# Coherence-length effects on the nucleation of superfluid transitions in liquid-helium mixtures

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A theory of the nucleation of superfluid transitions near container walls of 'He-'He mixtures is developed. The theory uses a local-continuum model, which accounts for wall-potential-induced concentration and pressure gradients, supplemented by an important coherence-length correction to account for the finite thickness of the nucleation films. An excellent one-parameter fit to the available experimental data is obtained. The theory provides support for the concept of universality in phase-transition phenomena.

## I. INTRODUCTION

In several recent papers' superfluid transitions in liquid-helium mixtures were studied within a continuum model which included concentration and pressure gradients induced in the systems by the Van der Waals forces acting between the helium and its container walls. The continuum model was a purely local one in which local equations of state derived from bulk properties of spatially uniform mixtures were used. Such a model is, of course, inexact in that it cannot rigorously describe systems in which properties vary significantly over distances of a few interatomic spacings.<sup>2</sup> From our point of view, however, this is not an extremely serious defect; local models have been widely used for many years (at least since the introduction of the wellknown Thomas-Fermi model for atoms) and are known to describe average properties of systems rather well. More serious here is the fact that coherence-length effects associated with continuous (i.e., second-order) phase transitions are improperly accounted for unless all lengths characterizing important variations in system properties are large compared to the coherence length in question. In particular, the local model for  $3$ He- $4$ He mixtures predicts that even for a mixture nominally quite rich in  ${}^{3}$ He, the local  ${}^{3}$ He concentration near a container wall will be rather small  $(88\%)$ . This leads to the conclusion that, for a certain range of temperatures  $T$  and  ${}^{3}$ He number concentrations  $X_0$ , a thin superfluid phase can exist near the walls in a system where the bulk remains normal. As  $T$  is lowered from a relatively large value,  $X_0$  being fixed, a point will be reached where the superfluid film will begin to appear. With continued lowering of  $T$ 

the film thickness will grow from zero to larger and larger values, bulk superfluidity finally being obtained. While superfluid film formation actually occurs, ' this description cannot be correct. Bulk properties of superfluid 'He, for example, certainly do not suffice to predict  $\lambda$  temperatures for thin <sup>4</sup>He films. The  $\lambda$  transition for bulk <sup>4</sup>He is 2.17 K, whereas transitions in very thin (thickness approximately equal to a few atomic layers<br>films occur at temperatures of the order of 0.1–0<br>K<sup>4,5</sup> The extent of the failure of the local confilms occur at temperatures of the order of  $0.1-0.2$ K.<sup>4,5</sup> The extent of the failure of the local continuum model is shown in detail in Sec. III.

One very fruitful way in which to look at the behavior of thin 4He films (or, more generally, 'He under conditions of restricted geometry) is to fix the temperature  $T$  and vary the thickness d from very small to larger and larger values. Given that  $T$  is not too small but nevertheless is less than 2.17 K, a thickness  $d_s(T)$  will be reached at which superflow in the film can first be observed. There is considerable evidence (discussed in Secs. III and IV) that this thickness is at least in essence proportional to the coherence length  $\xi(T)$  in bulk superfluid <sup>4</sup>He.

In the present work we use the idea that a film, predicted to be superfluid on the basis of local bulk properties alone, will not in fact become superfluid until it reaches a certain minimum thickness related to a coherence length. The precise relationship, which invokes careful spatial averages of the superfluid density  $\rho_s$  since system properties vary with position for our system, is given in Sec. II, where the local-continuum model of Ref. 1 is also reviewed.

Our results are described in detail in Sec. III. With a single fitting parameter we find excellent agreement with experiment for all temperatures greater than 0.87 K, the tricritical temperature

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for bulk mixtures under vapor pressure. This is consistent with, and provides evidence in favor of, the notion of universality<sup>6</sup> in the theory of critical phenomena. For  $T < 0.87$  K tricritical phenomena in the films near the walls begin to play an important role, the full nature of which will be explored in a future article. In implementing the coherence-length idea a new and widely applicable semiphenomenological expression for  $\rho_s$  is developed. Section IV summarizes our work and contains additional discussion.

