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[12] The experimental results on self-induced transparency of Ref. 4 show that for SF_6 a 2π pulse at an intensity level of approximately 10 W/cm^2 is about 200 nsec long. For a 2π pulse, $\Omega/2\pi$ times the pulse length is equal to 1; thus, 160 W/cm^2 is required to yield $\Omega/2\pi = 20$ MHz.

ISOTOPIC PHASE SEPARATION IN SOLID He^3-He^4 MIXTURES*

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The excess pressure due to phase separation of solid He^3-He^4 mixtures held at constant volume has been measured. From this we obtain the phase-separation temperature as a function of the mole fraction, the excess volume, and the pressure dependence of the critical temperature.

By observing the specific heat of a mixture of solid He^3 and He^4, Edwards, McWilliams, and Daunt[1] (EMD) discovered, in 1962, that these mixtures undergo phase separation below a certain temperature. Recently a microscopic theory of the phase separation has been presented by Mullin.[2] In this paper we report measurements of the excess pressure P^E due to phase separation of He^3-He^4 mixtures held at constant volume. In addition to the phase-separation temperature as a function of the mole fraction of He^3, x, the measurements give the excess volume v^E and the pressure dependence of the critical temperature, which are quantities predicted by the Mullin theory.

EMD found that the specific heat due to mixing could be represented quite well by the regular solution model. In this model[3] one employs various excess quantities which are generally defined as the increase in that quantity upon mixing. The excess Gibbs function is given by

$$g^E = x(1-x)\Delta E(P), \tag{1}$$

where ΔE is the energy of mixing. This leads to a critical temperature for phase separation of

$$T_c = \Delta E/2R. \tag{2}$$

The specific heat in the phase-separation region is expressed as

$$C = 2RT_c(1-2x_{ps})(dx/dT)_{ps}, \tag{3}$$

where the subscript "ps" means along the phase-separation curve. For the excess volume we have

$$v^E = (\partial g^E/\partial P)_T = x(1-x)2R(\partial T_c/\partial P). \tag{4}$$

Mullin finds that the solution is almost regular, but the essential feature is the difference in molar volumes of the pure He^3 and He^4, $v_3 - v_4$, which would be zero in the classical theory. He obtains for the excess Gibbs function

$$g^E = x(1-x)[a(P)-b(P)x], \tag{5}$$

where the small term in x in the square bracket causes a slight departure from the regular solution. The main effect of this extra term is to cause asymmetry in the phase-separation curve. For the excess volume Mullin obtains

$$v^E \simeq -\sigma x(1-x)/P, \tag{6}$$

with $\sigma/P \simeq 0.4$ cm^3/mole. He computes the T-x

curve for $P=35.8$ and for $P=55$ atm, finding a decrease in T_C of 0.02°K.

A decrease in the phase-separation temperature with increasing pressure has been reported by Zimmerman.[4] However, because of the sharp increase in the time constant for the phase separation, it is difficult to determine accurately the lowering of T_C by making measurements at higher pressures. As we show below, by combining excess-pressure measurements with specific heats, $\partial T_C/\partial v$ or $\partial T_C/\partial P$ can be determined from data at a single pressure.

To relate $\partial T_C/\partial v$ to the excess pressure P^E we require only the Maxwell relation $(\partial P/\partial T)_v = (\partial S/\partial v)_T$ and the fact that the experimental specific heat is represented quite well by Eq. (3) where $C=C(T/T_C)$. From this we have

$$(\partial P/\partial T)_v = -(C/T_c)(\partial T_c/\partial v). \tag{7}$$

Substituting for C from Eq. (3) and integrating from 0 to T gives for the excess pressure

$$P^E = -2Rx(1-x)\partial T_c/\partial v. \tag{8}$$

Thus, measurements of the excess pressure provide $\partial T_C/\partial v$ without the need for performing the experiment at different pressures. While Eq. (8) can also be obtained directly from the excess Helmholtz function a^E, using $P^E = -(\partial a^E/\partial v)_T$, the above approach does not rely as heavily on the regular solution theory. The excess volume and excess pressure are related by

$$v^E = -P^E(\partial v/\partial P)_T = vk_T P^E, \tag{9}$$

where k_T is the isothermal compressibility.

Measurements of pressure versus temperature at constant volume have been made for samples with $x=0.03$, 0.084, 0.166, 0.485, 0.928, and 0.9961. The samples were prepared in known proportions and the concentrations were also checked using a Varian Model 974-0036 partial pressure gauge. Portions of the apparatus have been described briefly elsewhere,[5] with the only change being the use of a Be-Cu capacitive strain gauge with a pressure sensitivity of 3×10^{-6} atm.

