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## Precipitous Onset for Superfluidity in $^3\text{He}$ - $^4\text{He}$ Mixtures

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The precipitous onset step of Kosterlitz, Thouless, and Nelson has been investigated in  $^3\text{He}$ - $^4\text{He}$  mixture films over the temperature range 1.3 K to  $T_\lambda$ . The constant relating the minimum value of superfluid density to the absolute temperature is found experimentally to be universal up to 30%  $^3\text{He}$ . Comparison is made with recent mixture theories. Measurements were made at 24 MHz with a quartz microbalance adsorptometer.

Recent theoretical work has culminated in certain remarkable predictions about two-dimensional superfluids.<sup>1-5</sup> In particular the two-dimensional superfluid density  $\sigma_s$  is predicted to have a certain minimum value,  $\sigma_s^0$ , which depends only on the mass,  $m$ , of the  $^4\text{He}$  atom and the absolute temperature  $T$ :

$$\sigma_s^0 = \frac{8\pi m^2 K}{h^2} T. \quad (1)$$

Here  $K$  and  $h$  are the universal constants of Boltzmann and Planck.

Thin adsorbed films of helium are the closest physical analog to a two-dimensional superfluid available. The superfluid density in such films does indeed drop precipitously to zero as the transition is crossed from the superfluid to the normal phase.<sup>6-8</sup> And the magnitude of the step found experimentally is that given by Eq. (1).

Recently the theory leading to Eq. (1) has been extended beyond simple thermodynamic equilibrium to include finite-frequency effects.<sup>9</sup> The resulting dynamic theory predicts an excess damping near the transition, with the result that the step assumes a width at finite frequencies. Bishop and Reppy<sup>6</sup> have performed an Androni-

kashvili-type experiment on  $^4\text{He}$  films at 2.5 kHz. A four-parameter fit to their data is in good agreement with the dynamic theory, and constitutes an important confirmation of its essential correctness. Not yet having available a finite-frequency theory for mixture films we offer here no analysis of the width of the onset jump we observe. A cursory estimate does indicate that our widths are comparable to those predicted for pure  $^4\text{He}$ .

In the Kosterlitz-Thouless-Nelson (KTN) theory<sup>1,2</sup> [Eq. (1)], the mass which enters is the  $^4\text{He}$  atomic mass. It is not some renormalized "effective mass."<sup>2</sup> Near the onset temperature  $T_o$ , the healing length is always comparable to the film thickness. Thus any such film can be viewed as two dimensional.<sup>10</sup> So Eq. (1) actually contains two predictions. Not only is  $\sigma_s^0/T_o$  predicted to be temperature independent, but the additional statement is made that it is universal. It contains no hidden dependence on onset thickness  $d_o$ , healing length, or bulk superfluid density  $\rho_s$ .

There is yet a further test of Eq. (1) that bears exploration. Is the constant also independent of  $^3\text{He}$  concentration? Solvable microscopic theories of  $^3\text{He}$ - $^4\text{He}$  mixture films are difficult to con-

struct, but a simple two-dimensional lattice-gas model has just been analyzed by Berker and Nelson.<sup>3</sup> For reasonable values of the parameters in their Hamiltonian, they predict that the magnitude of the superfluid onset jump will be the universal one, but only for small concentrations of <sup>3</sup>He. Their model predicts that a two-dimensional phase separation must occur before the concentration reaches 12%, reducing the magnitude of the onset jump. Other researchers<sup>4,5</sup> have reached similar conclusions.

We have performed a series of experiments on <sup>3</sup>He-<sup>4</sup>He mixtures using an rf microbalance adsorptometer. The experimental procedure closely followed that reported earlier in connection with measurements on pure <sup>4</sup>He.<sup>8,11,12</sup> The major difference in apparatus was the addition of a gas-handling system to prepare, admit, and recover the <sup>3</sup>He-<sup>4</sup>He mixtures. A comprehensive description of the experiment, covering all of its results, will be published elsewhere shortly.

