LOGARITHMIC SINGULARITIES

Chairman: C. N. Yang

The Lambda Transition in Liquid Helium*

W. M. Fairbank

Stanford University, Stanford, Calif.

and

C. F. Kellers

Wells College, Aurora, N.Y.

This talk is based on work done at Duke University several years ago by C. F. Kellers, M. J. Buckingham and myself [1]. A major part of the work constituted the Ph.D. thesis of Dr. Kellers [1b]. Much of what I will say has been previously reported [1]. Still, when Dr. Green asked me to give this talk I consented because I believed a review of the lambda transition in liquid helium is particularly pertinent to the present conference on critical phenomena. Not only were the Ehrenfest relationships for a second order transition enunciated first for the lambda transition in liquid helium, but liquid helium also served, through the experiments which I will describe, to first call attention to the logarithmic character of the lambda transition which is so much a part of the discussion of this conference.

The lambda transition in liquid helium is, of course, of particular interest because of the unique momentum ordering that takes place below the lambda point which makes it perhaps similar but certainly different from all other transitions. In addition, however, of all the cooperative transitions, it is the one which offers the best chance for experimental observations very close to the transition. Because liquid helium does not have any crystal structure, because it is a pure liquid changing at the lambda point only in its momentum ordering, it is apparently possible with sufficiently good thermal equilibrium to make measurements as close as experimental techniques permit to the lambda point without observing any of the broadening characteristics of other transitions.

Ever since London suggested that liquid helium represents in its superfluid phase a long-range order in momentum space, physicists have been puzzled by exactly what this means. London, Landau, Onsager, and Feynmann have suggested that liquid helium as a superfluid is required to be irrotational. If the helium rotates, it rotates according to the Onsager-Feynmann theory, with quantized vortex motion [2]. Helium rotating slowly enough is predicted to rotate in one single quantized state of rotation with each helium atom having angular momentum $\hbar$ regardless of the size of the bucket in which it is contained [2, 3]. The theoretical suggestion is then the following: If a bucket of helium no matter how large is cooled through the lambda transition into the superfluid state while rotating slowly enough, each superfluid helium atom will attain the same angular momentum. For the slowest stage the superfluid will stop rotating and the bucket, if suspended freely, will rotate faster [4]. This experiment has never been performed. Some physicists, including a graduate student George Hess at Stanford, are trying to perform this experiment [5]. This then is the type of order transition which is predicted for liquid helium, long-range order in momentum space of a very special kind.

Blatt, Butler, and Schafroth have taken another approach to the problem [6, 7, 8, 9]. They questioned whether a real liquid, as contrasted with an ideal Bose gas can exhibit arbitrarily long-range order or correlation in momentum space. They suggest that there may be a finite correlation length of approximately 10^{-5} cm over which helium atoms are correlated in momentum space, but beyond which there is no correlation at all. In particular, they developed a theory of a Bose liquid in which helium atoms are correlated over a distance of about 10^{-5} cm, the correlated atoms representing the superfluid. By calculating the amount of this correlated phase ρ, they calculate the ratio of uncorrelated to correlated phase (normal to superfluid, ρ_n/ρ_s) as a function of temperature, the correlation length being an adjustable parameter. With a cor-

*Supported in part by the National Science Foundation and the Army Research Office Durham.

relation length of 10^{-5} cm, they were able to accomplish something which to my knowledge had not been accomplished before. They were able to derive ρ_n/ρ_s as a function of temperature in agreement with experiment up through the lambda point and with the same theory, to derive the specific heat at the lambda point in quite good agreement with experiment both as to shape, magnitude, and temperature of the transition. Their theoretical curves are shown along with the then existing experimental data in figures 1 and 2. One characteristic new feature of this specific heat transition is that it is a quasi-transition. Instead of being a second-order transition with a discontinuity in the specific heat at the lambda point, the specific heat is rounded at the lambda point for a temperature interval of approximately 10^{-3} deg which is a direct result of a finite correlation length. This rounding could not have been observed in the then existing data. It also follows from their theory that curl $\mathbf{v}_s = 0$ is not an equilibrium property of superfluid helium II.

Feynmann [10] took issue with the premise that the correlation length in liquid helium could be cutoff at some finite distance, and suggested that the lambda point in liquid helium is not only sharp at one-thousandth of a degree from the lambda point but it is sharp at a hundred-thousandth of a degree. He believed that even though the correlation in liquid helium II gets weaker and weaker as the distance gets bigger, it still remains finite. Thus, no matter how large the bucket, liquid helium would still rotate in a single quantum state if the rotation were sufficiently slow.

