

# THE INFLUENCE OF $^3\text{He}$ ON THE LAMBDA TEMPERATURE

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**ABSTRACT** The observed change in the lambda transformation temperature with the concentration of  $^3\text{He}$  in  $^4\text{He}$  gives for low concentrations a negative linear shift of 1.48 degrees per mole concentration. The lambda transition is of the second order and there seems to be no appreciable change in the jump in the specific heat at the lambda point.

## INTRODUCTION

The interesting phenomenon of  $^3\text{He}$  not taking part in superfluid flow, observed by Daunt, Probst *et al.*, (1948) in dilute solutions of  $^3\text{He}$  in  $^4\text{He}$ , led to a new method of isotopic separation in the liquid phase by superfluid filtration and also to the conclusion that the lambda temperature of liquid  $^4\text{He}$  would be a function of the concentration of  $^3\text{He}$ . Accordingly the presence of  $^3\text{He}$  has a pronounced effect on the value of the lambda point, which is shifted towards lower temperature as was first shown by Abraham, Wemstock *et al.* (1949) for a mixture with  $^3\text{He}$  concentrations upto 28.2%. Similar experiments were performed by Eselsohn and Lazarev (1950) for a mixture with a concentration of about 1.5% and by Daunt and Heer (1950) for mixtures with high concentrations of 42% to 89%.

The specific heat measurements of the liquid mixtures of  $^3\text{He}$  in  $^4\text{He}$  have been made by the helium group of the Kamerlingh Onnes Laboratory, Leiden. Some of those results have already been published (Dokoupil, Kapadnis *et al.*, 1954 and 1959, Kapadnis and Dokoupil, 1955). In the present investigations the values of the lambda temperature of the mixture for low concentrations of  $^3\text{He}$  in  $^4\text{He}$  have been computed. For the sake of comparison numerical computations of the lambda temperatures line have also been made from the theories in vogue. The final check of the exact determination of the composition of the samples of the mixtures and its confirmation delayed appreciably these computations and the discussion of the results. In the mean time the temperature scale in the helium region has been changed. Therefore we have recalculated all our data using the new scale adopted internationally.

## EXPERIMENTAL

As the quantity of  $^3\text{He}$  at the disposal of the Leiden helium group was then very small, the method of a closed capsule completely filled with the condensed

mixture was developed and applied to the calorimetric determination of the specific heat (Dokoupil, Kapadnis *et al.*, 1954 and 1959; Kapadnis and Dokoupil, 1955). Special attention was paid to the accurate determination of the transition temperature. The temperature interval in which the lambda point of the mixture must lie, was first determined by a close scrutiny of the experimental points representing the specific heat of the mixture in the vicinity of the transition temperature. For the determination of its exact value separate runs through the transition point were made with a continuous heat supply, sometimes by the natural warming up only (especially when the bath level was very low). The values for the lambda temperature of the mixture were computed from the resulting discontinuities in the slopes of the corresponding curves of temperature versus time. Figure 1 shows such a typical plot used to compute the lambda temperature of the mixture.

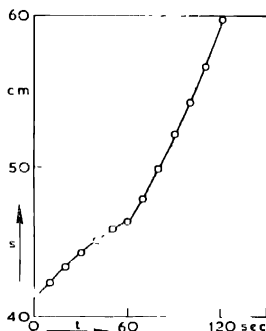


Fig 1 A typical plot of the Galvanometer reading vs Time ( $X=0.010$ )

#### RESULTS AND DISCUSSION

The values of the lambda temperature corresponding to the concentration of  $^3\text{He}$  in  $^4\text{He}$ ,  $X = 1.00, 2.50, 7.13, 21.1$  and  $41.7\%$  are  $T_\lambda^m = 2.162, 2.140, 2.072, 1.845$  and  $1.454^\circ\text{K}$  respectively, giving for low concentrations a negative shift of  $1.48$  degrees per mole concentration, which is linear. The lambda transition is of the second order and there seems to be no appreciable change in the jump in the specific heat at the lambda point.

The early experimental observations of the lowering of the lambda point of  $^4\text{He}$  due to an admixture of  $^3\text{He}$ , reported by Abraham *et al.*, (1949) for concentrations upto  $28.2\%$   $^3\text{He}$  by using a superleak formed around a platinum wire, seem to be largely in error, presumably due to the concentration gradient set up in the liquid mixture as a result of the heat flush effect. The value given for the lambda temperature for the concentration of about  $1.5\%$   $^3\text{He}$  by Eselsohn and Lazarev (1950)

is also somewhat low, probably due to the uncertainty of the concentration. They used an apparatus consisting of two reservoirs connected by a narrow capillary and made use of an observation of the supra-surface flow as the criterion for the existence of the lambda temperature. Daunt and Heer (1950) used in their experiments for concentrations from 42% to 89%  $^3\text{He}$ , the same criterion and determined the lambda temperatures at which the heat influx to the reservoir changed abruptly. They expected that their results were not seriously affected by the heat flush effect.

