# COMMENTS ON THE PAPER, "EVIDENCE FOR A NEW PHASE OF SOLID $He^3$" +)

by

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The interpretation of Osheroff, Richardson and Lee of their recent measurements of the solidification pressure of $He^3$ is reexamined. On the basis of a different assumption about the thermal coupling in the solid, it is concluded that the transition is unlikely to be of first order and is more likely the expected lambda transition to the ordered antiferromagnetic phase.

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In a recent publication[1], Osheroff, Richardson and Lee (ORL) have presented new and very interesting measurements of the solidification pressure of $He^3$ below 3 mK. They also give an interpretation of their results leading to the suggestion that at T≈2.7 mK, solid $He^3$ undergoes a first-order phase transition to a new phase whose entropy behavior, to use their own words, is unprecedented. In the present work we re-analyze their data and come to the conclusion that a more likely explanation is that the system undergoes the heretofore expected lambda transition to an ordered magnetic phase. To support our conclusions, we present calculations which yield a qualitative picture of the entropy and heat capacity of solid $He^3$ in the neighborhood of the transition from their experimental data.

We begin our discussion by deriving the basic thermodynamic equations which describe the experimental situation. We assume a quasi-stationary process, so that we may write

$$dP/dt = (dP/dT)*(dT/dt) \quad , \tag{1}$$

where dP/dT, the slope of the melting curve, is given by the Clausius-Clapeyron equation

$$dP/dT = - (S_{solid} - S_{liquid})/\Delta V \ . \tag{2}$$

Here $\Delta V = V_{liquid} - V_{solid} = 1.27$ cm$^3$/mole (it has been found to be nearly constant under the conditions of the experiment[2,3]) and $S_{solid}$ and $S_{liquid}$ are molar entropies. From the condition that the compression (or expansion) be adiabatic, one finds, in general

$$\frac{dT}{dt} = \frac{-T(S_{solid} - S_{liquid})}{[n_o - n(t)]\, C_{liquid} + n(t) C_{solid}} \frac{dn}{dt} \quad , \qquad (3)$$

where $n_o$ is the total number of moles of $He^3$, $n(t)$ is the number of moles of solid at time $t$, $C_{liquid}$ and $C_{solid}$ are molar heat capacities and $dn/dt$ is a constant for the given experimental conditions.

In their analysis, ORL assume that the solid once formed is decoupled from the system because of long thermal relaxation times; this is described by Eq. (3) without the term containing $C_{solid}$. At the transition $S_{solid} >> S_{liquid}$, so that they find from their experiments using Eqs. (1), (2) and (3) that

$$(dP/dt)_{above}/(dP/dt)_{below}$$

$$= (S_{solid\ above}/S_{solid\ below})^2 = 1.8,$$

which gives $S_{solid\ above} = (4/3)\, S_{solid\ below}$. Since equilibrium measurements[3] yield $S_{solid\ above} = 0.6$ R,

they find $S_{solid\ below} = 0.45\ R$. Their interpretation runs into difficulty if they integrate Eq. (2) down to zero temperature. They find

$$(\Delta P \Delta V)/R = \int_0^{T_{trans}} dT(S_{solid}/R) = 0{,}81\ mK,$$

which can be satisfied only if the entropy stays close to 0,45 R for most of the temperature range below 2.7 mK. This result is, as stated by ORL, strikingly abnormal behavior. We wish, therefore, to call into question the basic assumption of their interpretation, namely that the solid in their experiment stays at the temperature at which it was formed; i.e. that it is thermally decoupled from the system.

Although the spin-lattice relaxation time $T_1$ in the solid is increasing rapidly at low temperatures, the spin-spin relaxation time $T_2$ appears to stay at a constant value of about 0.1 $sec^4$. One would therefore expect that the spin system of the solid, which carries essentially all the entropy at these temperatures, must participate in the process of reaching equilibrium. Of course the lattice is essentially inert. It is then no longer justifiable to neglect $C_{solid}$ in Eq. (3) and a jump in this quantity will produce a discontinuity in dT/dt which in turn shows up in dP/dt even if dP/dT were continuous across the transition. This point of view is strenghened by the fact

that $dP/dT_{Pt}$ extracted from Fig. 3 of ORL shows no significant discontinuity across the transition.

Let us further consider Fig. 2 of ORL. Although the pressures measured at points A and B are the same both upon compression and expansion, the behavior of dP/dt is much more complicated. The relative change in slope near A and A' is the same; however, apart from the relative change, the slope is quite different - this is seen most clearly near the point C. These features lead one to the conclusion that there is a considerable amount of non-equilibrium behavior associated with the measurement as was clearly stated by ORL. Thus, although we believe that there is some cooling of the spin system of the solid after it is formed, it seems likely that the solid is not in true equilibrium.

To consider these points further, we have made several calculations of the entropy and heat capacity of the solid using the dP/dt data of ORL and Eqs. (1), (2) and (3). Typical results are shown in Fig. 1. In one calculation we assumed that the solid was in equilibrium with the liquid. In this way, we could calculate self-consistently the entropy and heat capacity of the solid. The results are shown in the curves labeled (a) in Fig. 1. To simulate the assumption that the solid was only in partial equilibrium

with the liquid, we also calculated the entropy and heat capacity self consistently assuming that only a small fraction of the solid (only a few percent of the total amount of $^3$He) should be included in Eq. (3). These results are shown as the curves labeled (b) in Fig. 1.

A glance at Fig. 1 shows that both curves (a) and (b) have the same qualitative features. In the first place, the heat capacity looks just like that of a second-order transition in the Ehrenfest sense. Such a result is quite reasonable as it is well known that lambda-transitions mimic second-order transitions if one goes through them fast enough. Further, in both cases the entropy is much too high. Again, this is consistent with non-equilibrium behavior, in which case all of the entropy would not be extracted by the compression.