At the expense of bringing you a free quote which was only told to me by Feynmann and therefore is not quoted from the literature, I am going to quote an exchange between Feynmann and Blatt because I think it illustrates the problem of long-range order very clearly for this conference. "Blatt, 'according to Feynmann', said to him, 'Feynmann, how is your intuition so good that you can say that there is a correlation between helium atoms separated by the size of the universe.' 'Feynmann said he replied', 'Blatt, how is your intuition so good that you can say a correlation which decreases as $1/r^2$ and is down by something like 10^{70} at the edge of the universe, doesn't exist'." That is just the point, this correlation falls off very rapidly. If the helium is to stop rotating in a rotating bucket when the bucket is cooled below the lambda point, then the rotational speed must decrease as $1/r^2$ as the size of the bucket becomes larger and larger.

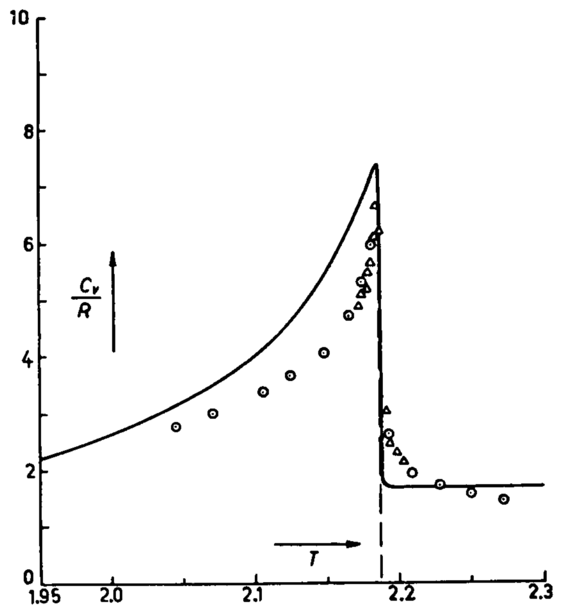

FIGURE 1. *The specific heat of liquid helium versus temperature in the neighborhood of the λ-point.*

The curve is theoretical, the experimental points are taken from Keesom: Helium (Amsterdam, 1942), Table 4.20. The theoretical curve is a continuous and differentiable function of temperature everywhere, represented by the same theoretical expression throughout the temperature region covered in ref. [8].

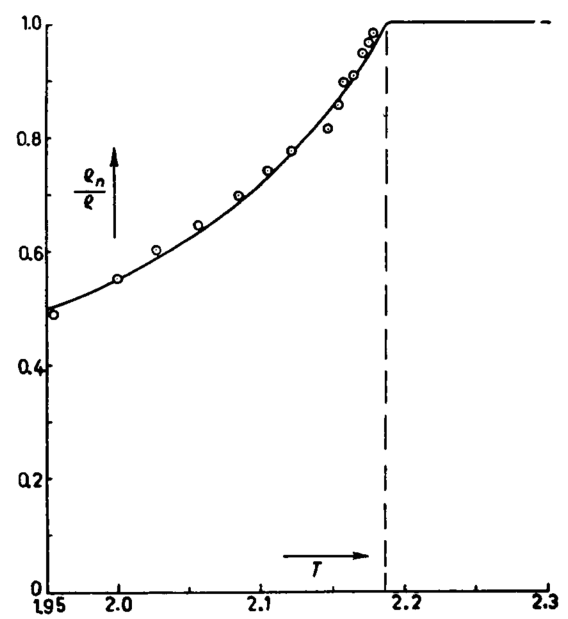

FIGURE 2. *The normal fluid concentration ρ_n/ρ in liquid helium versus temperature, in the neighborhood of the λ-point.*

The curve is theoretical, the experimental points are based on the second sound data of Pellam. The theoretical curve is a continuous and differentiable function of temperature everywhere, represented by the same theoretical expression throughout the temperature region covered in the ref. [9].

These conflicting theoretical predictions served as an incentive for us to perform a specific heat measurement to at least 10^{-5} deg of the lambda point. This experiment was performed by Buckingham, Kellers, and Fairbank, and has been reported in reference 1. Ultimately C_s was measured to within 10^{-6} deg of the lambda point T_λ, with equal precision both above and below T_λ. I would like to recall here just a few highlights of the experiment and comment on the possible significance of the data.

Many previous measurements have been made on the lambda point, including an unpublished master's degree thesis from our laboratory. The difficulty with all these earlier experiments was that reasonably large amounts of liquid helium was used in containers which were connected to the outside world.

To obtain the needed high temperature resolution it is essential that the attainment of equilibrium be unaffected by the drastic change of thermal conductivity of liquid helium at the lambda point, or by the onset of the creeping film. Both of these requirements were met by permanently sealing the helium (0.0587 g) in a cooper container (200 g), the inside of which was in the form of fins so placed that the helium was everywhere within 0.003 in. of the copper surface (fig. 3). With a heat input of 10 erg/s temperature equilibrium of greater than 10^{-6} deg could be obtained, even in the helium I region. In order to eliminate the need for removing exchange gas, the sealed container was suspended in a vacuum and a mechanical heat switch provided for contact with the bath when required.

