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Stability of quantum-fluid mixtures

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The absolute stability of quantum-fluid mixtures is examined in terms of long-range correlations in the wave function. Divergent long-range fluctuations in the Jastrow part of the ground-state wave function are shown to signal the location of the spinodal point, i.e., the limiting concentration for supersaturation. Sample results are presented for two-component boson fluids.

I. INTRODUCTION

Considerable interest has been shown recently in a number of different quantum fluids, including mixtures such as ^{3}He in ^{4}He, spin-aligned ^{3}He in ^{4}He, spin-aligned atomic hydrogen in ^{4}He, and the mixture of spin-aligned atomic hydrogen with spin-aligned atomic deuterium or tritium.[1-4] Of particular importance in the mixture case is the question of whether the components will actually mix at any finite concentration.[5]

It has long been known that isotopic bosons such as spin-aligned hydrogen and spin-aligned tritium will not mix at all at zero temperature.[6] Crudely speaking, the argument is that, since the interaction potential is identical between all of the isotopic particles, the enhanced zero-point motion of the lighter-mass particle when it is in solution, due to caging at the preferential higher number density of the heavier-mass particle, can only be overcome by complete phase separation. In the case where one (or both) of the isotopes is a fermion, this tendency to phase separate is partially compensated by the fact that the Fermi surface of the phase-separated state is larger than in the mixed case, thus favoring mixing.[3] Thus ^{4}He will admit up to a 6% concentration of ^{3}He at $T=0$. This limit is essentially established by the concentration at which the increased Fermi energy of the phase-separated system is equal to the increased kinetic energy due to caging in the mixed state.[3]

The complete phase separation of isotopic boson mixtures has been substantiated by Miller in his careful work on the quantum theory of corresponding states in mixtures.[1,2] It is also shown there that it is possible to have a finite concentration of one boson component in another if the relative strengths of the interaction potentials is favorable. In particular, an increased attraction between unlike particles promotes mixing.[1,7] To make those calculations practical, Miller used the average correlation approximation (ACA) in which the spatial correlations in the wave function are independent of the differences between the constituents. While this permits a survey of a wide range of systems, the results become less trustworthy as the physical differences between the constituents increases.[2]

In this paper we discuss a complementary theory which is particularly well suited to examining the properties of a given mixture, even in the case when the constituents are substantially different. We examine the generalization to multicomponent systems[8] of the paired-phonon analysis (PPA) or, equivalently, the Jastrow Euler-Lagrange analysis,[9,10] of the ground state of the system of interest. In contrast to the ACA, this provides a maximum flexibility in the correlation between the particles. An important conclusion of the present analysis is the ease with which it identifies the absolute mechanical instability with respect to phase separation, i.e., the location of the spinodal curve (at $T=0$). Knowing this determines the maximum concentration for a supersaturated solution. While this is relatively easy to obtain if one knows the enthalpy as a function of concentration, since it corresponds to the onset of negative curvature, that information is difficult to produce, except in the ACA, because of the necessity of converting from the density variable to the pressure variable.

II. STABILITY CRITERION

It has been shown that there is an intimate connection between the mechanical stability of a quantum fluid and the long-wavelength behavior of its collective mode and its liquid structure function. On the one hand, one knows that the zero-point motion of the phonons appears in the ground-state wave function in the form of a Jastrow factor.[11-13] Moreover, a structural instability of the fluid may be associated with the softening of the collective mode.[13] Similarly, the liquid structure function $S(k)$ is the susceptibility for coupling to the phonon, and thus will show an anomaly at a structural instability.

The latter effect is best known from the divergence

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of $S(0)$ at the liquid-gas critical point, producing critical opalescence. It has also been exhibited in recent theoretical work on the ground state of liquid ^{4}He (Ref. 14) and liquid ^{3}He,[9] where the mechanical instability occurs (as the density is lowered into the negative pressure regime) at the density where the compressibility K diverges. This cavitation instability would only be accessible experimentally by superexpanding the fluid. In contrast with finite temperature cases such as the liquid-gas critical point, the anomalous behavior of $S(k)$ does not appear in its value at $k=0^+$ (which must strictly vanish at zero temperature) but rather in the slope of $S(k)$ at $k=0$, which is proportional to $K^{1/2}$ and thus diverges at the density of the instability. Correspondingly one finds that the sound velocity c vanishes, producing a softening of the phonon energy $\hbar ck$. The connection between this long-wavelength behavior of $S(k)$ and the excitation spectrum $\epsilon(k)$ is given as always, by the Bijl-Feynmann relation[15]

$$\epsilon(k)=\frac{\hbar^2k^2}{2mS(k)} \quad , \tag{1}$$

which is exact in the long-wavelength limit.