## II. BASIC THEORY

The local-continuum theory<sup>1</sup> begins with the three exact equilibrium conditions for  ${}^{3}$ He $-{}^{4}$ He mixtures. These conditions require that the <sup>3</sup>He chemical potential  $\mu$ <sub>3</sub>, the <sup>4</sup>He chemical potential  $\mu$ <sub>4</sub>, and the temperature be constant throughout the system. Let  $U(\vec{r})$  be an external potential, the same for both helium isotopes. The theory approximates  $\mu$ <sub>3</sub> and  $\mu$ <sub>4</sub> by their local values plus contributions from  $U(\tilde{r})$ , i.e.,

$$
\mu_3 = \mu_{30}(P, X, T) + U(\overline{r}) = \text{const},\tag{1a}
$$

$$
\mu_4 = \mu_{40}(P, X, T) + U(\vec{r}) = \text{const.}
$$
 (1b)

Here  $T$  is everywhere constant, the pressure  $P$ and <sup>3</sup>He number concentration X vary with  $\bar{r}$ , and  $\mu_{i0}(P,X,T)$ ,  $i=3, 4$  is calculated for a uniform system at the given values of  $P$ ,  $X$ , and  $T$ .

Use of (1) and some straightforward thermodynamic calculations results in the two very practical equations

$$
vdP = -dU(\vec{r}),\tag{2}
$$

$$
d\ln\left(\frac{X}{1-X}\right) = -\frac{\Delta v}{\gamma k_B T} \, dP\,,\tag{3}
$$

in which

$$
v = Xv_3 + (1 - X)v_4, \tag{4}
$$

$$
\Delta v = v_3 - v_4,\tag{5}
$$

$$
\gamma(X,\,T,\,P)=\frac{X}{k_{B}T}\left(\frac{\partial\,\mu_{3}}{\partial X}\right)_{T,\,P}.\tag{6}
$$

We denote by  $v_3$  and  $v_4$  the respective specific volumes of <sup>3</sup>He and <sup>4</sup>He. The position dependence of all quantities is induced by the position dependence of  $U(\vec{r})$  in (2).

We are specifically interested in the case where  $U(\vec{r})$  is the potential felt by the system in the presence of a planar container wall. The wall is located at  $z = 0$ , and the system occupies the halfspace  $z > 0$ . Approximating the container by a continuous system whose atoms interact with the helium atoms via a Van der Waals potential yields

$$
U(\vec{\mathbf{r}}) = -\alpha/z^3. \tag{7}
$$

The value  $\alpha$  = 27 K/(layer)<sup>3</sup> for the Van der Waals constant' has been used in our computations.

Data from measurements on bulk systems give v,  $\Delta v$ , and  $\gamma$  as functions of T, X, and P. Putting  $\Delta v$  and  $\gamma$  in (3) and integrating allows one to express X in terms of P, T, and the pressure  $P_0$ , and concentration  $X_0$  of a reference state, chosen to be that of the bulk system far from the wall, where all  $z$  dependence is negligible. We calculate only for the case where  $P_0 \approx 0$  is the vapor pressure for nominal bulk concentration  $X_0$  and temperature  $T$ . The result for  $X$  is combined with the data for  $v(X, T, P)$  and placed in (2), which, in turn, is integrated from the state at  $z = \infty$  to a given point z. This gives  $P = P(z, X_0, P_0, T)$ , from which all other local thermodynamic quantities such as  $X(z, X_0, P_0, T)$  may be found.

Various phase boundaries, as defined by bulk measurements may be crossed in the course of the calculation just described. One boundary always reached, as a consequence of the very large attractive potential near the container wall is the solid-liquid phase boundary for the local values of  $P$ ,  $X$ , and  $T$ . When this happens we simply cut off the integration and take the system from  $z = 0$  to the cutoff value  $z = z$ , to be solid. We have no direct interest in any local properties of the always-present solid film. In terms of atomic layers (1 layer = 3.6 Å) z, varies little from a value of 1.5.

