraints. Also, this data was of lower accuracy because of the need to wait for thermal equilibration after each heat pulse. The data collected extends over the range $3 \times 10^{-9} < t < 2 \times 10^{-6}$. Because of its relatively low accuracy, a detailed comparison of this data with theory is not warranted. Nevertheless, we fit the data with Eq. (1), constraining all parameters except A to be the same as on the low temperature side. We found that the ratio A/A' of the leading coefficients was 1.054 ± 0.001 . This agrees well with the value found in the most recent experiment at the vapor pressure [15]. The curves shown in Fig. 1 are the best fits to the two branches of the data.

Above the transition, the calorimeter temperature initially overshoots its equilibrium value when a heat pulse is applied, due to the finite thermal conductivity of the helium. The resulting relaxation data and an estimate of the heat capacity allow us to derive values for the thermal conductivity, using the expression [34] $K = a^2 \rho C_p / 6.62\tau$, where τ is the relaxation time for the slowest thermal transients, ρ is the density, and *a* is the radius of the sample. In Fig. 3 we compare this data with the behavior predicted by a fit of the dynamic RG calculations of Dohm [35] to the measured thermal conductivity [36] further away from the transition. It can be seen that there is reasonable agreement with the functional form predicted by the model. In particular, the data in Fig. 3

TABLE I. Comparison of predicted and observed values for the heat capacity exponent.

Range of fit	Exponent value	Ref.
$2 \times 10^{-5} \leftrightarrow 3 \times 10^{-3}$	-0.026 ± 0.004	8
$10^{-6} \leftrightarrow 3 \times 10^{-3}$	-0.01 ± 0.01	26
$10^{-6} \leftrightarrow 3 \times 10^{-3}$	-0.0198 ± 0.0037	27, 28
$3 \times 10^{-6} \leftrightarrow 2 \times 10^{-3}$	$-0.017 \leftrightarrow -0.023$	29
$3 \times 10^{-8} \leftrightarrow 10^{-3}$	-0.0127 ± 0.0026	15
$10^{-9} \times 10^{-2}$	-0.01285 ± 0.00038	This work
Prediction	-0.007 ± 0.006	10
Prediction	-0.016 ± 0.006	11



FIG. 3. Comparison of predicted (Ref. [35]) and observed thermal conductivity in the region just above the transition.

imply that the effective amplitude [35] R_{λ} continues to increase as T_{λ} is approached. However, the best fit to our data would be about 10% higher than the curve shown. There is some need for caution in interpreting these flight results, which are the average of a number of measurements with wide scatter [37], and should be considered as provisional.

We wish to thank the NASA Office of Life and Microgravity Science and applications for its support with Contract No. 957448, M. Lee for his long support of the program, the members of the LPE group at Stanford and JPL for their many contributions to the experiment, and Ball Aerospace for the construction of major portions of the flight instrument. We also thank V. Dohm and G. Ahlers for helpful suggestions and a careful reading of the manuscript.

- [1] L. Onsager, Phys. Rev. 65, 117 (1944).
- [2] W. M. Fairbank, M. J. Buckingham, and C. F. Kellers, Proceedings of the Fifth International Conference on Low Temp. Phys., Madison, WI, 1957, p. 50.
- [3] M. I. Bagatskii, A. V. Voronel', and V.G. Gusak, Zh. Eskp. Teor. Fiz. 43, 728 (1962) [Sov. Phys. JETP 16, 517 (1963)].
- [4] C. Edwards, J. A. Lipa, and M. J. Buckingham, Phys. Rev. Lett. 20, 496 (1968).
- [5] G. Ahlers, Phys. Rev. Lett. 23, 464 (1969).
- [6] For the case $\alpha = 0$, a logarithmic function, $A \ln t + B$, is substituted.
- [7] J. A. Lipa, M. J. Buckingham, and C. Edwards, Phys. Rev. Lett. 25, 1086 (1970); M. R. Moldover, in *Phase Transitions, Cargese, 1980*, edited by M. Levy, J-C. Le Guillou, and J. Zinn-Justin (Plenum, New York, 1982), p. 63.
- [8] K. H. Mueller, G. Ahlers, and F. Pobell, Phys. Rev. B 14, 2096 (1976).
- [9] K.G. Wilson, Phys. Rev. B 4, 3174 (1971).
- [10] J. C. Le Guillou and J. Zinn-Justin, Phys. Rev. B 21, 3976 (1980).
- [11] D.Z. Albert, Phys. Rev. B 25, 4810 (1982).