We wish to show here that the analogous situation occurs in quantum fluid mixtures. In addition to the cavitation instability, there is also the possibility of a long-wavelength instability corresponding to absolute mechanical instability with respect to phase separation in a two-component mixture. Generally, the number of possible instabilities of this sort equals the number of components in the mixture. In order to discuss these instabilities in the mixture, we will briefly review the generalization to multicomponent systems of the relationships between the density fluctuation spectra, the structure of the ground-state wave function, and the liquid structure functions.[8,16] A matrix notation is used to simplify the discussion.

Each of the n components of the mixture has a density fluctuation operator $\rho^{\alpha}_{\vec{k}}$, $\alpha=1,\ldots,n$. In the long-wavelength limit, the operator producing each of the n collective modes is a linear combination of these density fluctuation operators. The generalization of the Bijl-Feynmann equation for the dispersion relations is obtained by diagonalizing the $n\times n$ matrix

$$\epsilon(k)=S^{-1/2}(k)\epsilon_0(k)S^{-1/2}(k) \quad , \tag{2}$$

where $\epsilon_0(k)=\mathrm{diag}(\hbar^2k^2/2m_\alpha)$, and $S(k)$ is the structure function matrix, with elements

$$S_{\alpha\beta}(k)=(N_\alpha N_\beta)^{-1/2}\langle\psi_0|\rho^{\alpha}_{\vec{k}}\rho^{\beta}_{-\vec{k}}|\psi_0\rangle \quad , \tag{3}$$

where ψ_0 is the normalized ground state. For a system with short-range interactions, the long-wavelength behavior of each component is linear[16]:

$$\lim_{k\to 0} S_{\alpha\beta}(k)=\gamma_{\alpha\beta}k \quad , \tag{4}$$

as is each of the dispersion relations[16]:

$$\lim_{k\to 0}\epsilon_\alpha(k)=\hbar kc_\alpha \quad . \tag{5}$$

In the case of a two-component system, the in-phase mode is a total density fluctuation mode, with velocity c_+, while the out-of-phase mode is the concentration fluctuation, with velocity c_-. Here we wish to point out that a long-wavelength instability to phase separation in a mixture is signaled by the vanishing of c_-, or, equivalently, by the divergence in the slopes $\gamma_{\alpha\beta}$ of the components of the liquid structure functions at long wavelengths.

To formulate this stability criterion in a manner useful for application in a calculation, we use the fact that, as in the single-component quantum fluid, the zero-point motion of the collective modes appears in the ground-state wave function as a (multicomponent) Jastrow factor. Thus, if we suppose that the system has ground-state wave function ψ_0, we can examine its stability against a small displacement of the Jastrow type:

$$\psi_u(\{\vec{r}_i\})=\left(\exp\frac{1}{2}\sum_{\alpha\leq\beta}^{n}\sum_{\langle i_\alpha,i_\beta\rangle}u_{\alpha\beta}(\vec{r}_{i_\alpha}-\vec{r}_{i_\beta})\right)\psi_0(\{\vec{r}_i\}) \quad . \tag{6}$$

A necessary condition that ψ_0 be the ground state is that is satisfy the matrix of conditions

$$\left.\frac{\delta}{\delta u_{\alpha\beta}(r)}\frac{\langle\psi_u|H|\psi_u\rangle}{\langle\psi_u|\psi_u\rangle}\right|_{u=0}=0 \quad , \tag{7}$$

which can be written in the convenient form[8]

$$S'(k)=\tfrac{1}{4}\{[1-S(k)]\epsilon_0(k)+\epsilon_0(k)[1-S(k)]\} \quad , \tag{8}$$

where the matrix S' is defined by

$$S'_{\alpha\beta}(k)=(N_\alpha N_\beta)^{-1/2}\langle\psi_0|V^*[\rho^{\alpha}_{\vec{k}}\rho^{\beta}_{-\vec{k}}-(N_\alpha N_\beta)^{1/2}S_{\alpha\beta}(k)]|\psi_0\rangle \quad , \tag{9}$$

and $V^*(\{\vec{r}_i\})$ is the Jackson-Feenberg function whose expectation value is the energy of the system

$$V^*(\{\vec{r}_i\}) = V(\{\vec{r}_i\}) - \sum_i \frac{\hbar^2}{4m_i} \nabla_i^2 \ln\psi_0^2 \ . \tag{10}$$

