## Comments on "Evidence for a New Phase of Solid He<sup>3</sup>"\*

H. Horner and L. H. Nosanow<sup>†</sup>

Institut für Festkorperforschung, Kernforschungsanlage, Jülich, West Germany (Received 22 May 1972)

The interpretation of Osheroff, Richardson, and Lee of their recent measurements of the solidification pressure of He<sup>3</sup> 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.

In a recent publication,<sup>1</sup> Osheroff, Richardson, and Lee (ORL) have presented new and very interesting measurements of the solidification pressure of He<sup>3</sup> below 3 mK. They also give an interpretation of their results leading to the suggestion that at  $T \approx 2.7$  mK, solid He<sup>3</sup> 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 reanalyze 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<sup>3</sup> 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 quasistationary process, so that we may write

$$dP/dt = (dP/dT)dT/dt,$$
(1)

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

$$dP/dT = -(S_{\rm sol} - S_{\rm lig})/\Delta V.$$
<sup>(2)</sup>

Here  $\Delta V = V_{1iq} - V_{sol} = 1.27 \text{ cm}^3/\text{mole}$  (it has been found to be nearly constant under the conditions of the experiment<sup>2,3</sup>) and  $S_{sol}$  and  $S_{1iq}$  are molar entropies. From the condition that the compression (or expansion) be adiabatic, one finds, in general

$$\frac{dT}{dt} = \frac{-T(S_{\rm sol} - S_{\rm lig})}{[n_0 - n(t)] C_{\rm lig} + n(t) C_{\rm sol}} \frac{dn}{dt},$$
(3)

where  $n_0$  is the total number of moles of He<sup>3</sup>, n(t) is the number of moles of solid at time t,  $C_{\text{liq}}$  and  $C_{\text{sol}}$  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  $C_{\rm sol}$ . At the transition  $S_{\rm sol} \gg S_{\rm lig}$ , so that they find from their experiments using Eqs. (1), (2), and (3) that

 $(dP/dt)_{above}/(dP/dt)_{below}$  $\simeq (S_{sol above}/S_{sol below})^2 \simeq 1.8,$ 

which gives  $S_{sol\ above} \simeq \frac{4}{3}S_{sol\ below}$ . Since equilibrium measurements<sup>3</sup> yield  $S_{sol\ above} \simeq 0.6R$ , they find  $S_{sol\ below} \simeq 0.45R$ . Their interpretation runs into difficulty if they integrate Eq. (2) down to zero temperature. They find

$$\frac{\Delta P \Delta V}{R} = \int_0^{T_{\rm tr ans}} dT \, \frac{S_{\rm scl}}{R} = 0.81 \, {\rm mK},$$

which can be satisfied only if the entropy stays close to 0.45R 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 diffusion constant<sup>4</sup> stays constant at a value around  $10^{-7}$  cm<sup>2</sup>/sec. Despite the smallness of this value we expect that at least a fraction of 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  $C_{sol}$  in Eq. (3), and a jump in this quantity will produce a discontinuity in dT/dtwhich in turn shows up in dP/dt even if dP/dTwere continuous across the transition. This point of view is consistent with 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 nonequilibrium 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. Starting at some initial temperature (5 to 10 mK) and using typical values for the fraction of the solid and the rate of compression given by ORL, we have integrated Eqs. (1), (2), and (3) to lower temperatures. Typical results are shown in Fig. 1. In one calculation we assumed that the solid was in equilibrium with the liquid. The resulting entropy and heat capacity of the solid 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 assuming that only a small fraction of the solid (only a few percent of the total amount of <sup>3</sup>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 aand 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 secondorder transitions if one goes through them fast enough. Further, in both cases the entropy is much too high. Again, this is consistent with nonequilibrium 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



FIG. 1. Entropy and heat capacity of the solid as functions 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.

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 cancelations in Eq. (3) which will hold even if the measurements were done in a quasistationary 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<sup>3</sup> would be a good spin- $\frac{1}{2}$  nearest-neighbor Heisenberg antiferromagnet, that the transition temperature would be 2.0 mK. However, the measurements of Kirk and Adams<sup>5</sup> have shown that this is certainly not the case. Recently, Zane<sup>6</sup> has suggested that three-body exchange, which would yield an effective next-nearest-neighbor exchange interaction that is ferromagnetic in sign could account for the Kirk-Adams results. Such a theory would also yield a transition temperature

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higher than 2.0 mK.

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## Nonsingular S = +1 Screw Disclination Lines in Nematics

C. Williams,\* P. Pierański, and P. E. Cladis Physique des Solides,†Université Paris-Sud, 91-Orsay, France (Received 10 April 1972)

We present experimental evidence to support the nonsingular model for screw disclinations of strength S = +1.

Constraining a nematic in a long cylinder so that the director has only a radial component (perpendicular to the cylinder axis) results in a splay deformation energy<sup>1</sup> which has a logarithmic divergence as  $r \rightarrow 0$  (r being the distance from the cylinder axis). This necessitates the introduction of a core region where the approximation of linear elasticity breaks down. It has recently been shown that the divergence in the splay energy can be avoided by the addition of a finite amount of bend deformation.<sup>2</sup> The resulting director field has a radial component which decreases as  $r \rightarrow 0$  but a z component (parallel to the cylinder axis) which increases as  $r \rightarrow 0$ . The net energy is then finite, smaller than a pure splay configuration, and does not require a core. Fig. 1(a) shows the theoretical configuration in the case of methoxybenzilidenebutylaniline (MB-BA) for which we have taken  $K_{11}/K_{33} = 0.7$ ;  $K_{33}$  is the bend elastic constant and  $K_{11}$  the splay elastic constant.1

This Letter gives experimental evidence which supports the nonsingular model.

A thin film of hexadecyltrimethyl ammonium bromide on a clean glass surface orients the director of MBBA molecules perpendicular to the surface. We have treated the inside surface of a glass capillary tube such that an S = +1 disclination line (rotation of  $2\pi$  of the director along a path coinciding with the boundary, r = R) is generated. The tube was then inserted in a layer of optical epoxy resin between two glass plates to avoid optical effects due to its cylindrical shape. The tube was studied with its axis perpendicular



FIG. 1. (a) The molecular orientation around a radial S=+1 line deduced from Cladis and Kleman's (Ref. 2) model for MBBA  $[K_{33}=8\times10^{-7}$  (cgs),  $K_{11}/K_{33}=0.7]$ . (b) Experimentally deduced values for  $\varphi$  as found from measuring the inclination of the extraordinary image with respect to the ordinary image.