Given the local values of  $P$  and  $X$  one can calculate (if it exists) the local value  $T_{\lambda}(P,X)$  of the  $\lambda$  temperature for the system.  $[T_{\lambda}(P,X)]$ =  $T_{\lambda}(z,X_0,P_0,T)$  varies with position because P and  $X$  vary]. According to the local continuum theory the system will be superfluid wherever  $T < T_{\lambda}(P,X)$ . As mentioned in the introduction, general features of our results are that  $X$  decreases with decreasing  $z$  (see Figs. 1 and 2). In the temperature range  $0.9 < T < 1.2$  K, of interest here, this has the consequence that  $T_{\lambda}(P, X)$  (see Figs. 3 and 4) is a monotonically increasing function of z until  $z = z_s$ , at which point  $T_{\lambda}$  drops abruptly to zero.<sup>8</sup> Therefore, a superfluid phase, if it is present, will be localized near the wall, its boundaries being determined by  $z = z_s$  and the point at which

$$
T = T_{\lambda}(P, X) = T_{\lambda}(z, X_0, P_0, T). \tag{8}
$$

Given that all  $T_{\lambda}$  curves of importance here possess the monotonicity property just mentioned,<sup>8</sup> it becomes clear that for fixed  $X_0$  and  $P_0$  the superfluid phase will first appear, as  $T$  is lowered from some relatively large value [say, a value larger than  $T_{\lambda}^{\max}=2.17$  (K), where (8) evidently has no solution], at  $z = z_s$  with zero thickness. The thickness  $e_s(T, X_0, P_0)$  will grow as T is fur-

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FIG. 1. Concentration X of <sup>3</sup>He and pressure P versus the distances from the walls calculated in the continuum model for a mixture of bulk concentration  $X_0$ =0.7 at a temperature  $T=0.965$  K. For distance layers the mixture is solid, the local pressure exceeding the local melting pressure.



FIG. 2. Same as Fig. 1 for a bulk concentration  $X_0$ = 0.6 and at a temperature  $T=1.07$  K.



FIG. 3. Local values of the superf cal values of the superfluid transition ten<br>  $\lambda(z) = T_{\lambda}(Xz)$ ,  $P(z)$ , T) and superfluid density  $\rho_s$  vs z, the distance from the walls, for  $X_0=0.7$ and  $T = 0.965$ . The temperature at which are done is represented as a dotted line.  $\rho_s$  is different from zero at distances such that  $T \le T_\lambda(z)$ .



IG. 4. Same as Fig. 3 for  $X_0 = 0.6$  and  $T = 1.07$ . As the case presented in Fig. 3 the evaluation of the integral of  $\rho_s$  gives  $\sigma_s/T = 0.27 \times 10^{-8}$ g/cm<sup>2</sup> K.

ther lowered, reaching infinity when  $T = T_{\lambda}(P_0, X_0)$ .

The problem with the local-continuum model is that it invokes the idea that the  $\lambda$  temperature of a system is independent of its thickness. This, as discussed in Sec. I, is in complete contradiction with the data for  $\frac{4}{1}$ He films<sup>4,5</sup> as well as with modern theory.<sup>9,10</sup> What we shall do here is to modern theory. What we shall do here is to construct a modified continuum model. The numerical results of the continuum model are retained with the addition of the idea that superfluidity does not appear near the walls until  $e_s(T, X_o, P_o)$  attains a value proportional to a superfluid coherence length, at which point the entire film in the range  $z_s \leq z \leq e_s$  becomes superfluid. The constant of proportionality is fixed by fitting the theory to one experimental data point.

It is a general result of theories for pure  ${}^{4}$ He, notably the works of Ginsburg and Pitaevskii and<br>Mamaladze,<sup>9</sup> and Kosterlitz and Thouless,<sup>10</sup> that  $M$ amaladze,<sup>9</sup> and Kosterlitz and Thouless,<sup>10</sup> that for fixed  $T$  a system of reduced dimensionality characterized by a length  $d$  (e.g., a film of thickness  $d$ , or a fluid in a pore of radius  $d$ ) will not hess *a*, or a fluid in a pore of radius *a*) will no<br>display superfluidity for  $d < d_s$   $\sim \xi$ , where  $\xi$  is a coherence length and  $d_s$  is the critical length. Further,  $\xi$  is related to the superfluid densit by the equation

$$
\rho_s \xi / T = \text{const.} \tag{9}
$$

Putting together the two ideas produces

$$
\rho_s d_s / T = \text{const.} \tag{10}
$$

For films in which spatial variations in  $\rho_s$  play a role (the case for all very thin films), it is certainly appropriate to generalize (10) to read

$$
\sigma_s / T = \text{const} \equiv \gamma_c \,, \tag{11}
$$

where  $\sigma_s$  is the superfluid density per unit area.