For most concentrations, measurements have been made at several different molar volumes. These were usually taken in the warming direction, although some data were taken while cooling. Figure 1 shows a typical set of data where ΔP vs T is given for $x=0.485$. Each curve is labeled by the minimum pressure on the sample in the region above the phase-separation temperature, with the pressure changes relative to this value. The phase-separation temperature is taken to be at the inflection point of each curve. In the region of temperature covered here the lattice contribution to ΔP is negligible,[6] and the exchange contribution[5] has been subtracted for the sample with $x=0.9961$. The excess pressure is the negative of the total pressure increase at the lowest temperature, where the sample is essentially completely separated. The section of the curve labeled bcc-hcp will be discussed later.

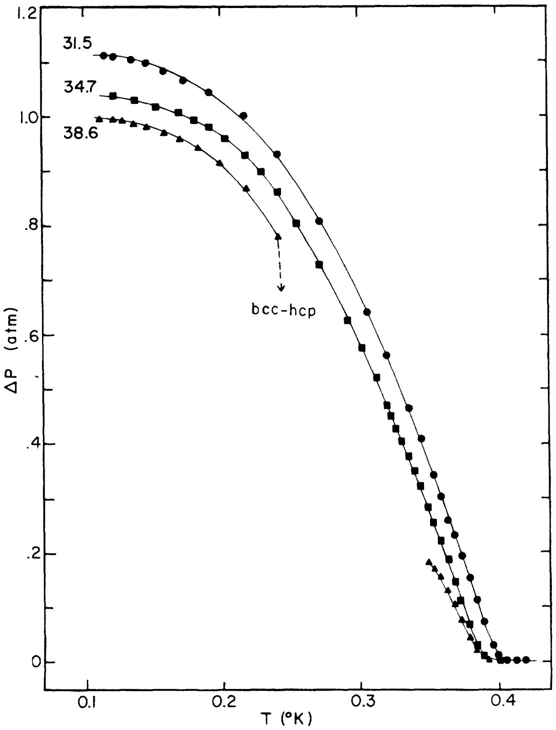

FIG. 1. The pressure change due to phase separation for $x=0.485$ at the indicated sample pressures. The line labeled bcc-hcp is explained in the text.

A summary of the results is given in Table I. We see that for $x=0.9961$, 0.928, and 0.485 our phase-separation temperatures are in close agreement with those of EMD. However, for $x=0.03$, 0.084, and 0.166 we find the temperature to be somewhat higher than the symmetrical curve of the regular solution theory. Thus we would conclude that the separation curve, as shown in Fig. 2, is slightly asymmetrical in the direction indicated by Mullin. This is not inconsistent with the findings of EMD since they studied only samples with $x>0.2$ where there is little departure from the symmetric curve. The relatively large temperature error bars for our data

Table I. Various phase-separation properties of solid He^3-He^4 mixtures.

x	T_{ps} (°K)	T_{ps}(REG)[a]	P (atm)	$-P^E$ (atm)	$-V^E$ (cm^3/mole)	$-V^E(M)$[b] (cm^3/mole)	$-\frac{dT_c}{dP}$ (m°K/atm)
0.03	0.270	0.202	28.6[c]	0.175	0.0138	0.0116	2.80
0.03	0.249		33.9	0.099	0.0066		1.40
0.084	0.310	0.267	33.5	0.436	0.0288	0.0307	2.28
0.166	0.345	0.312	35.4	0.662	0.046	0.0555	2.02
0.485	0.380	0.377	31.5[c]	1.11	0.109	0.010	3.04
0.485	0.374		34.7	1.05	0.0927		2.58
0.485	0.369		38.5	0.998	0.0758		1.70
0.928	0.249	0.252	32.7	0.301	0.0345	0.0267	2.75
0.928	0.255		33.9	0.288	0.0314		2.50
0.9961	0.138	0.138	33.8	0.0139	0.00168	0.00155	2.58
0.9961	0.138		37.0	0.0104	0.00111		1.70

[a] Phase-separation temperatures at 35.8 atm given by the regular solution model; see Ref. 1.

[b] Excess volumes at 35.8 atm predicted by Mullin, Ref. 2.

[c] To prevent the He^3-enriched phase from melting, these samples were not cooled below 0.21 and 0.10°K, respectively.

in Fig. 2 result from the difficulty in locating precisely the inflection point on the curves of P vs T.