Measurements were made by means of a carbon resistor with a minimal detectable change representing 2.10^{-7} deg. It was possible to make measurements both while increasing and decreasing the temperature.

Figure 4 shows the data. These of course are not new data just taken, but the reason I was asked to present them is that perhaps they are very similar to data which are now being obtained on other transitions. The specific heat, C_s, is plotted as ordinant versus $|T - T_\lambda|$. In order to show the nature of the transition very near the lambda point, the data are shown on successively expanded temperature scales. To aid in a visualization of the very large amount of expansion of each successive curve, a small vertical line has been drawn just above the origin. The width of the line indicating the fraction of the curve which is shown expanded in the curve directly to the right. The ratio of the expansion between

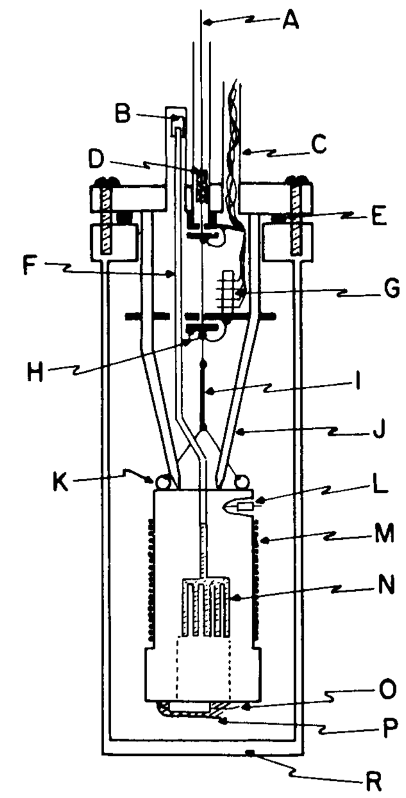

FIGURE 3. *Schematic diagram of adiabatic chamber for specific heat measurements [1c]: (A) stainless steel wire for closing heat switch; (B) brass cap on filling capillary; (C) wires connecting to heater and resistors on sample; (D) cotton plug dyed with carbon black as radiation trap; (E) indium "O" ring; (F) filling capillary; (G) Kovar seal used as thermal short for wires to sample; (H) radiation shield and thermal short for heat switch; (I) nylor cord; (J) three prongs of heat switch (copper); (K) indium coating and suspension for sample; (L) temperature-sensitive resistor; (M) heater; (N) sample cavity; (O) temperature-sensitive resistor; (P) copper shield over resistor; (R) calorimeter wall.*

Figure taken from ref. [1c].

the first and last curve is about 5×10^4. Thus if the projected slide of the first diagram is 10 ft, it would have to be expanded to 100 miles to obtain a 10 ft projection of the third figure.

It can be seen that as the specific heat is displayed on a more and more expanded scale, it maintains the same geometric shape. There is certainly no indication of a rounded quasi-transition as predicted by the theory of Blatt, Butler, and Schafroth.

Figure 5 shows the data in a form with which you are all now familiar. C_s has been plotted as ordinant against $T - T_\lambda$ in degrees Kelvin on a logarithmic scale. It is seen in this kind of a plot that near the lambda point on each side there is a factor of 10^4 in $T - T_\lambda$ over which the data fall in two

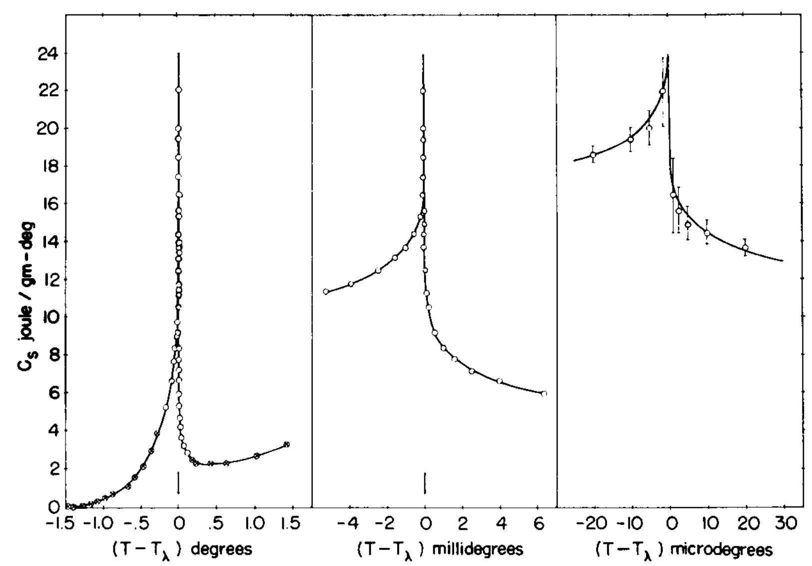

FIGURE 4. *Specific heat of liquid helium versus* $T - T_\lambda$ *in °K* [1].
0 represent data of Kellers, Fairbank and Buckingham [1]. ⊗ represent, above 1.5 °K, data of Hill and Lounasmaa [20] and Lounasmaa and Kojo [18] and, below 1.5 °K, data of Kramers, Wasscher, and Gorter [21]. Solid line near λ-point represents eq (1). Width of small vertical line just above origin indicates portion of diagram shown expanded (in width) in the curve directly to the right. Figure from ref. [1c].