This set of equations is the multipcomponent generalization[8] of the Euler-Lagrange equations for the optimum Jatrow factor in the ground state of a quantum fluid.[17,18] The prime in Eq. (9) signifies that $S'_{\alpha\beta}$ can be obtained as the ξ derivative of $S_{\alpha\beta}(k,\xi)$ defined for the wave function $\psi = \exp\frac{1}{2}(\xi V^*)\psi_0$. Note from Eq. (9) that $S'(k)$ is a fluctuation function, and thus might be expected to exhibit anomalous properties at an instability. Moreover, it can be seen from the right side of Eq. (8) that $S'(k)$ vanishes as k^2 at small k. As in the theory of critical points, it is better to work with direct correlation functions (i.e., non-nodal functions) $X_{\alpha\beta}(k)$ and their ξ derivative $X'_{\alpha\beta}(k)$:

$$X(k) = 1 - S^{-1}(k) \ , \tag{11}$$

$$X'(k) = S^{-1}(k)S'(k)S^{-1}(k) \ . \tag{12}$$

Then Eq. (8) can be converted to the useful result

$$(\epsilon_0^{1/2}S^{-1}\epsilon_0^{1/2} - \tfrac{1}{2}\epsilon_0)^2 = 2\epsilon_0^{1/2}X'\epsilon_0^{1/2} + \tfrac{1}{4}\epsilon_0^2 \equiv \Omega(k) \ . \tag{13}$$

In particular, the right side of (13) must be positive definite. Since, in the long-wavelength limit, $S^{-1}(k) \sim k^{-1}$ and $\epsilon_0 \sim k^2$, we see that

$$\lim_{k\to 0} (\epsilon_0^{1/2}S^{-1}\epsilon_0^{1/2})^2 = \lim_{k\to 0} 2\epsilon_0^{1/2}X'\epsilon_0^{1/2} \ . \tag{14}$$

Thus the right side of Eq. (14) must be positive, i.e., must have all positive eigenvalues, and is proportional to k^2. Moreover, it follows that the determinant of the right side must be positive and proportional to k^{2n}. Since $\mathrm{Det}\epsilon_0$ is also positive and proportional to k^{2n}, it follows that

$$\lim_{k\to 0} \mathrm{Det}X'(k) > 0 \ . \tag{15}$$

The actual value of the left side of (15) may be determined from Eq. (14) by using the invariance of the determinant under permutation of the factors in a product matrix, together with the relationship in Eq. (2), to give

$$\lim_{k\to 0} \mathrm{Det}X'(k) = \prod_{\alpha=1}^{n} m_\alpha c_\alpha^2 \ , \tag{16}$$

which is the generalization of the one-component result[17]

$$\lim_{k\to 0} X'(k) = mc^2 = \frac{1}{\rho K} \ . \tag{17}$$

Thus it is clear that the positive value in (15) is directly related to the absolute mechanical stability of the system.

In the particular case of a two-component system, the result is

$$\mathrm{Det}X'(0^+) = m_1 m_2 c_+^2 c_-^2 \ . \tag{18}$$

The velocities of the density fluctuation and concentration fluctuation are evaluated elsewhere.[16] The result for the product which appears on the right of Eq. (18) is

$$\mathrm{Det}X'(0^+) = \rho_1\rho_2(e_{11}e_{22} - e_{12}^2) \ , \tag{19}$$

where $e_{\alpha\beta}$ is the second derivative of the ground-state energy density with respect to the number densities ρ_γ. The right side of Eq. (19) is related to more familiar forms of the thermodynamic stability[1,2] by noting that[16]

$$e_{11}e_{22} - e_{12}^2 = e_{22}\left.\frac{\partial\mu_i}{\partial\rho_1}\right|_{\mu_2} = e_{11}\left.\frac{\partial\mu_2}{\partial\rho_2}\right|_{\mu_1} \ , \tag{20}$$

so that the vanishing of Eq. (16) with $e_{\alpha\beta} \neq 0$ implies that $\partial\mu_\alpha/\partial\rho_\alpha$ vanishes.