The experimental results of King and Fairbank (1953) using the disappearance of second sound at the lambda point as a criterion, for concentrations below 4.2%  $^3\text{He}$  revealed a linear dependence of the lambda temperature of the mixture on the concentration of  $^3\text{He}$ , giving negative slope of 1.5 degrees per mole concentration. These observations led to the suspicion that the earlier observations were erroneous. It was confirmed by our measurements of the specific heats of liquid mixtures of  $^3\text{He}$  and  $^4\text{He}$  and later on by the oscillating pendulum experiments performed by Dash and Tailor (1955), who obtained from their measurements of the behaviour of a torsion pendulum immersed in the liquids, transition temperatures of the individual solutions by locating the discontinuity in slope of the torsion period versus temperature. This is equivalent to finding the temperature at which the normal fluid density becomes equal to the total density. Their experiments also gave a negative linear shift of  $1.47 \pm 0.03$  degrees per mole concentration for the whole concentration range (upto  $X = 0.092$ ) they studied.

It may be pointed out that in the experiments of King and Fairbank, those of this research and of Dash and Tailor the principles involved in the methods to determine the lowering of the lambda point of liquid  $^4\text{He}$  due to the presence of  $^3\text{He}$  in the mixture, are entirely different from each other. In spite of that the excellent agreement of the results suggests that for low concentrations at least the observed negative shift of 1.48 degrees per mole concentration may be treated as an established fact.

Figure 2 shows the ratio of the lambda temperature of the mixture to that of pure  $^4\text{He}$  as a function of the concentration of  $^3\text{He}$  upto  $X = 0.11$ . The observed data as well as the numerical evaluations made from different theories in vogue have been included in the figure.

It is evident from the work of Stout (1948) that the Gibbs' function written according to the laws for perfect classical solutions leads to the second order lambda transition for liquid  $^4\text{He}$ , the lambda point of liquid  $^4\text{He}$  remaining unaffected by the admixture of  $^3\text{He}$ . In order to account for the experimentally observed change in the lambda transformation temperature with the concentration of  $^3\text{He}$ , using classical thermodynamics it is necessary to inject a reflection of the quantum nature of liquid helium through some experimentally observed phenomenon. It was accounted for, in the theory of de Boer and Gorter (1950) by introducing the

hypothesis put forward by Taconis *et al.* (1949), namely that the  $^3\text{He}$  can be regarded as being in solution with the 'normal' constituent of helium II only. The lambda temperature of the mixture in this case is given by

$$1 - X = \exp [ S_\lambda(T - T_\lambda)/RT ]$$

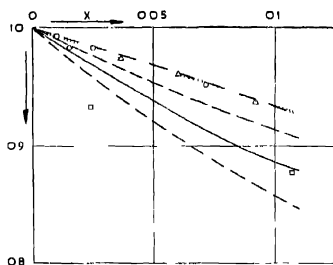


Fig. 2.  $T_\lambda^m$  vs. Concentration of  $^3\text{He}$  in  $^4\text{He}$

- This Research,
- Abraham *et al.*,
- △ Dash and Taylor, and
- ◇ Eiselsohn and Lazarev
- de Boer and Gorter.
- Heer and Daunt;
- Mikura,
- Nanda.

For given  $X$  the value of  $T$  satisfying the above expression corresponds to  $T_\lambda^m$ , the lambda temperature of the mixture. The choice of the Gibbs' function made in this theoretical model does not seem to be the proper one. The expression one gets after making use of the functional dependence of the Gibbs' function for pure  $^4\text{He}$  on the normal fluid fraction gives temperature independent entropy for liquid helium I. This implies that liquid helium I has zero specific heat. Moreover, the Gibbs' function for the  $^4\text{He}$  component as a function of temperature (linear or quadratic in  $T$ ) and the normal fluid fraction of the total number of  $^4\text{He}$  atoms in their theory does not help much in bringing the theoretical  $T_\lambda^m - X$  curve in close agreement with the experimentally observed values. The theory also gives much higher values for the initial slope  $\left(\frac{\partial T_\lambda^m}{\partial X}\right)_{X \rightarrow 0}$  in both the cases of the Gibbs' function for the  $^4\text{He}$  component. The modification of this theory made by Nanda (1955) leads to

$$1 - X = \exp \left[ -\frac{1}{3} \left( \frac{T_\lambda}{T} \right)^{1/6} - \left( \frac{T}{T_\lambda} \right)^{1/6} + \frac{3WX^2}{RT} \right]$$