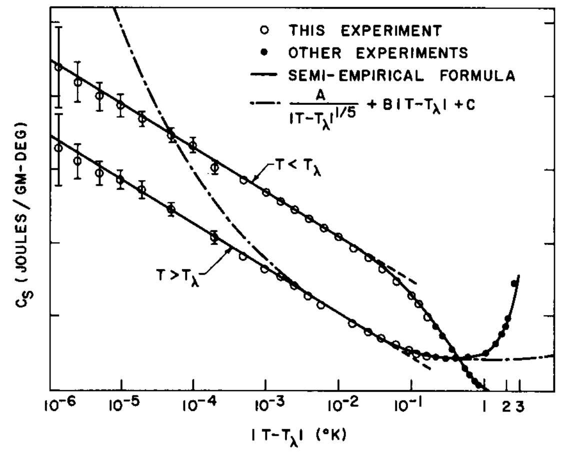

FIGURE 5. *The specific heat of liquid helium versus log*$|T-T_\lambda|$ [1].
0 represent data of Kellers, Fairbank and Buckingham [1]. ● represent, above 1.5 °K data of Hill and Lounasmaa [20] and Lounasmaa and Kojo [18] and, below 1.5 °K, data of Kramers, Wasscher, and Gorter [21]. Two parallel straight lines represent eq (1). The broken line represents formula shown in the figure. Figure from ref. [1c].

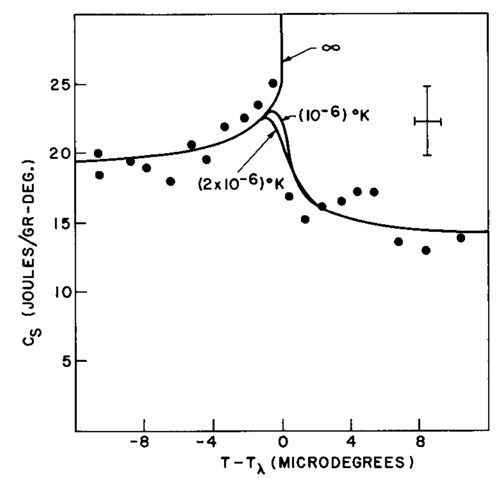

FIGURE 6. *Plot of* C_s *versus* $T - T_\lambda$ *very near λ-point.*
Data taken from one single run randomly selected from five runs where there were no disturbing influences. Figure from ref. [1b].

parallel straight lines which are branches of the expression

$$C_s = 4.55 - 3.00 \log_{10}|T - T_\lambda| - 5.20\Delta \qquad (1)$$

where $\Delta = 0$ for $T < T_\lambda$, and $\Delta = 1$ for $T > T_\lambda$. One reason that this was very interesting and exciting at the time, is that Onsager had published the exact solution of the two-dimensional Ising model [11]. The two-dimensional Ising model gives a logarithmic transition but only for an exact solution.

Figure 6 shows previously unpublished data from a single run taken from C. F. Keller's Ph.D. thesis [1b]. About 20 runs were taken back and forth through the lambda point over ±10 μdeg per minute. About half of the runs were discarded because the rate of cooling or heating at the beginning or end of the run was different. In about half of the remaining runs there was a big spike in the data due to someone opening the door and so forth. Of the remaining 5 runs, the one shown in figure 6 is typical and picked at random. The data is

averaged over 1 μdeg so the lambda point is still clearly observable. The three solid curves are placed in the diagram to show the effect of averaging over finite temperature intervals 10^{-6} deg Kelvin and 2×10^{-6} deg Kelvin. The curve labeled infinity is the curve given in eq (1) and represents infinite resolution. As stated above these data were averaged over 1 μdeg interval and should correspond to that curve labeled 10^{-6}.

Thus we see from the experimental data that the lambda point, instead of being a rounded quasi-transition as suggested by the theory of Blatt, Butler, and Schafroth assuming a finite correlation length, is in fact, sharp to at least 2 orders of magnitude closer in temperature to the lambda point than predicted by their theory. The simple logarithmic behavior of the data shown in figure 4 which extrapolates to a logarithmic singularity of the lambda point has stimulated more detailed experimental and theoretical investigation of the lambda point, as witnessed so completely by this conference. Although the two-dimensional Ising model has been solved exactly by Onsager giving such a logarithmic singularity, it is obvious from this conference that an exact solution to the three-dimensional Ising model will not be easily found.