The location of the spinodal points is given by the vanishing of $\mathrm{Det}X'(0^+)$ in which case

$$e_{11}e_{22} - e_{12}^2 = 0 \ , \tag{21}$$

or, equivalently, either c_+ or c_- vanishes. In fact, it can be seen from the expressions in Ref. 16 that $c_-^2 = 0$ whenever (21) is satisfied, although c_+^2 vanishes only at the pont where $e_{11} = e_{12} = e_{12} = 0$. If $c_-^2 = 0$ but $c_+^2 > 0$, the concentration mode has completely softened at $k = 0$, so that the spinodal point corresponds to an absolute mechanical instability with respect to phase separation. When c_+ also vanishes, the density fluctuation mode has also completely softened, which corresponds to the cavitation instability found at the maximum metastable negative pressure in a one-component system.[14]

Expressions for the long-wavelength behavior of $S_{\alpha\beta}(k)$ at $T = 0$ (Ref. 17) show that the slope diverges $(e_{11}e_{22} - e_{12}^2)^{-1/2}$ as the spinodal point is approached from within the metastable region. However, if the spinodal point is the phase-separation instability so that c_+ does not vanish, there is a linear combination of $S_{\alpha\beta}(k)$ which has a nondiverging long-wavelength slope, corresponding to the normalization of the density fluctuation eigenstate.

At $T \neq 0$ the components of the liquid structure factor have a finite value at $k = 0$, proportional to T with a coefficient which diverges as $(c_-)^{-2}$ as $c_- \to 0$. This is the concentration fluctuation effect analogous to critical opalescence at a liquid-gas critical point.

III. APPLICATIONS

An advantage of the formulation of the stability criterion in terms of $X'(0)$ is that $X'(k)$ is the key function in certain methods for solving the ground-state problem. As a practical matter, one does not actually know ψ_0 as was presupposed in Sec. II. However, experience has shown that Jastrow functions [i.e., functions of the form of Eq. (6) with ψ_0 being a constant for boson systems or a Slater determinant for fermion systems] provide good approximations to ground-state wave functions of strongly correlated systems. Moreover, the long-wavelength properties of such systems are accounted for very well by a Jastrow function if that function is the solution of the Euler-Lagrange equation for the energy extremum[17–20]:

$$\frac{\delta}{\delta u(r)}\langle\psi_u|H|\psi_u\rangle/\langle\psi_u|\psi_u\rangle=0 \ . \tag{22}$$

Thus the appearance of long-wavelength instabilities, when the wave function is the optimum Jastrow function, may be taken as a strong indication of the same instabilities in the exact ground state of the system.

Alternative procedures have been developed to solve the Euler-Lagrange equation for $u(r)$ [Eq. (22), (8), or (13)] which makes use of the function $X'(k)$, in effect attempting to reduce the difference between the left and right sides of Eqs. (8) and (13) with successive iteration. One of these procedures, the paired-phonon analysis (PPA),[17,18] has been generalized to multicomponent systems.[8] The PPA is initialized with a convenient choice of Jastrow function $u^0_{\alpha\beta}(r)$. The first iterate is defined by $u^1_{\alpha\beta}(r)=u^0_{\alpha\beta}(r)+\Delta u_{\alpha\beta}(r)$, where $\Delta u_{\alpha\beta}(r)$ is chosen to minimize the expectation value of H, in an approximate way, which is equivalent to diagonalizing the Hamiltonian in the paired-phonon basis via a Bogoliubov transformation.[8,17] The matrix elements of H in this basis depend in a simple way on $X'(k)$. Thus the iteration scheme is closely tied to a physical basis, the diagonalization procedure. During the PPA iteration process, the condition that the diagonalization produces real excitation energies at small k is that $\Omega(k)$ [Eq. (13)] be a positive matrix [even though it is no longer equal to the left side of Eq. (13) unless the PPA has been iterated to convergence]. Moreover, upon taking the determinant of $\Omega(k)$, this condition reduces to the absolute stability condition of Eq. (15) in the long-wavelength limit. The relationship between this determinant and the excitation velocities [Eq. (16)] also holds for the optimized Jastrow function as long as the c_α are the velocities which appear in the slopes of excitation spectrum and $S(k)$ at small k.[21]