giving the variation of the lambda temperature with the concentration of  $^3\text{He}$ . This expression, as seen from the figure 2, fails to improve the

situation. An approach to this problem was made by Trikha and Nanda, with the assumption that  $^3\text{He}$  dissolves in  $^4\text{He}$ , normal as well as superfluid. They substituted the experimentally observed value of the initial slope in their expression for the lambda temperature of the solution in order to assign a value to the parameter called the interchange energy. This adjustment did not lead to the desired agreement with the experimental results. An appropriate modification of the form of the Gibbs' function for pure  $^3\text{He}$  or an entirely different approach to introduce the quantum nature of the liquid in the classical thermodynamic treatment is necessary.

Starting from an interpretation of the lambda point in liquid helium as a consequence of the Bose-Einstein condensation, Heer and Daunt (1951) put forward the idea that the behaviour of the mixtures of  $^3\text{He}$  and helium II could be described as an ideal mixture of a degenerate Bose-Einstein gas and a nondegenerate Fermi-Dirac gas. Thus, they assumed the statistical independence of the Bose-Einstein and the Fermi-Dirac systems in solutions of  $^3\text{He}$  and  $^4\text{He}$  and showed that the lambda temperature of the mixture is given by the degeneracy temperature of the Bose-Einstein systems. The expression for the lambda temperature of the mixture in this case is

$$\frac{T_\lambda^m}{T_\lambda} = \left[ \frac{1-X}{1+X\left(\frac{V_3^0}{V_4^0}-1\right)} \right]^{2/3}$$

The agreement with the experimental  $T_\lambda^m$  of the mixture improves a bit in this case as seen from figure 2. However the theory was criticized by de Boer and Gorter

(1952), who showed that the distribution function  $\frac{C_V}{C_L}$  must have a discontinuity at the lambda point as a consequence of the finite jump in the specific heat of liquid helium at this temperature without regard to the model used, in opposition to the results of Heer and Daunt. Mikura (1954 and 1955) modified this theory by introducing a Bose-Einstein liquid model for liquid helium II in which He II is composed of Bose-Einstein particles in a smoothed potential well. Between the ground state and the first excited state there is an energy gap  $K\Delta$ ,  $k$  being the Boltzmann constant, and the total number of particles are clustered together in groups of atoms. He introduced  $^3\text{He}$  in this system as an ideal Fermi-Dirac gas in a smoothed potential well. It appeared necessary to introduce still another assumption

$$\Delta = \Delta_0 N_4 V_4^0 / (N_3 V_3^0 + N_4 V_4^0)$$

regarding the dependence of  $\Delta$  on  $^3\text{He}$  concentration to get agreement with experimental data on the lambda temperature of the mixtures. The refinement of the treatment made by Mikura leads to a very close, rather a very good fit of his  $T_\lambda^m$  vs  $X$  curves with the experimental data, as a result of the adjustment of the

parameters in his theory to that effect. The lambda temperatures of the mixtures are given in this case, by the expression

$$\frac{T_{\lambda}^m}{T_{\lambda}} = \left[ \frac{1-X}{1+X} \left( \frac{V_3^0}{V_4^0} - 1 \right) \cdot \frac{F_{3/2}(\Delta_0/T_{\lambda})}{F_{3/2}(\Delta/T_{\lambda})} \right]^{2/3}$$

One gets according to Pomeranchuk's model (1949) a negative shift of about 0.8 degrees per mole concentration, by using the effective mass derived from second sound experiments and taking for the lambda point the temperature at which the mass of the excited particles becomes equal to the total mass. (This value is rather too low)

The experimental lambda temperatures are in very good agreement with

$$T_{\lambda}^m = T_{\lambda} (1-X)^{2/3}$$

given by Goldstein (1954) from the asymptotic Bose-Einstein model by neglecting the effect of  $^3\text{He}$  on the total liquid density.

A number of phenomenological and molecular theories are available at present. The phenomenological approach made by de Boer and Gorter does not give any clue to the understanding of the molecular origin of the deviations from the laws of perfect solutions. A different choice of the Gibbs' function and an introduction of a non-ideality parameter make no appreciable improvement in the original theory. The root cause of the disagreement with the experiments seems to lie in the basic assumptions of the theory.

The theories based on the difference in the statistics are also unsatisfactory, though the approach made by Mikura leads to rather very good agreement with the experimental results. The parameters involved in this model render it rather easy to arrive at the desired results by their suitable adjustment. One can also obtain the results in reasonable agreement with the experiments, simply by making  $S_{\lambda}$  and either  $E$  or  $n$  concentration-dependent.

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