In figure 5 the curve of the form

$$\frac{A}{|T-T_\lambda|^{1/5}} + B|T-T_\lambda| + C$$

which has been fitted to the data for $T - T_\lambda$ between 2×10^{-1} and 2×10^{-3} as shown. This is the fundamental form suggested by the Padé approximation of the three-dimensional Ising model. It is seen that although it fits the data rather well for $(T-T_\lambda) > 2 \times 10^{-3}$°K, it departs very dramatically for temperatures closer to the lambda point. Unless data are obtained close enough to the lambda point one could not differentiate between this expression and the logarithmic singularity.

Much of this conference will be taken up with the exact nature of other kinds of lambda transitions. However, I wish to compare quickly two results by Robinson and Friedberg [12] and Skalyo and Friedberg [13] on the lambda transition in hydrated nickel and cobalt chloride with specific heat data on liquid helium.

Figure 7 shows the specific heat data of nickel chloride to within 0.07 °K of the lambda point from above, and within 0.2 °K from below the transition temperature. Figure 8 shows the same data with C_p plotted against $\log|T - T_\lambda|$. If one defines a reduced temperature by $t = |T - T_\lambda|/T_\lambda$ and compares the curve for nickel chloride with the helium curve, one sees for the same value of t both curves show nonparallel straight lines.

Figure 9 shows the data for cobalt chloride taken to smaller values of t. These data show two parallel straight lines and then a flattening out of the data.

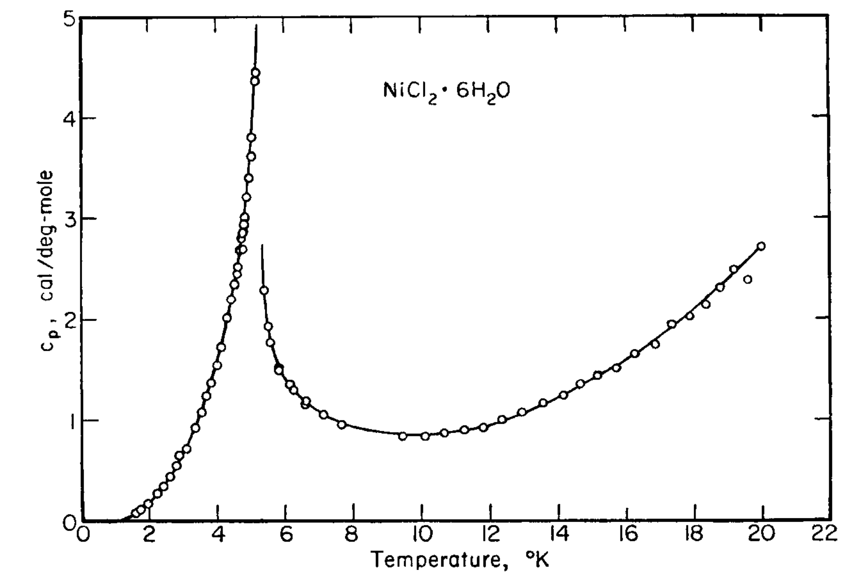

FIGURE 7. C_p *versus* $|T-T_\lambda|$ *and* C_p *versus log*$|T-T_\lambda|$ *for* $NiCl_2 \cdot 6H_2O$.
Data by Robinson and Friedberg taken from ref. [12].

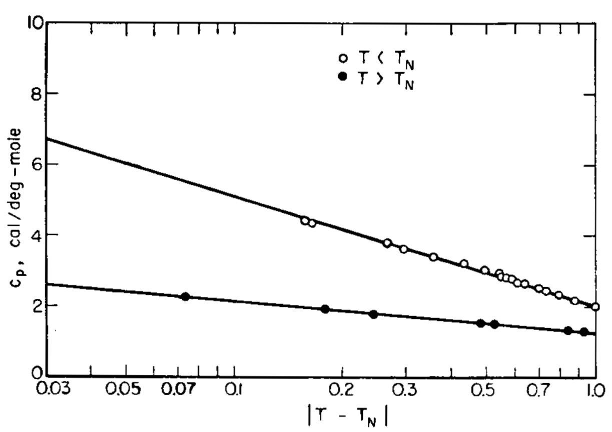

FIGURE 8. C_p *versus* $|T-T_N|$ *and* C_p *versus* $\log|T-T_N|$ *for* $NiCl_2 \cdot 6H_2O$.
Data by Robinson and Friedberg taken from ref. [12].

A comparison with helium indicate agreement as to the two parallel straight lines. The flattening out at values of t closer to the lambda point would be expected in either an experiment or theoretical model where long-range correlation is cutoff. These two sets of data are then seen to be consistent with the lambda transition for helium and indicates the possibility that other lambda points have the same form as the helium transition.

