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OF He⁴

Berkeley, California

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THE HEAT CAPACITY OF THE γ PHASE OF He⁴ *

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About two years ago Vignos and Fairbanks reported a new solid phase, identified as the γ phase, in He⁴,¹ and soon thereafter Schuch and Mills established that this phase has a body-centered cubic (bcc) structure.² Recently Grilly and Mills established the PVT relations for the transition from the α to the γ phase, and from the α and γ phases to the liquid.³ The α phase in He⁴ has an hexagonal close-packed (hcp) structure. Both an hcp and a bcc structure also exist in He³, and the heat capacity of both of these phases has been measured by Heltemes and Swenson for this isotope.⁴ The Debye θ of the bcc phase was found to be about 20% smaller than that of the hcp phase at the same density, the isotope effect on θ was observed to be only slightly larger than the $m^{0.5}$ ratio in the hcp phase, and an anomaly of unknown origin was found in the heat capacity of the bcc phase. The work presented here was undertaken primarily to determine if a similar difference in the lattice heat capacity of the two phases also exists in He⁴, to determine the isotope effect in the bcc phase, and to see if a similar anomaly exists in the heat capacity of γ He⁴.

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Experimental investigations of the properties of the γ phase of He^4 are difficult because of the small volume and temperature range over which this phase exists. In order to measure the heat capacity, temperature increments of 1 or 2×10^{-3} $^\circ\text{K}$ had to be used. It was nonetheless possible to obtain a precision of the order of 1% in the heat capacities by using a germanium thermometer which was calibrated against the vapor pressure of He^4 . The sample was introduced into a beryllium-copper container through a stainless steel capillary at 4.2 $^\circ\text{K}$. The capillary was then blocked by cooling the bath, to which it was attached, to 1.2 $^\circ\text{K}$. Thereafter the sample was cooled by means of a mechanical heat switch.

It is an inherent limitation of this method that only molar volumes less than 20.98 cc are accessible to measurements. For any greater volume there is no freezing in the capillary, and the sample cannot be kept at constant volume. For this reason the data obtained here are limited to the higher-density region of the γ phase.

In view of the small range of densities involved in these measurements, it was not possible to make a direct determination of the sample density with meaningful accuracy. Rather the molar volume was inferred from the α -to-liquid or γ -to-liquid transition temperature and the data of Grilly and Mills.³

Some of the experimental results in the α phase and most of the results in the γ phase are presented in Figure 1. Within experimental accuracy, the Debye θ of the α phase decreases linearly with increasing temperature over the temperature range covered here. The Debye θ of the γ phase is 16.9 $^\circ\text{K}$. One set of measurements on the α phase was made at a molar volume of 19.87 cc,

at which direct comparison with existing data is possible. The data presented here agree well with those of Keesom and Keesom⁵, but differ from those of Webb et al.⁶ by about 12% in the heat capacity. The heat capacity of solid He⁴ has also been measured by Dugdale and Simon⁷ and by Heltemes and Swenson.⁴ However, Dugdale and Simon's data are at smaller molar volumes, and comparison is difficult. Heltemes and Swenson's data include the molar volume range studied here, but are supposedly reliable only below 1.1 °K. Thus comparison is again not straightforward. Since the θ values reported herein are temperature-dependent and only extend down to 1.3 °K or $(T/\theta) \approx 0.052$, an extrapolation to 1.1 °K or $(T/\theta) \approx 0.044$ must be made. Comparison with Debye θ values of other substances on which accurate measurements are available over a large temperature range shows that a linear extrapolation in this relative temperature range is not likely to be in error by more than 1 or 2%. This type of extrapolation agrees well with Heltemes and Swenson's data at 20.9 cc/mole, but differs from their data by 4% in θ at 19.87 cc/mole.