As listed in Table I, the magnitudes of the excess pressures range from about 1 atm[7] for $x = 0.485$ to 10^{-2} atm for $x = 0.9961$. These generally show a decrease as the pressure increases. The excess volumes were obtained from P^E using Eq. (9), with the compressibility taken as the appropriate average of that of the pure constituents.[6,8] The excess volumes are generally in remarkably good agreement with those calculated by Mullin, Eq. (6), for $P = 35.8$ atm.

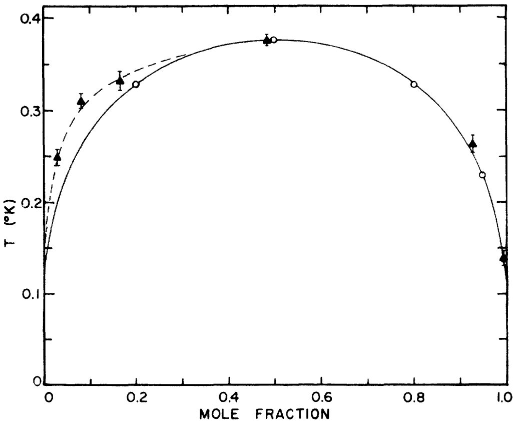

FIG. 2. Phase-separation temperature versus mole fraction of He^3. Triangles, this work; circles, Edwards, McWilliams, and Daunt (Ref. 1); solid line, regular solution theory.

The last column of Table I gives $\partial T_c/\partial P$, which is obtained from the excess volumes using Eq. (4). We find $\partial T_c/\partial P \simeq -2$ mdeg K/atm, with a smaller absolute value as the pressure is increased. This is in qualitative agreement with the average value of -1 mdeg K/atm between 35.8 and 55 atm predicted by Mullin.

All of the measurements reported here have been complicated by crystallographic changes as indicated by the broken portion of one of the curves in Fig. 1. At the pressures and temperatures involved here the crystal structure of pure He^3 is bcc while that of pure He^4 is hcp. Hence, we would expect to see a crystallographic change in one of the two almost pure phases below the phase-separation temperature. These changes were always observed. If the structure of the mixture were bcc, on increasing the temperature a given amount we first saw the decrease in P due to the partial mixing, followed by a continual decrease in P due to the bcc-hcp transition in the He^4-rich phase. When the mixture structure was hcp, on warming there was, after the decrease in P due to partial mixing, an increase in P due to the hcp-bcc transition in the He^3-rich phase. It was found that the crystallographic transition occurs at a much slower rate than the phase separation, hence, it was possible to distinguish between the two processes. The measurements were taken at a rate such that the changes in P due to the crystallographic changes were small compared with those due to the phase separation.

Also several instances of crystallographic changes in the mixed or partially mixed samples have been observed. No attempt to locate these phase boundaries has been made since attaining equilibrium conditions would require waiting for very long times. For some concentrations, the phase separation has been observed at one pressure for which the mixed structure was bcc and at a higher pressure for which the mixed structure was hcp. This difference in crystal structure appears to have very little if any effect on the phase-separation temperature.

For a given concentration, the time constant for the phase separation always increased with increasing pressure. For example, in the $x = 0.485$ sample, τ increased from ~30 sec at 30.5 atm to ~500 sec at 38.5 atm. Although we do not have conclusive quantitative results concerning the dependence of τ on the temperature and concentration, some qualitative conclusions appear valid. For a fixed concentration the time constant is longer in the vicinity of the phase-separation temperature than at lower temperatures, and is longer on cooling than on warming. Also τ appears to be longer for the smaller He^4 or He^3 concentrations, particularly the latter. EMD find $\tau \sim 15$ sec at 35.8 atm, independent of x and T. Apparently the time constant is affected by the dimensions of the sample since that of EMD was contained in pores of 10-μ diam while the smallest dimension of our sample was 0.8 mm.

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MAGNETIC THERMOMETRY WITH CERIUM MAGNESIUM NITRATE AND ITS RELATION TO THE PROPERTIES OF He^3†

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Direct comparison of the magnetic temperature of one cerium-magnesium-nitrate powder thermometer with that of a single-crystal sphere shows that the former is less than the latter by $\Delta = +(0.46 \pm 0.05)$ mdeg K. Comparison of other powdered thermometers suggests that Δ may vary from thermometer to thermometer by 0.1 to 0.2 mdeg K. These values of Δ do not lead to serious difficulties when comparing experimental properties of He^3 with theories of the Fermi liquid.

We have made a direct comparison of the magnetic temperatures indicated by a powder (size less than 420 μ) of cerium magnesium nitrate (CMN) in the form of a right circular cylinder with diameter equal to height and by a sphere of CMN. The results, obtained over the range 10-38 mdeg K, are described by

$$T_S^* = T^* + \Delta, \tag{1}$$