The suggestion that the specific heat of helium might become infinite at the lambda transition was first made by Tisza [14]. The first experimental evidence concerning the logarithmic nature of the lambda transition was obtained by Atkins and Edwards [15]. They suggested that a logarithmic term could be used to derive the result of their measurements of the thermal expansion coefficient below the lambda point. Figure 10 shows the data on the expansion coefficient of Atkins and Edwards, Chase and Maxwell, and Kerr and Taylor. It is seen that the data can be represented by two parallel straight lines from the plot of the expansion coefficient versus $\log(T-T_\lambda)$. This curve is reproduced from the paper by Kerr and Taylor [16]. Figure 11 shows the relative molar volume of helium in the vicinity of the lambda point as given by Kerr and Taylor. It is seen that the lambda point comes where the slope is infinite rather than at the point where the molar volume is minimum.

Buckingham [1c] has derived rigorously the thermodynamic consequences of lambda transitions characterized by the absence of a latent heat, but at which the specific heat at constant pressure becomes infinite. Pippard [17] had previously considered such a transition and worked out thermodynamic relationships based on the assumption that the entropy surface is cylindrical near the lambda point. The thermodynamic relationships worked out by Buckingham and Pippard can be

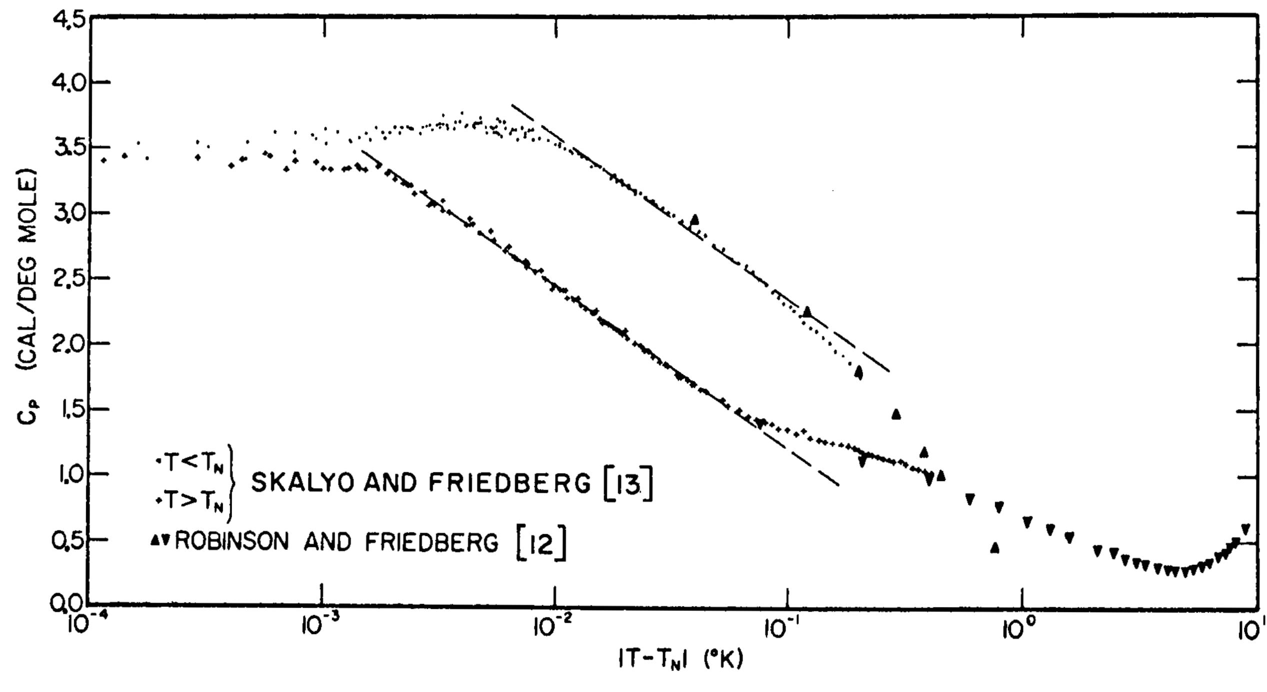

FIGURE 9. *The heat capacity,* C_p, *of* $CoCl_2 \cdot 6H_2O$ *as a function of* $\ln|T-T_N|$ *for* $T_N = 2.2890$ °K.
Figure taken from ref. [13].