In order to compare the Debye θ values of the α and the γ phases at the same molar volume, again an extrapolation is necessary because the two-phase temperature range at constant volume is about 0.2 °K. Extrapolation from $(T/\theta) \approx 0.060$ to $(T/\theta) \approx 0.067$ must be made in this case, and again a linear extrapolation for the α phase is not likely to introduce a prohibitive error. This extrapolation yields a difference between the Debye θ values for the two phases of 24%, which is about the same as that found in He³ for the phases of the same crystal structures. It therefore appears that the larger lattice heat capacity of the bcc phase is not a peculiarity of He³, but rather that it is associated with the structure of the lattice.

Heltemes and Swenson pointed out that the ratio of the Debye θ values for the two isotopes below 1.1 $^{\circ}$ K in the hcp phase is 1.19, which is only slightly higher than $(4/3)^{0.5} = 1.16$. A similar comparison for the bcc phase is difficult because the He^4 γ phase can be investigated only at $(T/\theta) \approx 0.1$, and the data on He^3 refer to relatively much lower temperatures. The temperature dependence of θ in He^4 is not established at all. But again comparison with other solids shows that θ is in general close to a minimum at $(T/\theta) \approx 0.1$, and that it is between 8 and 15% lower than at 0 $^{\circ}$ K. One can thus estimate that, if the phase were stable, it would have a Debye θ of 18.4 to 19.9 $^{\circ}$ K at 0 $^{\circ}$ K. Comparison with Heltemes and Swenson's value for bcc He^3 gives a ratio of 1.33 to 1.23 for the Debye θ 's of the two isotopes in the bcc phase. Whereas the uncertainties in the above comparison are rather large, there is a definite indication that the Debye θ ratio is larger than that indicated by simple theory. A similar effect has been observed in solid hydrogen and deuterium, where the ratio is 1.6 rather than 1.4.⁸ It seems reasonable to attribute the effect to the large anharmonicity due to the large zero-point energy.

An anomalous contribution to the heat capacity of bcc He^4 was not observed in this work. Of course the existence of an anomaly can not be ruled out entirely, because of the rather small temperature range over which the phase can be investigated. In bcc He^3 at a molar volume of 20.9 cc Heltemes and Swenson's data do not in themselves establish the existence of an anomaly. The deviation of the heat capacity from a Debye function can in this case be adequately explained in terms of the natural deviation of the Debye θ from a constant value and is indeed quite similar to that found in hcp He^4 at the same

volume and Debye θ . Only at much larger molar volumes does this deviation become larger than would ordinarily be expected, and there is some support for postulating an anomaly. If there were an anomalous contribution to the heat capacity in both isotopes, then the isotope effect would be more uncertain, and could be closer to the $m^{0.5}$ value. If one attributes the total heat capacity to the lattice for the bcc phase of both isotopes, then the isotope effect as reflected in the θ ratio at 1.70 °K is 1.23, which is still considerably larger than the $m^{0.5}$ ratio. The curve for θ vs T corresponding to Heltemes and Swenson's total heat capacity at 20.9 cc/mole is shown in Figure 1 for comparison.

At present neither the data on He³ nor those on He⁴ are sufficient to clearly establish the relative magnitudes of possible anomalous and lattice contributions to the heat capacity of the bcc phases of the two isotopes. In He⁴ there is not much hope for improvement because of the natural limitations set by the existence range of the phase. The bcc phase of He³ can perhaps be studied more thoroughly, and one would hope that more precise data over a larger temperature range would shed some light on the problem.

Considerable information was gained during this work on the detailed behavior of He⁴ along the various phase boundaries. In particular, the difference between the λ point in the liquid at the melting line and the upper triple point was found to be 10×10^{-3} °K, in agreement with Vignos and Fairbanks¹ and in disagreement with Grilly and Mills.³ Work on the $\alpha - \gamma$ two-phase region is still in progress, and a comprehensive report on all the work will be given elsewhere.

The author is indebted to Professor Norman E. Phillips for many stimulating discussions during the course of this work.

