Measurement of the Heat of Transition of He^3 from one Liquid Phase of an He^3-He^4 Solution to Another

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The measurement of the heat of transition of He^3 from a solution phase enriched in He^3 to a phase enriched in He^4 has been carried out in the temperature range 0.02 to 0.70°K. Below 0.05°K, the heat of transition is equal to $(80\pm3)T^2$[J/mole−°K]; for 0.54°K, it reaches a maximum value of 4.5 J/mole. The heats of transition are in good agreement with the available data on the specific heats of He^3, He^4, and solutions of He^3 in He^4, and also with the solution stratification curve.

KNOWLEDGE of the heat of transition of He^3 from a solution of one concentration into a solution of a different concentration is of interest since, like the stratification curve, it connects the thermodynamic quantities in the equilibrium states of the two phases. Moreover, knowledge of the heat of transition is necessary for calculations of the low-temperature efficiency of solution cryostats.

The heat of transition is defined as the heat q which must be supplied to the upper, He^3-enriched phase in order to transfer one mole of He^3 from the upper phase to the lower, which is He^4 enriched, at constant temperatures, pressures and concentrations of both phases, i.e., $q = \delta Q/\delta n_3$, where δn_3 is the number of moles of He^3 which move from one phase to the other, and δQ is the heat necessary for this transfer. It must be noted that such a process occurs under equilibrium conditions only because the lower phase of the He^3 $= He^4$ solution is a superfluid and therefore the removal of atoms of He^3 and the delivery of atoms of He^4 takes place without entropy loss, as the counterflow of the normal and superfluid parts of the solution.

The measurement of the heat of transition was carried out in a He^3–He^4 solution cryostat by a method similar to that described earlier.[1] The bath of the solution had a volume of 18 cm^3 and was connected to the vaporization bath by coaxial German-silver tubes of length 15 cm, which went from the bottom of the solution both to the bottom of the vaporization bath. In the circulation mode, the returning flow of He^3 passed along the inner tube, of diameter 0.2 mm, with a wall thickness of 0.15 mm. In the one-shot operation, the He^3 contained in the tube (~1–2% of the amount of He^3 in the solution bath) was rapidly transferred to the lower phase of the solution bath and did not have any effect on the further process. The outer tube, along which He^3 moved into the evaporation chamber, had an inner diameter of 1 mm and a wall thickness of 0.3 mm.

In the measurement of the heat of transition, an amount of the upper phase in the solution bath was brought to the necessary value by circulation of the He^3, and cooling to the required temperature was then carried out. The minimum temperature reached in the circulation was 0.85°K. The circulation was then stopped, a heater located in the upper solution chamber was switched on, and pumping of the gas evaporating from the chamber in the measurement volume was begun. The heater produced a heat flow that was homogeneous over the cross section of the chamber. The heater current was regulated so that the temperature of the lower phase in the solution chamber remained constant. At temperatures above 0.2°K, because of the very small thermal conductivity in the upper phase, significant temperature gradients developed, especially in the vicinity of 0.4°K. Therefore, each experiment was carried out until the upper phase was completely exhausted, and the heat of transition was determined from the value of the power of the heater at the moment of disappearance of the upper phase, as revealed by the sharp decrease in the power needed to maintain the same temperature in the solution chamber.

According to estimates, the parasitic heat influx into the solution chamber was less than 3×10^{-8} W, which, at very low temperatures, was less than 1.5% of the released power. The rate at which the helium was drawn from the vaporization chamber ranged from 7×10^{-8} mole/sec at 0.02°K to 1.8×10^{-5} mole/sec at 0.7°K, owing to the longer times required to establish equilibrium at the high temperatures. The amount of He^3 transferred from the upper phase to the lower was determined by the number of moles of helium which entered the measurement volume and by its concentration. However, the obtained relation was not linear.