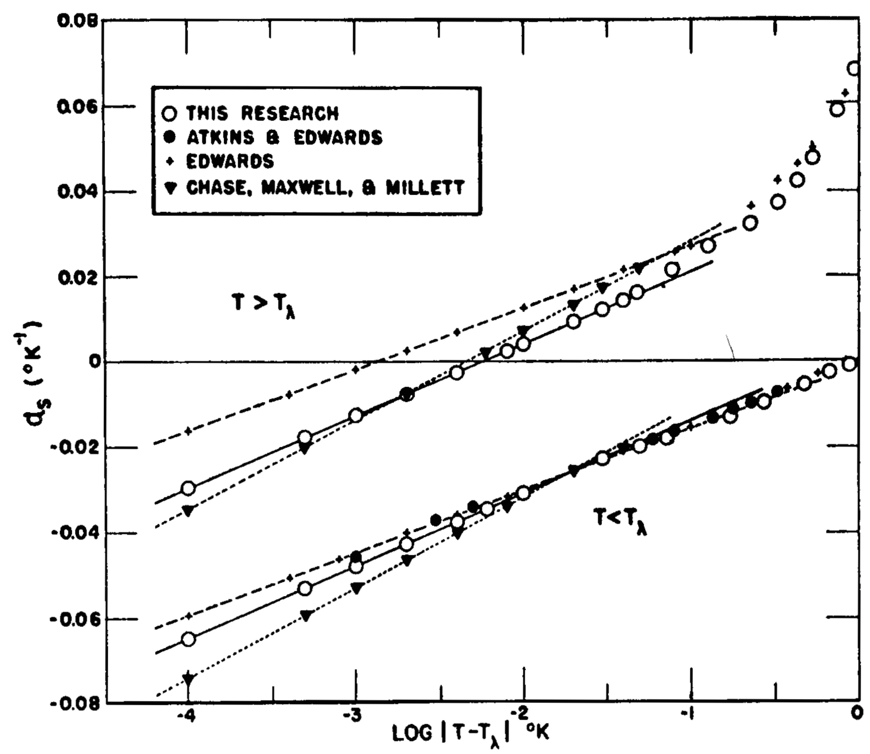

FIGURE 10. *The expansion coefficient,* α_s, *near the* λ*-point as a function of* $\log|T-T_\lambda|$.
Curve from Kerr and Taylor. Figure taken from ref. [16].

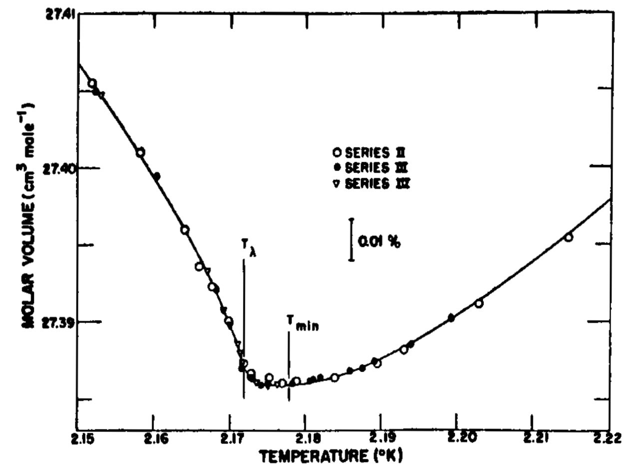

FIGURE 11. *The molar volume of* He^4 *in the vicinity of the* λ*-point.*
Data by Kerr and Taylor. Figure from ref. [16].

used to compare the behavior of various thermodynamic properties in the neighborhood of the lambda line [1c]. In reference 1c and 16 the specific heat and coefficient of expansion are compared using the Buckingham relationship. The slope of the lambda line $(\partial S/\partial T)_\lambda$ is taken from the data of Lounasmaa and Kojo [18] and Lounasmaa and Kaunisto [19]. The data can be plotted in a parametric plot which should give asymptotically a single straight line. Two parallel straight lines are obtained by Kerr and Taylor using the data shown in figures 4 and 10. This shows that the fundamental form of the data is correct but the details need to be refined. It is difficult to obtain exactly the same experimental conditions in different experiments very close to the lambda point. It seems worthwhile to make simultaneous specific heat and expansion coefficient measurements to within 10^{-6} °K of the lambda transition.

The behavior of the velocity of sound in the vicinity of the lambda point can also be predicted using specific heat and other thermodynamic data [1c]. Here again there is approximate but not exact agreement. Since there will be a separate talk on this subject I will not present further data here.

So in summary, the specific heat expansion coefficient and velocity of sound all give evidence to the logarithmic nature of the lambda transition in helium although refined details of comparison still need to be worked out. I would like to end by again mentioning that liquid helium presents the unique opportunity to obtain data close to the lambda point. The question naturally arises, is the transition in liquid helium similar to lambda transition in other substances, and does liquid helium present an idealized experimental model for all lambda transitions? At first glance one might think that liquid helium doesn't. I would, however, like to point out a sense in which liquid helium does present such a model; a sense that has been emphasized by Buckingham in private discussions.

When one goes from the normal liquid to the superfluid in liquid helium, one gets a change from a normal system to something which is suddenly ordered throughout the whole container. In this particular case, it is an ordering in the superfluid. The superfluid atom in one part of the container knows what is happening to the superfluid atom in a different part of the container, and how long that order is depends only on the size of the container. One can disturb the system without destroying this order throughout the entire system provided the disturbances, for example rotational speeds, become smaller and smaller as the size of the system becomes larger and larger.