The constant in  $(11)$  will depend on the geometry. If one uses the relation  $d_s = \pi \xi$  of Ginsburg and Pitaevskii for planar films and the standard definition $11$  of the coherence length, one obtains

$$
\gamma_c = \frac{m_4^2 k_B}{4\hbar^2} = \text{const} = 0.138 \times 10^{-8} \text{g/cm}^2 \text{ K}.
$$
 (12)

In view of the rather unsettled theoretical situation (the theories of Refs. 9 and 10 are based on rather different physical ideas), we view (12) as providing solely an order-of-magnitude estimate. This viewpoint is certainly supported by the available data on transitions in superfluid systems of reduced dimensionality. Scholtz, Mclean, and Rudnick<sup>4</sup> fit a collection of data for planar  ${}^{4}$ He films for  $T \ge 1$  K with  $\gamma_c = 0.298 \times 10^{-8} \text{g/cm}^2$  K. Thomlinson, Ihas, and Pobell<sup>12</sup> have done a detailed study of superfluid flow of  ${}^{3}$ He $-{}^{4}$ He solutions in cylindrical pores, where the geometrical correction of Ref. 9 changes the theoretical  $\gamma_c$ 

to  $0.213 \times 10^{-8}$ g/cm<sup>2</sup>K. Their experimental result is  $\gamma_c = 0.66 \times 10^{-8} \text{g/cm}^2 \text{K}$  independent of <sup>3</sup>He concentration. The data of Chan, Yanof, and Reppy' for 'He films in Vycor pores (where the geometry is not well characterized) may be fit with a  $\gamma_c \approx 0.7 \times 10^{-8} \text{g/cm}^2 \text{K}.$ 

The local-continuum model allows us to calculate  $\sigma_s$  by calculating  $\rho_s(P,X,T) = \rho_s(z,P_0,X_0,T)$ given  $\rho_s$  as a function of P, X, and T in bulk sysgiven  $\rho_s$  as a function of P, X, and T in bulk<br>tems, and then integrating.<sup>13</sup> Therefore, we replace (11) by

$$
\int_{z_{s}}^{e_{s}(T,X_{0},P_{0})} \frac{\rho_{s}(z,P_{0},X_{0},T)}{T} \ dz = \gamma_{c}.
$$
 (13)

This completes our modification of the local continuum theory. The critical constant  $\gamma_c$  is obtained by fitting to a single data point, i.e., a point for fixed  $P_0$ ,  $X_0$ , and T. That the theory should be expected to correctly fit the data for other concentrations is the essence of the concept of universality.<sup>6</sup> Calculational details and our explicit results, which do indeed agree with universality, are given in Sec. III.

## III. CALCULATIONAL DETAILS AND RESULTS

Prior to describing our results, we shall summarize the experimental data used as input for our calculations. In deriving the functions  $P(z, X_0, P_0, T)$  and  $X(z, X_0, P_0, T)$ , we needed experimental data for the quantities  $\gamma(X, T, P)$ ,  $v_3(X, T, P)$  and  $v_4(X, T, P)$ . For  $\gamma(X, T, P)$  $=(X/k_BT)(\partial \mu_3/\partial X)_T$ , we have used the experimental results of de Bruyn Ouboter *et al*,<sup>14</sup> w mental results of de Bruyn Ouboter  ${\it et\ al,}^{14}$  who give an empirical expression for  $\mu$ , to derive