We assume that a volume δv_1 is transferred from the upper phase to the lower in the solution chamber; then the volume in the vaporization chamber decreases by δv_2 and δn_3 moles of He^3 and $\delta\bar{n}_4$ moles of He^4 enter the measurement volume. One can then show that $\delta n_3 = x\delta v_1/v^{u}$ moles of He^3 are transferred from the upper phase to the lower, where x is the concentration of He^3 in the upper phase and v^{u} is the molar volume of the upper phase. We set up the balance of the entire gas and of He^3 alone:

$$\delta v_1/v^{u}+\delta v_2/v^{l}=\delta v_1/v^{l}+\delta\bar{n}_3+\delta\bar{n}_4,$$

$$x\delta v_1/v^{u}+z\delta v_2/v^{l}=y\delta v_1/v^{l}+\delta\bar{n}_3,$$

where y and z are the molar concentrations of He^3 in the lower phase in the solution bath and in the vaporization baths v^{l} is the molar volume of the lower phase in the solution bath and in the vaporization bath. If we multiply the first equation by z and subtract it from the second, we get

$$(x-z)\delta v_1/v^{u}=(y-z)\delta v_1/v^{l}+(1-z)\delta\bar{n}_3-z\delta\bar{n}_4.$$

Taking into account the fact that $\delta v_1/v^{u} = \delta n_3/x$, we have

$$(1-z/x)\delta n_3=(y-z)v^{u}\delta n_3/xv^{н}+(1-z)\delta\bar{n}_3-z\delta\bar{n}_4$$

or

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$$\delta n_3 = \frac{(1-z)\,\delta\bar{n}_3 - z\,\delta\bar{n}_4}{1 - z/x - (y-z)\,v^{\mathrm{u}}/xv^{l}} \tag{1}$$

At temperatures below 0.1°K, we have $\delta n_3 \approx 1.1\ \delta\bar{n}_3$; for 0.5°K, we have $\delta n_3 \approx 1.3\ \delta\bar{n}_3$.

The accuracy of determination of δn_3 amounts to about 1.5% at temperatures below 0.2°K and depends principally on the accuracy of determination of the concentration of the evacuated gas from the mass spectrometer. At higher temperatures, the error amounted to 3–4% because of the uncertainty of the concentration of helium in the vaporization bath, which was determined from the vapor pressure on the basis of[2] and data on thermal osmosis.[3] The value of the molar volume v^{u} in the upper phase was assumed to be the same as in[4,5] at T = 1.2°K; in the lower phase, v^{l} was taken from the data of[3].

The temperature in the solution chamber was measured by means of a bronze resistance thermometer and the magnetic susceptibility of cerium magnesium nitrate. The bronze resistance thermometer was calibrated against the vapor pressure of He^3 (1962 scale) in the range 0.45–1.5°K with an accuracy to within 0.003° and the readings of the cerium magnesium nitrate was tied-in with it. To verify the stability of the thermometer, a sample of pure cadmium in the form of a wire of diameter 60 μ ($R_{300}/R_{4.2}$ = 1100) was placed in the solution chamber. The transition temperature of cadmium, determined by the bronze thermometer, remained constant and equal to 0.506°K (for H = 1.0 Oe). The temperature below 0.45°K was measured by means of a ballistic galvanometer and a sphere compressed from 13 g of fine cerium magnesium nitrate powder, with effective density 1.9 g/cm³. To make the cerium magnesium nitrate scale more precise, demagnetization was carried out from $T = 8 \times 10^{-3}$°K and H = 650 Oe in the almost complete absence of the upper phase in the solution chamber. The magnetic temperature, equal to 2.06×10^{-3}°K, was maintained for more than half an hour. The conversion of the magnetic temperature to the magnetic temperature of a single-crystal sphere, i.e., allowance for the slight ellipticity (axis ratio 1.07) and averaging of the anisotropy in the powder lead to $T = 3.54 \times 10^{-3}$°K, which agrees, within the limits of error, with $T = 3.62 \times 10^{-3}$°K from[9]. It can be assumed that the error in the determination of the temperature amounted to about 1.5%.