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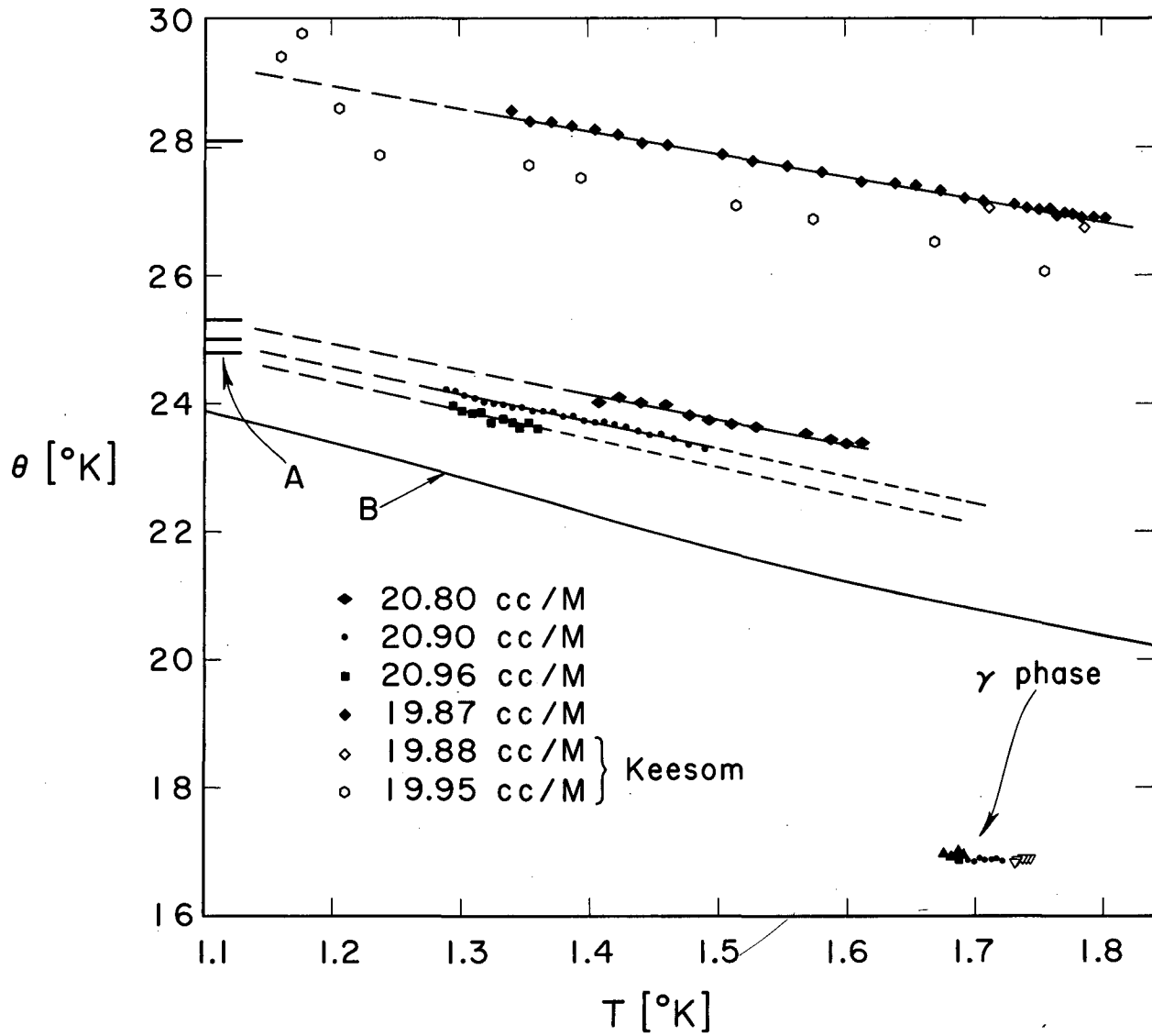
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Figure Caption

Figure 1. Debye theta vs temperature for the α and γ phases of solid He⁴.

A indicates the values obtained by Heltemes and Swenson for the volumes of He⁴, used in this experiment and

B indicates the Debye θ calculated from Heltemes and Swenson's total heat capacity for bcc He³ at 20.9 cc/mole.



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Fig. 1

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