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

J.-P. Laheurte and J.-P. Romagnan

Laboratoire de Physique de la Matière Condensée, Université de Nice, Parc Valrose, 06034-Nice, France

W. F. Saam*

Department of Physics, Ohio State University, Columbus, Ohio 43210 and Laboratoire de Physique de la Matière Condensée, Université de Nice, Parc Valrose, 06034-Nice, France (Received 13 December 1976)

A theory of the nucleation of superfluid transitions near container walls of ${}^{3}\text{He}{}^{4}\text{He}$ 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 papers¹ 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.² 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 ³He-⁴He mixtures predicts that even for a mixture nominally quite rich in ³He, the local ³He concentration near a container wall will be rather small ($\leq 8\%$). This leads to the conclusion that, for a certain range of temperatures T and ³He 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,³ this description cannot be correct. Bulk properties of superfluid ⁴He, for example, certainly do not suffice to predict λ temperatures for thin ⁴He films. The λ transition for bulk ⁴He is 2.17 K, whereas transitions in very thin (thickness approximately equal to a few atomic layers) films occur at temperatures of the order of 0.1–0.2 K.^{4,5} 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, ⁴He 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 ⁴He.

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 ρ_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 universality⁶ 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 ρ_s is developed. Section IV summarizes our work and contains additional discussion.

II. BASIC THEORY

The local-continuum theory¹ begins with the three exact equilibrium conditions for ³He⁻⁴He mixtures. These conditions require that the ³He chemical potential μ_3 , the ⁴He chemical potential μ_4 , and the temperature be constant throughout the system. Let $U(\mathbf{\hat{r}})$ be an external potential, the same for both helium isotopes. The theory approximates μ_3 and μ_4 by their local values plus contributions from $U(\mathbf{\hat{r}})$, i.e.,

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

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

Here T is everywhere constant, the pressure P and ³He number concentration X vary with \vec{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

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

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

in which

$$v = Xv_3 + (1 - X)v_4,$$
 (4)

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

$$\gamma(X, T, P) = \frac{X}{k_B T} \left(\frac{\partial \mu_3}{\partial X}\right)_{T, P}.$$
 (6)

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

We are specifically interested in the case where $U(\mathbf{r})$ 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

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

The value $\alpha = 27 \text{ K/(layer)}^3$ for the Van der Waals constant⁷ has been used in our computations.

Data from measurements on bulk systems give v, Δv , and γ as functions of T, X, and P. Putting Δv and γ 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 $z = z_s$ 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_s 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 λ 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_{λ} drops abruptly to zero.⁸ 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

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

Given that all T_{λ} curves of importance here possess the monotonicity property just mentioned,⁸ 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 ³He 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 distances $z \le 1.5$ atomic 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 superfluid transition temperatures. $T_{\lambda}(z) = T_{\lambda}(\mathbf{x}z)$, P(z), T) and superfluid density ρ_s vs z, the distance from the walls, for $X_0 = 0.7$ and T = 0.965. The temperature at which the calculations are done is represented as a dotted line. ρ_s is different from zero at distances such that $T \leq T_{\lambda}(z)$.



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

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 λ temperature of a system is independent of its thickness. This, as discussed in Sec. I, is in complete contradiction with the data for ⁴He films^{4,5} as well as with modern theory.^{9,10} 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 $e_s(T, X_0, P_0)$ 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 ⁴He, notably the works of Ginsburg and Pitaevskii and Mamaladze,⁹ and Kosterlitz and Thouless,¹⁰ 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 display superfluidity for $d < d_s \sim \xi$, where ξ is a coherence length and d_s is the critical length. Further, ξ is related to the superfluid density 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 ρ_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 σ_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 definition¹¹ 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 Rudnick⁴ fit a collection of data for planar ⁴He films for $T \ge 1$ K with $\gamma_c = 0.298 \times 10^{-8}$ g/cm² K. Thomlinson, Ihas, and Pobell¹² have done a detailed study of superfluid flow of ³He-⁴He solutions in cylindrical pores, where the geometrical correction of Ref. 9 changes the theoretical γ_c to 0.213×10^{-8} g/cm² K. Their experimental result is $\gamma_c = 0.66 \times 10^{-8}$ g/cm² K independent of ³He concentration. The data of Chan, Yanof, and Reppy⁵ for ⁴He films in Vycor pores (where the geometry is not well characterized) may be fit with a $\gamma_c \approx 0.7 \times 10^{-8}$ g/cm² K.