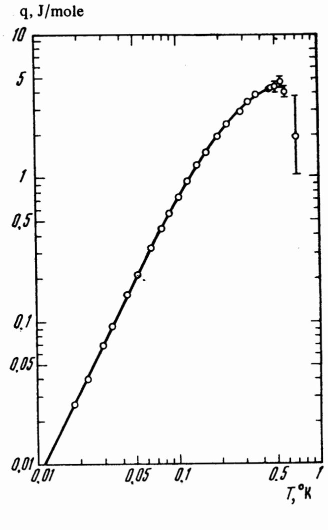
The results of measurement are shown in the table and in the figure. As seen from the drawing, the heat of transition has maximum at T = 0.54°K where it is equal to q = 4.5 J/mole. At temperatures below 0.05°K, the dependence of the heat of transition on the temperature becomes quadratic, in agreement with the theory, and is given by

$$q = (80 \pm 3[\mathrm{J/mole\text{-}°K^2}])\,T^2.$$

Vilches and Wheatley[7] determined the heat of transition for temperatures below 0.05°K and obtained

$$q = (83 \pm 4[\mathrm{J/mole\text{-}°K^2}])\,T^2,$$

but they did not take into account the difference between the quantity δn_3 of He^3 transferred from one phase to another, and $\delta\bar{n}_3$ the evaporating He^3, which amounts to $\delta n_3 = 1.08\ \delta\bar{n}_3$ for these temperatures. Therefore their recalculated data give



Heat of transition of He^3: O—experimental data, continuous curve—calculation from Eq. (2).

T, °K	q, J/mole	T, °K	q, J/mole	T, °K	q, J/mole
0.0180	0.0262	0.0866	0.561	0.323	3.45
0.0224	0.0396	0.101	0.739	0.370	3.95
0.0290	0.0675	0.116	0.952	0.457	4.26
0.0340	0.0915	0.139	1.250	0.470	4.38
0.0437	0.1512	0.161	1.55	0.497	4.42
0.0516	0.211	0.195	1.98	0.543	4.75
0.0643	0.329	0.227	2.42	0.585	4.09
0.0769	0.448	0.283	2.96	0.693	1.96

$$q = (77 \pm 4[\mathrm{J/mole\text{-}°K^2}])\,T^2,$$

which agrees with our results within the limits of error.

The heat of transition q can be determined from the entropies of the two phases:[8]

$$q = T(S^{l} - S^{\mathrm{u}}), \tag{2}$$

where S^{l} and S^{u} are the entropies of the lower and upper phases per mole of He^3. We do not yet have complete data along the stratification curve. But the theories that have been developed allow us to compute the entropy by means of existing data. According to the Landau-Pomeranchuk theory,[9] the entropy of the lower phase is equal to the entropy of an ideal Fermi gas S_F,

$$S^{l} = S_F(T/T_F),$$

with a Fermi temperature $T_F = (3\pi^2 n_3)^{2/3}\hbar^2/2m^*k$, where n_3 is the number of atoms of He^3 per cm³, ħ is Planck's constant, k is Boltzmann's constant, and m* is the effective mass, approximately equal to 2.4 helium atomic masses. The weak interaction between the He^3 quasiparticles leads to a dependence of the effective mass on the concentration.[10] Taking this dependence into account, Radebaugh[3] calculated the entropy in detail as a function of temperature and concentration. In the determination of the entropy along the equilibrium curve, the concentration of He^3 at 0°K was taken to be 6.6% from his tables.[11,12] The properties of the upper phase differ little from the properties of ideal solutions.[13] Neglecting the entropy of pure He^4, which amounts to less than 0.1% of the entropy of He^3, the entropy of the upper phase can be computed from the formula

$$S^{\mathrm{u}} = S_3^{0} - R[x\ln x + (1-x)\ln(1-x)]/x, \tag{3}$$

where S_3^0 is the entropy of pure He^3 and R the gas constant. The entropy of pure He^3 was taken from the work of Radebaugh,[3] who calculated it on the basis of experimental data on the heat capacity of pure He^3[13-16].

The dependence of the heat of transition q on the temperature, determined in this fashion from Eq. (2), is shown in the figure by the continuous curve. As is seen, it agrees completely with the experimental data with accuracy within the experimental error. This shows the excellent agreement beteen our data on the heat capacity, heat of transition of He^3 from one liquid phase to the other, and the shape of the stratification curve.

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214