To illustrate the points discussed above we have done several calculations on two-component boson mixtures in the HNC/0 (hypernetted-chain) approximation as in Ref. 18. We use the Lennard-Jones potential for all interactions:

$$V_{\alpha\beta}(r)=4\epsilon_{\alpha\beta}\left[\left(\frac{\sigma_{\alpha\beta}}{r}\right)^{12}-\left(\frac{\sigma_{\alpha\beta}}{r}\right)^{6}\right] \ . \tag{23}$$

We follow Miller by parametrizing the potential with an excess ϵ parameter e (Ref. 1):

$$e\equiv\epsilon_{12}-\tfrac{1}{2}(\epsilon_{11}+\epsilon_{22}) \ . \tag{24}$$

For simplicity we choose $\epsilon_{11}=\epsilon_{22}\equiv\epsilon$ and define the enhancement parameter κ by

$$\epsilon_{12}=(1+\kappa)\epsilon \ . \tag{25}$$

Then e reduces to

$$e=\epsilon\kappa \ , \tag{26}$$

so that κ is a unitless scale for e allowing us to vary the relative strength of the interspecies interaction.

Results for the finite k-dependent stability condition are shown in Fig. 1 for the final solutions of the iterative procedure. The system under consideration is a 6% concentration of *boson* ^{3}He in ^{4}He at a density of 0.0218 Å^{-3} with helium parameters in the Lennard-Jones potential, except that the ϵ_{12} enhancement parameter κ is in the range 0.050 down to 0.015. Note that the finite k-dependent stability condition [$\mathrm{Det}\,\Omega(k)>0$] is violated for κ less than approximately 0.0215. Note, also, that it is first violated at $k=0$, as should be expected for an infinite wavelength instability. The absolute stabililty condition [$\mathrm{Det}\,X'(0)>0$] is examined as a function of κ in Fig. 2. The fact that it appears to be a linear func-

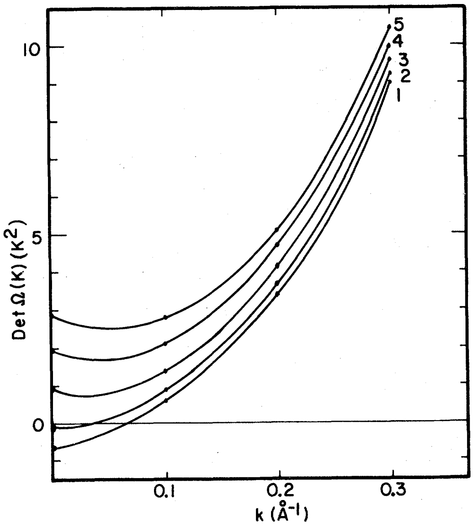

FIG. 1. Det $\Omega(k)$ [Eq. (13)] for a 6% concentration of *boson* ^{3}He in ^{4}He at total denstiy $\rho=0.0219$ Å^{-3} for several values of κ: 1, $\kappa=0.015$; 2, $\kappa=0.02$; 3, $\kappa=0.03$; 4, $\kappa=0.04$; 5, $\kappa=0.05$.

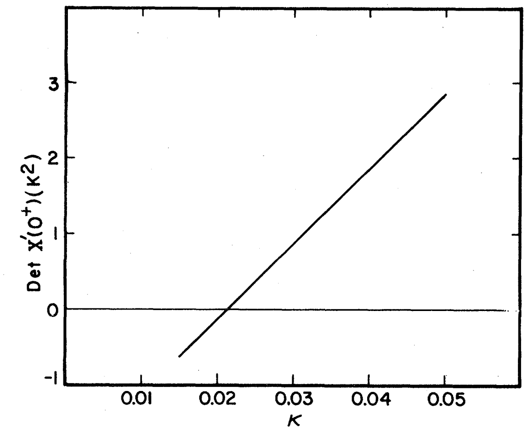

FIG. 2. Det $X'(0^+)$ as a function of κ for the system described in Fig. 1.

tion of κ makes it easy to obtain the critical κ value for this density and concentration without being obliged to approach it arbitrarily closely.