We offer here our experimental results on the onset step of superfluidity in <sup>3</sup>He-<sup>4</sup>He mixtures. The measurements span temperatures from 1.3 K to  $T_\lambda$ . In this temperature range we explored all of the concentration, exhibiting superfluidity. We represent by  $c$  the molar concentration of <sup>3</sup>He in the mixture of gases which condenses to form the film. The molar concentration of <sup>3</sup>He just inside the surface of this film is represented by  $\beta$ . The liquid concentration  $\beta$  is determined by the gas concentration  $c$  and the temperature  $T$ . In equilibrium the liquid's vaporization (boiling) pressure must equal the liquification (dew) pressure of the gas.<sup>13</sup> That is, given  $T$  and  $c$ , the concentration  $\beta$  is determined by

$$P_b(\beta, T) = P_d(c, T). \quad (2)$$

The subscripts  $d$  and  $b$  indicate "dew" and "boiling," respectively. From this notion, utilizing our own measurements as well as values from the literature,<sup>14</sup> we deduce that we are exploring films characterized by surface concentrations  $\beta$  ranging from 0 to 0.3 moles of <sup>3</sup>He per mole of mixture.

To measure the total adsorbed mass per unit area,  $\sigma$ , and the adsorbed mass of superfluid per unit area,  $\sigma_s$ , we employed a quartz-crystal microbalance adsorptometer. This consists of a flat-wafer  $Y$ -cut quartz crystal oscillating in its fundamental thickness-shear mode at about 24 MHz. A loading adsorbed mass per unit area,  $\sigma_m$ , on both faces of this crystal reduces its resonance frequency by an amount  $\Delta f$  which directly

measures  $\sigma_m$  by virtue of the relation

$$\sigma_m = -S\Delta f, \quad (3)$$

where the constant  $S$  depends only upon the properties of the quartz crystal.<sup>11</sup> In our case  $S$  is known to be 0.385 g/cm<sup>2</sup>GHz. Since any superfluid in the film contributes nothing to the loading we thus obtain a direct measure of  $\sigma - \sigma_s = \sigma_m$ , the difference between the total mass per unit area adsorbed and the areal mass of superfluid. The quantity  $\sigma_m$  is measured experimentally at many pressures  $P$  of the gas mixture producing the adsorbed film for each temperature  $T$  and concentration  $c$  of the gas. That is, we experimentally determine  $\sigma_m = \sigma_m(P, c, T)$ .

It is possible to deduce the total film mass  $\sigma$  even in regions where a portion of that mass, the superfluid, does not load the adsorptometer. This is illustrated in Fig. 1. When data points from many different temperatures, but approximately the same  $\beta$ , are plotted in the reduced fashion shown, they coalesce to form a single, "universal" curve throughout the nonsuperfluid regions. The dark circles are such data points. Under the assumption that the presence of super-

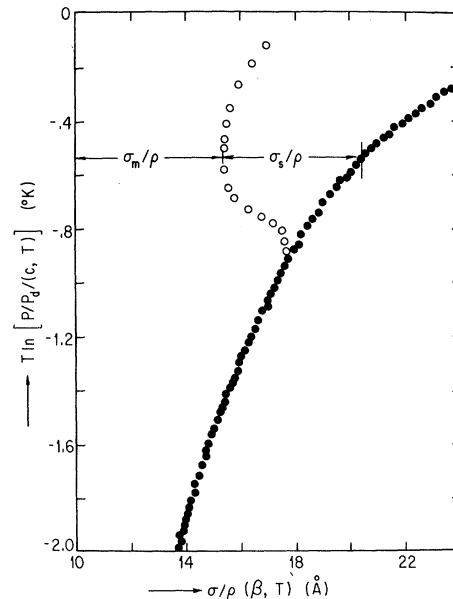


FIG. 1. Data from a variety of concentrations ( $0.0 < \beta < 0.1$ ) and temperatures ( $1.5 < T < 2.1$ ) coalesce to form a "universal" curve (dark circles). One run (open circles) exhibiting superfluidity ( $\beta = 0.05$ ,  $T = 1.3$  K) illustrates how  $\sigma_s$ , the areal superfluid density, is determined from the measured  $\sigma_m$ . The abscissa  $\sigma_s/\rho$ , approximates the film thickness. The ordinate measures the van der Waals potential at that thickness.

fluid does not significantly alter it, the total film mass is given by this curve even when  $\sigma_s$  is non-zero. Data from a single run exhibiting superfluidity are also shown (open circles). The basis of the "universality" exhibited in this plot is that the horizontal axis approximates the film thickness, while the vertical (logarithmic) axis is a measure of the van der Waals potential at the film's surface.<sup>13</sup>