The local-continuum model allows us to calculate σ_s by calculating $\rho_s(P, X, T) = \rho_s(z, P_0, X_0, T)$ given ρ_s as a function of P, X, and T in bulk systems, and then integrating.¹³ 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 γ_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.⁶ 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 $\gamma(X, T, P) = (X/k_BT)(\partial \mu_3/\partial X)_{T,P}$ we have used the experimental results of de Bruyn Ouboter *et al*,¹⁴ who give an empirical expression for μ_3 , to derive

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

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

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

$$v_4 = B_4 - 2A_4P, \tag{16}$$

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 ³He and pure ⁴He.¹⁵

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 (cm³), and P (atm).

	A	В	A ₄	B ₄
$0 \le p \le 2$ atm	0.053 08	1.126	0.01576	3.319
$2 \leq P \leq 5 \text{ atm}$	0.02797	1.025	0.01576	3.319
$5 \leq P < 10 \text{ atm}$	0.01504	0.8964	0.01241	3.298
$10 \leq P \leq 15 \text{ atm}$	0.008 66	0.7695	0.00971	3.244
$15 \le P < 25$ atm	0.004 964	0.6587	0.007 39	3.174

variations occur within about six atomic layers from the wall. The curves terminate at a distance of approximately 1.5 layers from the wall, where the theory predicts solidification. The predicted solid-layer thickness is quite consistent with previous experimental estimates of the solidlayer thickness in pure ⁴He.^{4,5} (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_{λ} . This is taken from bulk measurements of $T_{\lambda}(X,P)$,¹⁶ which yield the approximate analytical expressions

 $T_{\lambda} = 2.5256 - 2.4577X - 0.0059P; \quad 0.50 \le X \le 0.673,$ $T_{\lambda} = 2.38 - 2.17X - 0.0059P; \quad 0.35 \le X \le 0.50.$

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

 $T_{\lambda} = 2.17 - 1.41X - 0.0127P; \quad 0 \le X \le 0.161.$ (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 ρ_s are lacking over the rather extensive ranges of X, T, and P required here. Only solutions dilute in ${}^{3}\text{He}, {}^{17}$ and the λ and tricritical 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 λ region are not so important as to warrant use of the well-known Tdependence $\rho_s \sim (T_{\lambda} - T)^{2/3}$ 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 ³He atoms denoted by ρ_{rn} and ρ_{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 form¹⁸

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

$$\alpha = 8.7 + 0.065P \quad (cgs units), \tag{20}$$

$$\Delta = 8.65 - 0.0082P \quad (P \text{ in atm}),$$

derived from the data given in the Wilks book.¹⁸ 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 λ transition. Neglecting the weak dependences of ρ_{3n} and ρ on *T*, we may combine (18), (19), and (21) to arrive at the result

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

Note that this result submerges all of the X dependence of ρ_{3n} (and hence that of ρ_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 γ_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 γ_c once again. The resulting T-vs-X curve is shown in Fig. 5, along with the available experimental data points.¹

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 K 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 $\sigma_{s}/T = 0.27 \times 10^{-8}$ g/cm² 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 λ 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 λ line near the container walls is reversed. The local λ temperature falls rather than rises as the container wall is approached, and the λ transition commences in the bulk far from the walls, a film of normal fluid remaining near the walls.²⁰ Of course, the present modified theoretical curve has the same intersection with the λ line as does that of the unmodified theory; on the scale of the figure this is impossible to depict, however.

The value of γ_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 2 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 $\times 10^{-8}$ g/cm² K obtained by Scholtz, Mclean, and Rudnick⁴ for pure unsaturated planar ⁴He 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,⁶ especially when it is noted that the ⁴He films possess a free surface, whereas the mixture films dealt with here do not.

Our theory also yields the thickness $e_s(T, X_0, P_0)$ 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 ${}^{3}\text{He}-{}^{4}\text{He}$ 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 γ_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 γ_c is essentially the same as that found for unsaturated pure ⁴He 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 ρ_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-

*Work supported in part by NSF Grant No. DMR75-21866.

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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 important.¹

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- 44, 1837 (1963) [Sov. Phys.-JETP <u>17</u>, 1235 (1963)]. ¹⁷See, e.g., D. O. Edwards and C. Ebner, Phys. Rep. <u>2c-2</u>, 78 (1971).
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- ¹⁹C. M. Surko and R. E. Slusher, Phys. Rev. Lett. <u>30</u>, 1111 (1973); R. L. Woerner, D. A. Rockwell, and T. J. Greytak, *ibid.* <u>30</u>, 1114 (1963); J. M. Rowe, D. L. Price, and G. E. Ostrowski, *ibid.* <u>31</u>, 510 (1973). The work of these authors verifies our statement for X < 0.31. 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 ρ_s .
- ²⁰This crossover behavior has been observed in the experiments of C. A. Gearhart, Jr., and W. Zimmerman, Jr., Phys. Lett. A 48, 49 (1974).

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