Figure 3 shows the $T=0$ $S_{\alpha\beta}(k)$ for several values of κ. Note that the closer κ approaches the instability, the larger the slope of $S_{\alpha\beta}(k)$ at small k, approaching an infinite slope at the instability. Note also that the variation of $S_{\alpha\beta}(k)$ with κ shows up only at very small k, owing presumably to the rather small values of κ tested.

Included in Figs. 1 and 3 are apparent solutions of the Euler-Lagrange equation from within the absolutely unstable values of κ. It should be stressed that these are not solutions in the strict sense of the variational problem, but are an artifical effect due to the choice of the step size in k space in our numerical procedure. There are, in fact no solutions for these values of κ. Moreover, the analysis of Sec. II makes it clear that, if this situation persists in the exact ground-state solution, then the spinodal value of κ is the limiting point for the existence of a uniform ground-state wave function. Beyond this spinodal point, the ground state must be a state of two-phase equilibrium. It should be added, however, that there exists a well-defined energy function $E(\kappa)$ for κ beyond the spinodal point. In this unstable region, $E(\kappa)$ is the infimum of the quantity $\langle\psi|H|\psi\rangle/\langle\psi|\psi\rangle$ within the space of all uniform trial functions ψ, and can be approached arbitrarily closely from the above by suppressing the long-wavelength fluctuations which arise in the solution of the corresponding Euler-Lagrange equation.

Our stability results are in semiquantitative agreement with those of Miller where they can be compared,[1] though our method is very different. His results are obtained by evaluating the curvature of the enthalpy. Since he uses short-ranged Jastrow functions, the instabilities cannot show up in long-wavelength behavior of the $S_{\alpha\beta}(k)$, which in that

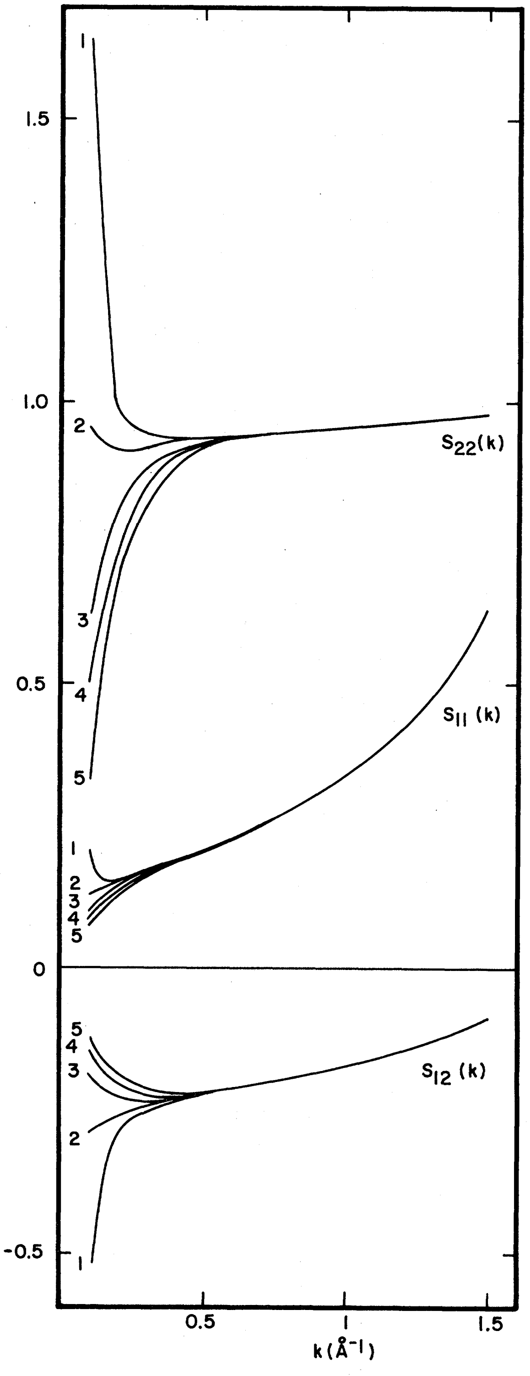

FIG. 3. Theoretical optimal structure functions $S_{\alpha\beta}(k)$ for a 6% concentration of *boson* ^{3}He in ^{4}He at total denstiy $\rho=0.0219$ Å^{-3} for the values of κ given in Fig. 1.

case approach a finite value at $k=0$. However, the absence of the correct long-wavelength behavior only slightly affects the energy, the essential ingredient in the excess enthalpy.