We wish to comment further on the shape of the P(t) curve found by ORL. A lambda-transition is to be expected and it should give a P(t) curve with much more curvature in the neighborhood of the transition. We believe, in fact, that if the measurements were done under equilibrium conditions, that this would be observed above the transition. In fact, there is already some curvature there, although it is small. However, as can be seen from our calculations, quite

a large curvature in the heat capacity below the transition will give essentially linear behavior for P(t). This result is due to cancellations in Eq. (3) which will hold even if the measurements were done in a quasi-stationary manner. We also wish to point out that a first-order transition with some thermal coupling to the solid would lead to dP/dt = 0 over a finite time interval at the transition.

Finally, we wish to comment on the measured value of the transition temperature. It has been expected, on the assumption that solid $He^3$ would be a good spin-(1/2) nearest neighbor Heisenberg antiferromagnet, that the transition temperature would be 2.0 mK. However, the measurements of Kirk and Adams[5] have shown that this is certainly not the case. Recently, Zane[6] has suggested that three-body exchange, which would yield an effective next-nearest neigbor exchange interaction that is ferromagnetic in sign could account for the Kirk-Adams results. Such a theory would also yield a transition temperature higher than 2.0 mK.

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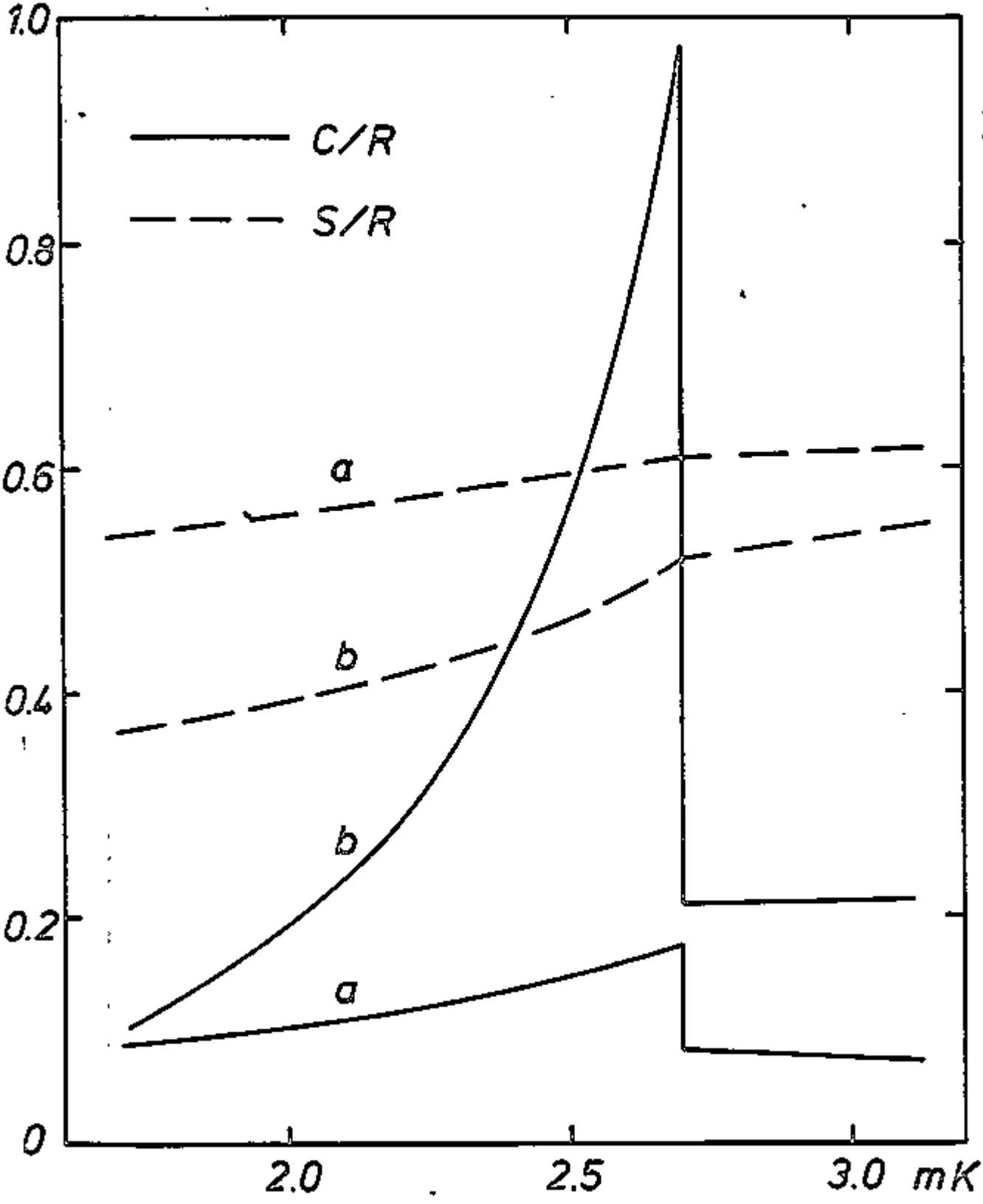
FIGURE CAPTION

Fig. 1 Plots of the entropy and heat capacity of the solid as a function of temperature in the region of the transition as calculated from the data of ORL. For curve (a) true equilibrium of the solid was assumed; whereas, for curve (b) partial equilibrium was assumed.


1.0
0.8
0.6
0.4
0.2
0
C/R
S/R
a
b
b
a
2.0
2.5
3.0 mK