- [12] Nippon Mining Co., Japan, grade 6N (99.9999% pure).
- [13] J.A. Lipa, B.C. Leslie, and T.C. Wallstrom, Physica (Amsterdam) **107B**, 331 (1981); T.C.P. Chui, D.R. Swanson, M.J. Adriaans, J.A. Nissen, and J.A. Lipa, Temp. Its Meas. Control Sci. Ind. **6**, 1213 (1992); T.C.P. Chui, D. Swanson, M.J. Adriaans, J.A. Nissen, and J.A. Lipa, Phys. Rev. Lett. **69**, 3005 (1992).
- [14] J. A. Lipa, T. C. P. Chui, J. A. Nissen, and D. R. Swanson, Temp. Its Meas. Control Sci. Ind. 6, 949 (1992).
- [15] J. A. Lipa and T. C. P. Chui, Phys. Rev. Lett. 51, 2291 (1983).
- [16] D. Petrac, U.E. Israelsson, and T.S. Luchik, Adv. Cryo. Eng. **39**, 137 (1994).
- [17] J. A. Lipa, in 75th Jubilee Conference on Helium-4 edited by J. G. M. Armitage (World Scientific, Singapore, 1983), p. 208.
- [18] T. P. Chen and F. M. Gapsparini, Phys. Rev. Lett. 40, 331 (1978); M. Coleman and J. A. Lipa, Phys. Rev. Lett. 74, 286 (1995).
- [19] G. Ahlers, Phys. Rev. 171, 275 (1968).
- [20] M.E. Fisher and P.E. Scesney, Phys. Rev. A 2, 825 (1970).
- [21] V. Privman, P. C. Hohenberg, and A. Aharony, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, New York, 1991), Vol. 14, p. 1.
- [22] The best fit parameter values are $\alpha' = -0.012850$, A' = 5.7015, B' = 456.28, D' = -0.0228, E' = 0.323.
- [23] The exponent Δ has previously been shown to be close to its theoretical value of 0.5, which was used in the fitting process.
- [24] For the ground results, the transition temperature has been taken as the point where the calorimeter is first full of normal fluid [See G. Ahlers, Phys. Rev. A 3, 696 (1971)].
- [25] T. C. P. Chui and J. A. Lipa, in 75th Jubilee Conference on Helium-4, edited by J. G. M. Armitage (World Scientific, Singapore, 1983), p. 206.
- [26] See Ref. 24. Best fit exponents in the range 0.0 to -0.02 are quoted.
- [27] F. M. Gasparini and A. A. Gaeta, Phys. Rev. B 17, 1466 (1978).
- [28] F. M. Gasparini and M. R. Moldover, Phys. Rev. B 12, 93 (1975).
- [29] T. Takada and T. Watanabe, J. Low Temp. Phys. 41, 221 (1980).
- [30] B. D. Josephson, Proc. Phys. Soc. 92, 269 (1967).
- [31] L. S. Goldner, N. Mulders, and G. Ahlers, J. Low Temp. Phys. 93, 131 (1992).
- [32] D. R. Swanson, T. C. P. Chui, and J. A. Lipa, Phys. Rev. B 46, 9043 (1992); D. Marek, J. A. Lipa, and D. Philips, Phys. Rev. B 38, 4465 (1988).
- [33] G. Ahlers, in *The Physics of Liquid and Solid Helium*, edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, 1976), Part 1, p. 85.
- [34] M. Necati Ozisik, Boundary Value Problems of Heat Conduction, (Dover, New York, 1989).
- [35] V. Dohm, Phys. Rev. B 44, 2697 (1991); Z. Phys. B 61, 193 (1985).
- [36] W. Y. Tam and G. Ahlers, Phys. Rev. B 32, 5932 (1985).
- [37] This scatter appears to be primarily an artifact of the curve fitting used to extract the relaxation time, and is being investigated further.