$$
\gamma(X,\,T,\,P)=1-3.08(1-X)X/T.\tag{14}
$$

We estimate this analytic expression to be valid to within roughly 10% throughout the domain  $(X \le 0.75, 0.9 \le T \le 1.2 \text{ K}, P \le 25 \text{ atm}) \text{ of concen-}$ trations, temperatures, and pressures of interest here. The quantities  $\Delta v = v_3 - v_4$  and  $v_4$  have been used in the forms

$$
\Delta v = B - 2AP, \tag{15}
$$

$$
v_4 = B_4 - 2A_4P, \t\t(16)
$$

where the constants  $B$ ,  $A$ ,  $B_4$ , and  $A_4$  take on different numerical values for different pressure intervals, as given in Table I. These constants are temperature and concentration independent to within effects of the order of  $1\%$  in the domains of interest to us here; the data in Table I are derived from results for pure <sup>3</sup>He and pure<br><sup>4</sup>He.<sup>15</sup>  $^{4}$ He.<sup>15</sup>

Our results for  $X(z, X_0, P_0, T)$  and  $P(z, X_0, P_0, T)$ for several values of T and  $X_0$  and  $P_0 = 0$  are plotted in Figs. 1 and 2. Note that the important

TABLE I. Values of the parameters used in the calculations of He molar volumes in several ranges of pressure  $V_4/R = B_4 - 2A_4P$  and  $(V_3 - V_4)/R = B - 2AP$  with R =8.32,  $V_4$  and  $V_3$ (cm<sup>3</sup>), and P (atm).

	А	B	$A_{\Lambda}$	$B_{4}$
$0 \leq P \leq 2$ atm	0.05308	1.126	0.01576	3.319
$2 \leq P < 5$ atm	0.02797	1.025	0.01576	3.319
$5 \leq P < 10$ atm	0.015 04	0.8964	0.01241	3.298
$10 \leq P < 15$ atm	0.00866	0.7695	0.00971	3.244
$15 \leq P < 25$ atm	0.004964	0.6587	0.00739	3.174

variations occur within about six atomic layers from the wall. The curves terminate at a distance of approximately 1.<sup>5</sup> layers from the wall, where the theory predicts solidification. The predicted solid-layer thickness is quite consistent with previous experimental estimates of the solidpredicted solid-layer thickness is quite cons<br>with previous experimental estimates of the<br>layer thickness in pure <sup>4</sup>He.<sup>4,5</sup> (Note that the system studied here is almost pure 4He when the predicted solidification occurs. )

The next quantity calculated is the local value of  $T<sub>1</sub>$ . This is taken from bulk measurements of of  $T_{\lambda}$ . This is taken from bulk measurements of  $T_{\lambda}(X,P),$ <sup>16</sup> which yield the approximate analytical expressions

 $T_{\lambda} = 2.5256 - 2.4577X - 0.0059P$ ;  $0.50 \le X \le 0.673$ .

 $T_{\lambda} = 2.38 - 2.17X - 0.0059P$ ;  $0.35 \le X \le 0.50$ .

 $T_{\lambda} = 2.214 - 1.68X - 0.0127P$ ;  $0.161 \le X \le 0.35$ ,

 $T_{\lambda} = 2.17 - 1.41X - 0.0127P$ ;  $0 \le X \le 0.161$ .  $(17)$ 

Two typical results for  $T_{\lambda}(z, X_0, P_0, T)$  are plotted in Figs. 3 and 4. The temperature  $T$  is also plotted in order that one may easily see the region of superfluidity predicted by the local-continuum theory.

In order to apply the correlation-length corrections to the local-continuum theory we require an expression for the superfluid density  $\rho_s(X, T, P)$ . At present both data and systematic theory for  $\rho_s$  are lacking over the rather extensive ranges of  $X$ ,  $T$ , and  $P$  required here. Only soranges of X, T, and P required here. Only s<br>lutions dilute in  ${}^{3}$ He,<sup>17</sup> and the  $\lambda$  and tricritica regions of the phase diagram (see, e.g., Ref. 12), have received careful attention. Fortunately, we do not require extreme accuracy throughout the  $X, P, T$  domain of interest in computing the integral in (13). For example, contributions to (13) coming from the  $\lambda$  region are not so important as to warrant use of the well-known  $T$ dependence  $\rho_s \sim (T_\lambda - T)^{2/3}$  there. The exponential T dependence arising from rotons is sufficient. Our approach is to use a semi-empirical model, which we believe to be entirely new, in which the normal fluid contains only contributions from rotons and <sup>3</sup>He atoms denoted by  $\rho_{rn}$  and  $\rho_{3n}$ , respectively; phonon contributions are neglected in the temperature range considered. Thus, we write