The result of performing the operations described above is illustrated in Fig. 2. Data from two different experimental trials is shown to illustrate the scatter. After an initial precipitous step,  $\sigma_s$  rises linearly with thickness. The slope with which it rises is significant. We always find it to be  $\rho_s = \rho_s(\beta, T)$ , the three-dimensional superfluid density of the bulk mixture. Figure 2 also illustrates how  $d_o$ ,  $d_I$ , and  $\sigma_s^0$  were determined. The onset thickness  $d_o$  is taken as the point of inflection of the steplike portion of the curve. The intercept thickness  $d_I$  is found by extrapolating the straight-line (slope  $\rho_s$ ) portion of the curve to  $\sigma_s = 0$ . This is a measure of the excess normal-fluid density associated with the surface. The value of the jump,  $\sigma_s^0$ , is clearly geometrically related to the other parameters by  $\sigma_s^0 = (d_o - d_I) \times \rho_s(\beta, T)$ .

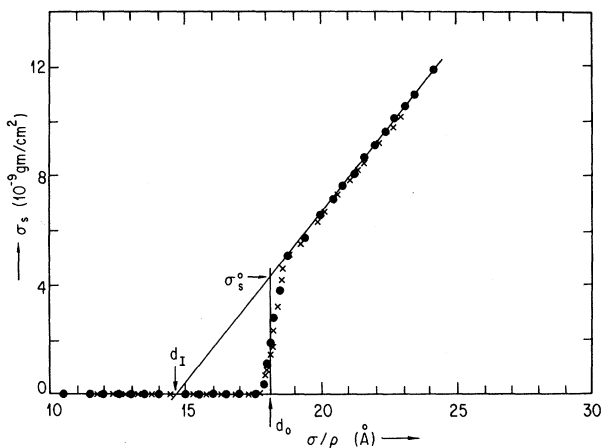


FIG. 2. The superfluid areal density  $\sigma_s$  is plotted as a function of  $\sigma/\rho$ , the approximate film thickness, for two different experimental trials (circles, crosses) at  $T = 1.3$  K and  $\beta = 0.05$  (5%  $^3\text{He}$ ). The data exhibit a precipitous onset, followed closely by a linear region with slope  $0.124$  g/cm<sup>3</sup>, which is the bulk superfluid density for that concentration and temperature. The onset thickness  $d_o$  is taken at the inflection point of the onset "step." The thickness of film which never displays superfluidity  $d_I$  is found by extrapolating the linear region to  $\sigma_s = 0$ . The magnitude of the jump,  $\sigma_s^0$ , is the value of this extrapolated line at  $d_o$ .

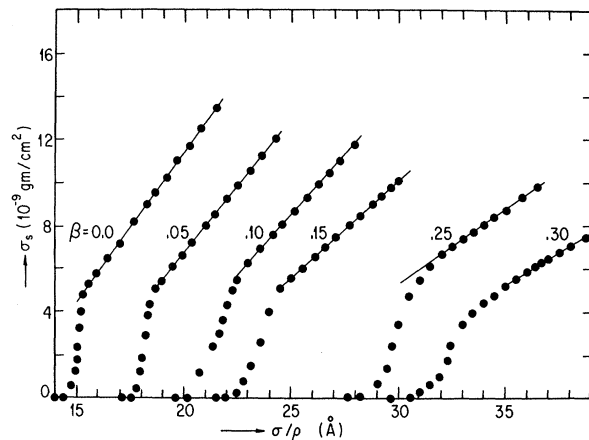


FIG. 3. The areal superfluid density  $\sigma_s$  as determined by subtraction (see Fig. 1) is shown as a function of  $\sigma/\rho$ , the approximate film thickness, for a variety of concentrations,  $\beta$ , at 1.3 K. Each curve exhibits a relatively precipitous rise, followed by a linear region with slope  $\rho_s(\beta, T)$ . As summarized quantitatively in Table I and in Fig. 4 the heights of the steps appear to be roughly equal indicating the universality of the KTN theory.

Figure 3 shows our results for a number of different concentrations,  $\beta$ , measured at the same temperature  $T = 1.3$  K. Higher values of  $\beta$  do not exhibit superfluidity at this temperature. Our results for a range of temperatures and concentrations are compiled in Table I.