In the case of superconductors one has a quantized magnetic flux over supposedly the distance of the length of the wire in superconducting magnets that may be thousands of feet long. The length of the ordering depends only on the size of the superconducting loop and especially in the case of superconductors, exists in the presence of large quantities of impurities. Now there is different kind of order parameter in superconductors and that is the length of the correlation between electrons in a pair. This is only 10^{-4} cm, but the two electrons in a pair have equal and opposite momentum. The momentum of each pair is zero and therefore every pair throughout the whole superconductor has the same zero momentum. Thus there is a long-range order in momentum throughout the entire superconductor even though each pair is paired over 10^{-4} cm.

One doesn't have this momentum ordering in any other kinds of transition besides superconductors and liquid helium. But on the other hand, as Buckingham is going to point out at the end of the morning, one has a transition from a state where ordering exists within a small cluster in a particular part of a container, to a system where one has suddenly, for example in a liquid gas transition, order throughout the entire container. Part of the atoms have one special density and they are all together in the container, and all the rest of the atoms have a different density. This sudden transition is to a state of order which depends only on the size of the container.

I want to close with this question. Is the lambda transition in liquid helium, except for the fact that one has a lambda line, an exact model for other transitions when long-range correlations are not cutoff in other experiment systems or theoretical models?

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The Specific Heat of He^3 and He^4 in the Neighborhood of Their Critical Points*

M. R. Moldover and W. A. Little

Stanford University, Stanford, Calif.

Introduction

Recently Bagatskiĭ, Voronel', and Gusak [1] showed that the specific heat at constant volume of argon exhibited what appears to be a logarithmic singularity at the critical temperature (T_c) for measurements taken at a density near the critical density. This singular behavior is in sharp contrast to the predictions of the traditional view of this phase transition by Landau and Lifshitz [2]. However, the behavior is precisely that to be expected for the so-called "lattice gas" model for the liquid-gas transition. Lee and Yang [3] have shown that the partition function of a classical gas of particles moving on a discrete lattice with a repulsive force preventing double occupancy of any site, and a nearest neighbor attraction can be mapped precisely onto the partition function of an Ising model of a spin system in an external magnetic field. The specific heat for this Ising model in zero field exhibits a logarithmic singularity at the Curie point. The specific heat for the corresponding lattice gas on the critical isochore exhibits a logarithmic singularity at the critical point. The measurements on argon then indicate that for a *real* gas the specific heat behaves in a similar manner to that of a lattice gas. We have investigated this point further by studying the specific heat at constant volume (C_V) of both He^3 and He^4 at densities close to the critical density. We have done this for two main reasons. Firstly, to see whether the behavior observed for argon is also observed for helium, for which quantum effects should be important, and secondly, to investigate the detailed nature of the singularity in the pressure-density plane, not only *on* the critical density, but also in its immediate neighborhood. Yang and Yang [4] have conjectured that the quantum effects would reduce the magnitude of the singular contribution to the specific heat in helium. Our results confirm this view.

*Work supported in part by the National Science Foundation and the Office of Naval Research.

Experimental Procedure

There are several practical advantages to using helium rather than another noble gas for C_V measurements near the critical point. The low heat capacity of metals at liquid helium temperature permits one to use a massive calorimeter of large surface to volume ratio. Thus the path for heat transfer through the helium may be kept short. In addition, more nearly adiabatic conditions and high resolution thermometry are most easily attained at liquid helium temperatures.

Our calorimeter was built of two OFHC copper parts. The helium was contained in the lower part in 50 slots. Each slot was 0.01 cm wide to facilitate good thermal contact between the calorimeter and the helium. The slots were made only 0.3 cm deep in an effort to minimize possible gravitational effects. [In contrast a thin stainless steel shell 10 cm high and 4 cm in diameter containing a magnetic stirrer was used for the work on argon [1, 5]. This construction was necessary to obtain a low ratio of heat capacity of the calorimeter to its contents while maintaining constant volume at the high critical pressure of argon.] The lower part of our calorimeter was wound with a constantan heater and had a carbon resistor clamped and cemented to it. The calorimeter was supported on nylon threads in an evacuated chamber. Thermal contact to the bath was made with a mechanical heat switch. A stainless steel filling capillary 5 in. in length and 0.006 in. I.D. led from the calorimeter to a needle valve. The dead volume was about 1/2 percent of the total volume of the calorimeter. The helium was admitted to the calorimeter via a Toeppler pump which was used to measure the volume of gas to an accuracy of about 0.2 percent.

One of the precautions taken was the measurement of the stray heat input to the calorimeter before and after each data point. The approach to temperature equilibrium of the calorimeter was observed after each heating interval. As T_c was