$$
\rho_s = \rho - \rho_{3n} - \rho_{rn} \,. \tag{18}
$$

The roton part has the form<sup>18</sup>

$$
\rho_{rn} = \left[ \alpha(P)/T^{1/2} \right] e^{-\Delta(P)/T}.
$$
 (19)

The function  $\alpha(P)$  depends upon universal constants and the effective mass  $\mu_r(P)$ , and  $\Delta(P)$  is. the roton gap. Both  $\mu_r(P)$  and  $\Delta(P)$  are taken to depend upon P alone, their dependences on X and T being known to be weak.<sup>19</sup> We use  $T$  being known to be weak. $^{19}$  We use

$$
\alpha = 8.7 + 0.065P \text{ (cgs units)},
$$
  
\n
$$
\Delta = 8.65 - 0.0082P \text{ (P in atm)},
$$
\n(20)

 $\Delta = 0.0002F$  ( $F$  in atm),<br>derived from the data given in the Wilks book.<sup>18</sup> The condition that  $\rho_s = 0$  at  $T = T_\lambda(X, P)$  yields

$$
\rho_{3n} = \rho - \frac{\alpha(P)}{T_{\lambda}^{1/2}(X,P)} e^{-\Delta(P)/T_{\lambda}(X,P)}, \qquad (21)
$$

at the  $\lambda$  transition. Neglecting the weak dependences of  $\rho_{3n}$  and  $\rho$  on T, we may combine (18), (19), and (21) to arrive at the result

$$
\rho_s = \frac{\alpha(P)}{\left[T_\lambda(X,P)\right]^{1/2}} e^{-\Delta(P)/T_\lambda(X,P)} - \frac{\alpha(P)}{T^{1/2}} e^{-\Delta(P)/T}.
$$
\n(22)

Note that this result submerges all of the  $X$  dependence of  $\rho_{3n}$  (and hence that of  $\rho_s$ ) in  $T_\lambda(X,P)$ , for which we have already given an expression in Eq. (17). Two typical curves for  $\rho_s(z,X_0,P_0,T)$ are plotted in Figs. 3 and 4.

Given the calculated values for  $\rho_s(z, P_0, X_0, T)$ we can numerically compute the integral given in (13), which provides us with the correlationlength correction to the local-continuum theory. The critical constant  $\gamma_c$  has been fixed by calculating the value of the integral for one of the experimentally determined points at which wall film superfluidity first appears. This point has coordinates  $X_0 = 0.65$ ,  $T = 1.01$  K. All points have  $P_0 \approx 0$  so we omit further reference to values of  $P_0$ . To obtain the remaining points we fixed  $X_0$ and varied  $T$  until the integral took on the value  $\gamma_c$  once again. The resulting T-vs-X curve is shown in Fig. 5, along with the available experimental data points. '

The agreement between theory and experiment is very good over most of the range of applicability of the theory. The agreement is least satisfactory for the lower temperatures. However, this is to be expected since for temperatures near and below 0.87 <sup>K</sup> tricritical and phase separation effects will become important, invalidating a theory based on a coherence length related to superfluidity alone.



FIG. 5. Phase diagrams of  ${}^{3}\text{He}-{}^{4}\text{He}$  mixtures. The experimental measurements of superfluid onset temperature for a given bulk concentration {circles, Ref. 20 and other data, Ref. 1) are presented with results of our calculations (dotted line) using  $\sigma_s/T = 0.27 \times 10^{-8} \text{ g/cm}^2 \text{ K.}$ In the insert, the dashed line shows the calculated superfluid onset curve, neglecting coherence-length effects.