Figure 4 shows some of our values for the magnitude of the onset jump. On the left side is re-plotted previously published data at zero concentration of  $^3\text{He}$  ( $\beta = 0$ ). Our data at the temperature  $T = 1.3$  K for  $^3\text{He}$  concentrations ranging out

TABLE I. Summary of our experimental results. The average of all of our values for the quantity  $\sigma_s^0/T_o$  is  $3.33$  g/cm<sup>2</sup> K. The KTN theoretical value is  $3.49$  g/cm<sup>2</sup> K.

$\beta$	$T$ (K)	$\rho_s$ (g/cm <sup>3</sup> )	$d_I$ (Å)	$d_o$ (Å)	$\sigma_s^0/T_o$ (g/cm <sup>2</sup> K)
0.05	1.8	0.084	24.1	30.9	3.17
0.05	1.7	0.096	21.5	27.1	3.18
0.05	1.6	0.105	19.0	24.1	3.25
0.05	1.5	0.113	17.6	21.9	3.17
0.05	1.4	0.118	16.0	19.4	3.07
0.05	1.3	0.123	14.6	18.8	3.23
0.10	1.3	0.106	17.0	21.3	3.50
0.15	1.3	0.089	18.8	23.6	3.46
0.25	1.3	0.064	21.5	30.0	4.00
0.30	1.3	0.053	24.3	32.8	3.30

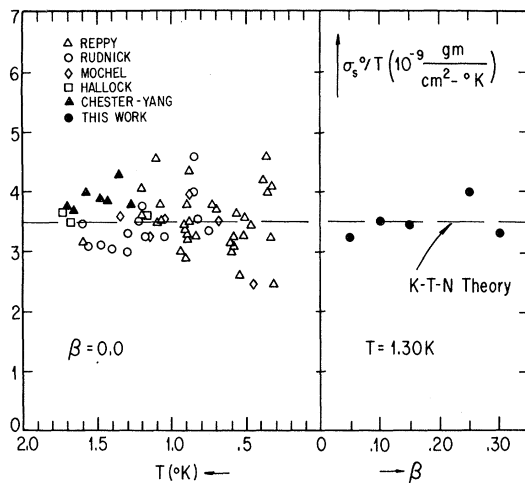


FIG. 4. Measured values of the ratio  $\sigma_s^0/T$ , characterizing the superfluid areal density at onset are exhibited for comparison with the KTN theoretical value (dashed line). Points on the left are replotted from previously published pure  ${}^4\text{He}$  experiments showing the temperature explored. Our values of the step height at 1.3 K are displayed in the graph to the right, showing the range of concentrations  $\beta$  over which we observed superfluidity. At higher temperatures no superfluidity is observed for  $\beta = 0.3$ .

to 30% are plotted on the right. It is clear that, to within the accuracy of former experiments, the universality of the onset step is preserved.

Experimental errors were mainly of the systematic variety. In an individual run measurements of  $\sigma_m$  were consistent within  $1 \times 10^{-10}$  g/cm<sup>2</sup>. Errors due to oscillator frequency drift, nonlinear pressure measurement, and imprecise determination of the dew point contributed an error, varying from run to run, of about  $3 \times 10^{-10}$  g/cm<sup>2</sup>. Influencing all measurements equally were global systematic errors related to crystal roughness and other adsorptometer properties, estimated as  $\pm 10\%$  in measured densities.

We conclude that the jump in superfluidity at the "precipitous onset" in  ${}^3\text{He}$ - ${}^4\text{He}$  mixtures is characterized by a universal constant in the manner given by Eq. (1). The  ${}^3\text{He}$  concentration is a neutral variable for the superfluid transition in the quasi-two-dimensional region, as predicted.<sup>3,4</sup> However, we have seen no evidence for two-dimensional phase separation and no diminution of

the onset jump for concentrations beyond the 12% limit mentioned by Berker and Nelson. Possibly surface inhomogeneities inhibit the phase separation. Another extreme is also possible. Vertical (perpendicular to surface) phase separation may occur. This latter possibility seems unlikely in view of the following: Although the universality of the onset jump is preserved even with substantial  ${}^3\text{He}$  concentrations, the other characteristics of the system are severely modified by the addition of this impurity. The onset thickness varies markedly with  $\beta$ , as does the onset temperature. Additionally, the linear regions of Fig. 3 all exhibit slopes  $\rho_s(\beta, T)$  characteristic of homogeneous bulk mixtures of concentrations  $\beta$ . This verifies that the films do have the compositions we deduce from Eq. (2). Regardless of the explanation proposed, the range of universality that we observe far exceeds that predicted.

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