Isotopic mixtures correspond to the choice $\kappa=0$, all constitutents having the same interactions. As must be the case from more general arguments,[6] we find no solutions in the isotopic boson mixture ^{3}He bosons in ^{4}He (as can be seen from Figs. 1 and 2) as well as in the case of spin-aligned hydrogen in spin-aligned tritium (^{1}H-^{3}H). The absolute stability condition Eq. (15) is strongly violated at all concentrations. It is interesting to note that it takes somewhat more than a 2% enhancement of the attraction between ^{3}He (boson) and ^{4}He atoms to overcome the tendency to phase separate in a 6% solution.

It should be emphasized that in those cases where the absolute stability condition is not violated, i.e., where we find a solution to the Euler-Lagrange equations, we cannot decide without calculating the excess enthalpy whether the solution is a metastable system lying in the two-phase region, or is stable.

Since our calculations have dealt exclusively with boson mixtures, we should point out that the same analysis with only slight modifications[20] can be applied to mixtures having one or more fermion component, which includes a number of physically interesting examples. An interesting complication occurs there because of Landau damping of one or more of the collective modes, which should be enhanced by the softening of the collective mode as the instability is approached. Analysis of the one-component fermion liquid has, however, demonstrated the existence of absolute instabilities at negative pressures,[9] a result which will have a straightforward generalization to a multicomponent system.

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[1] M. D. Miller, Ann. Phys. 127, 367 (1980).

[2] M. D. Miller, Phys. Rev. B 18, 4730 (1978).

[3] R. A. Guyer and M. D. Miller, Phys. Rev. B 22, 142 (1980).

[4] J. Phys. (Paris) 41, C7-1 (1980).

[5] R. A. Guyer and M. D. Miller, Phys. Rev. Lett. 42, 1754 (1979).

[6] G. V. Chester, Phys. Rev. 100, 446 (1955).

[7] K. E. Kürten and M. L. Ristig (unpublished).

[8] C. E. Campbell, Ann. Phys. 73, 43 (1977).

[9] J. W. Owen, Phys. Rev. B 23, 2169 (1980); J. W. Owen, in *Recent Progress in Many-Body Theories,* Lecture Notes in Physics, Vol. 142, edited by J. G. Zabolitzky, M. de Llano, M. Fortes, and J. W. Clark (Springer-Verlag, New York, 1981).

[10] L. J. Lantto and P. J. Siemens, Phys. Lett. 68B, 308 (1977).

[11] H. W. Jackson and E. Feenberg, Ann. Phys. 15, 266 (1961).

[12] E. Feenberg, *Theory of Quantum Fluids* (Academic, New York, 1969).

[13] C. E. Campbell and F. J. Pinski, Nucl. Phys. A328, 210 (1979).

[14] A. D. Jackson, A. Lande, and L. J. Lantto, Nucl. Phys. A317, 1 (1979).

[15] R. P. Feynmann, Phys. Rev. 94, 262 (1954).

[16] C. E. Campbell, Low Temp. Phys. 4, 433 (1971).

[17] C. E. Campbell and E. Feenberg, Phys. Rev. 188, 396 (1969).

[18] C. C. Chang and C. E. Campbell, Phys. Rev. B 15, 4238 (1977).

[19] E. Krotscheck, R. A. Smith, and J. W. Clark, in *Recent Progress in Many-Body Theories,* Lecture Notes in Physics, Vol. 142, edited by R. Guardiola and J. Ros (Springer-Verlag, New York, 1981).

[20] E. Krotscheck, Phys. Rev. A 15, 397 (1977); E. Krotscheck and W. Kundt, Phys. Lett. 71B, 19 (1977).

[21] However, it has been demonstrated in the one-component fluid that these velocities may no longer correspond to the thermodynamic derivatives, so that Eq. (19) may no longer be valid (Refs. 21 and 14).