It is instructive to compare the predictions of the local-continuum theory alone, without the modifications developed here. These predictions are shown as a dashed line in Fig. 5 and lie well above the experimental data. The reason that the dashed line intersects the  $\lambda$  line is discussed in detail in the second of Refs. 1. In essence what happens is that the relative importance of P and X in determining the local  $\lambda$  line near the container walls is reversed. The local  $\lambda$  temperature falls rather than rises as the container wall is approached, and the  $\lambda$  transition commences in the bulk far from the walls, a film of mences in the bulk far from the walls, a fill<br>normal fluid remaining near the walls.<sup>20</sup> Of course, the present modified theoretical curve has the same intersection with the  $\lambda$  line as does that of the unmodified theory; on the scale of the figure this is impossible to depict, however.

The value of  $\gamma_c$  determined by the procedure just described is  $\gamma_c = 0.27 \times 10^{-8} \text{g/cm}^2 \text{K}$ . This is well within a factor of <sup>2</sup> of the estimate based on previous theories [see Eq. (12}]. More importantly, the result is quite close to the previously discussed (in Sec. II) value of 0.298  $\times10^{-8}$ g/cm<sup>2</sup>K obtained by Scholtz, Mclean, and Rudnick<sup>4</sup> for pure unsaturated planar  $4$ He films for  $T \ge 1$  K. The difference between the two values is within the approximate 10% accuracy of the data used as input for the local-continuum theory. The essential equality of the two values should be regarded as an important verification of the universality concept,<sup>6</sup> especially when it is noted that the 'He films possess a free surface, whereas the mixture films dealt with here do not.

Our theory also yields the thickness  $e_s(T, X_o, P_o)$ of the superfluid wall films at the point at which they become superfluid. The results are plotted in Fig. 6.

### IV. SUMMARY AND DISCUSSION

We have presented a theory of the nucleation of superfluid transitions in liquid  ${}^{3}$ He $-{}^{4}$ He mixtures near container walls. The theory is based on the local-continuum model supplemented by coherence-length corrections accounting for the reduced dimensionality of the nucleating wall films. Choosing the critical constant  $\gamma_c$  of the theory to fit a single data point was sufficient to provide an excellent fit to all the data in the range of applicability of the theory, in agreement with the idea of universality. That the soobtained  $\gamma_c$  is essentially the same as that found for unsaturated pure 'He films provides further evidence in favor of universality.



FIG. 6. Thickness of the superfluid phase on the superfluid onset curve, shown as the dotted curve in Fig. 5.

To our knowledge the application of the localcontinuum theory to account carefully for the spatial variation of  $\rho_s$  within the wall films is the first time such spatial variations have been carefully treated in the field of superfluid film physics. The importance of these variations is evi-

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- <sup>1</sup>The first of these papers, by M. Chester, J. P. Laheurte, and J. P. Romagnan [Phys. Rev. B 14, 2812 (1976)] developed the theory for solutions dilute in either  ${}^{3}$ He or  ${}^{4}$ He. The second paper, by J. P. Laheurte, J. P. Romagnan, and H. Dandache in [J, Phys. (Paris) 38, 59 (1977)] generalized the work to the case of arbitrary <sup>3</sup>He concentrations.
- $2$ In this case important nonlocal effects occur. A recent work giving a theory of such effects for classical fluids is C. Ebner, W. F. Saam, and D. Stroud [Phys. Rev. A. 14, 2264 (1976)]. This paper contains many further references to work on both quantum and classical systems.
- 3A11 data available to date are reviewed in Ref. l.
- <sup>4</sup>J. H. Scholtz, E. O. Mclean, and I. Rudnick, Phys. Rev. Lett. 32, 147 (1974).
- <sup>5</sup>M. H. W. Chan, A. W. Yanof, and J. D. Reppy, Phys. Rev. Lett. 32, 1347 (1974).
- ${}^{6}$ L. P. Kadanoff, in Proceedings of the International School of Physics "Enrico Fermi" Course LI, edited by M. S. Green (Academic, New York, 1973); R. B. Griffiths, Phys. Rev. Lett. 24, 1479 (1970).
- This is actually the value appropriate for Vycor glass [see E. S. Sabisky and C. H. Anderson, Phys. Rev. Lett. 30, 1122 (1973)]. However, it is quite close to values calculated for metals (Cu containers were used in the experiments relevant to our theory) by E. S. Sabisky and C. H. Anderson [Phys. Rev. <sup>A</sup> 7, 790  $(1973)$ , and small changes in  $\alpha$  are not very significant producing only changes in the length scale determined by  $\alpha^{1/3}$ .
- ${}^{8}$ This is not precisely true in that at the higher temperatures considered here there is maximum in  $T_{\lambda}(P,x)$ very near the liquid-solid boundary (see Fig. 4). This fact does not materially affect our arguments.
- $9V.$  L. Ginzburg and L. P. Pitaevskii, Zh. Eksp. Teor. Fiz. 34, 1240 (1958) [Sov. Phys.-JETP 7, 858 (1958)]; Yu. G. Mamaladze, ibid. 52, 729 (1967) [ibid. 25, 479  $(1967)$ ].
- $^{10}$ J. M. Kosterlitz and D. J. Thouless, J. Phys. C  $6$ ,

dent in Figs. 3 and 4.

We hope to extend our work to the case of wall film nucleation at lower temperatures ( $T \le 0.87$  K) where accounting for tricritical and phase separation phenomena in the films becomes impor $tant.<sup>1</sup>$ 

1181 (1973).

- <sup>11</sup>R. A. Ferrell, N. Menyhard, H. Schmidt, F. Schwabl, and P. Szépfalusy, Ann. Phys. (N.Y.) 47, 565 (1968); B. I. Halperin and P. C. Hohenberg, Phys. Rev. 117, 952 (1969); M. E. Fisher, M. N. Barber, and D. Jasnow, Phys. Rev. A 8, 1111 (1973).
- $12W$ . C. Thomlinson, G. G. Ihas, and F. Pobell, Phys. Rev. B 11, 4292 (1975).
- $^{13}$ At low temperatures and for thin films this would not be an appropriate procedure. In this case phonons having spectra crucially dependent on the thickness of the film would play an important role. See, e.g., W. F. Saam and M. W. Cole, Phys. Rev. B 11, 1086 (1975}. Calculations and comments on the thin-filr' case are also given by M. Chester and L. Eytel, ibid. 13, 1069 (1976}.
- <sup>14</sup>R. de Bruyn Ouboter, J. J. M. Beenakker, and K. W. Taconis, Physica (Utr.) 25, 1162 (1959).
- $15$ See, e.g., J. Wilks, The Properties of Liquid and Solid Helium (Clarendon, Oxford, 1967), Tables A2 and A10.
- <sup>16</sup>T. A. Alvaselo, P. M. Bergland, S. T. Islander, G. R. Pickett, and W. Zimmerrnan, Jr., Phys. Rev. <sup>A</sup> 4, 2354 (1971); C. LePair, K. W. Taconis, R. De Bruyn Ouboter, P. Das, and E. De Jong, Physica (Utr.) 5, 764 (1965); K. N. Zinov'eva, Zh. Eksp. Teor. Fiz.
- 44, 1837 (1963) [Sov. Phys. —JETP 17, 1235 (1963)] .  ${}^{17}$ See, e.g., D. O. Edwards and C. Ebner, Phys. Rep. 2c-2, 78 (1971).
- $^{18}$ See Ref. 15, Chap. 5 and Table A5.
- $^{19}$ C. M. Surko and R. E. Slusher, Phys. Rev. Lett.  $30$ , 1111 (1973); R. L. Woerner, D. A. Rockwell, and T. J. Greytak, ibid. 30, 1114 (1963); J. M. Rome, D. L. Price, and G. E. Ostrowski, ibid. 31, 510 (1973}. The work of these authors verifies our statement for  $X<sub>0.31</sub>$ . We simply assume no important concentration dependence for the larger values of  $X$  involved in our work, noting (see Figs. 3, 4) that smaller values of  $X$ are rather heavily weighted in the integrals over  $\rho_s$ .
- $20$ This crossover behavior has been observed in the experiments of C. A. Gearhart, Jr., and W. Zimmerman, Jr., Phys. Lett. <sup>A</sup> 48, <sup>49</sup> (1974).