

Behavior of the thermal conductivity of dilute ^3He - ^4He mixtures in the superfluid phase

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We present a new solution to the Khalatnikov two-fluid hydrodynamic equations which suggests that thermal conduction experiments in superfluid ^3He - ^4He mixtures measure the diffusive conductivity. This implies that for small ^3He concentrations the measured conductivity approaches a constant. The singular behavior predicted by Khalatnikov's original solution can be eliminated using a constraint imposed by the tight coupling of concentration and temperature gradients. Microscopically, we find that diffusive mass transport in bulk superfluid is limited by the ability of the diffusing atoms to get rid of the thermodynamic diffusion energy they carry, leading to an apparent diffusivity which can be much smaller than the isothermal mass diffusivity. We also describe a preliminary experiment to observe two effects predicted by our analysis. In agreement with our predictions, we find that (1) the thermal relaxation time in dilute mixtures is very long and (2) the temperature gradient in a superfluid mixture is caused by ^3He mass diffusion. The latter observation gives rise to a new technique for measuring the effective thermal conductivity which avoids the main difficulties encountered by the previous method.

I. INTRODUCTION

The effective thermal conductivity K_{eff} of very dilute superfluid ^3He - ^4He mixtures is predicted^{1,2} to have a $1/c$ divergence when c , the concentration of the ^3He , approaches zero. However the measurements of Dingus, Zhong, Tuttle, and Meyer³ (DZTM), show a major deviation from this behavior. Some indications of this discrepancy were first reported by Tanaka *et al.*,⁴ although Ptukha's original work⁵ showed no anomaly in this region. Recently, using a new method of measuring K_{eff} not involving heat flow, we have verified⁶ the results of DZTM, at least near $c = 10^{-7}$ where the discrepancy with theory is large. The difficulty appears to be of subtle nature, since Khalatnikov's two-fluid hydrodynamics² (KTFH), used in the analysis, is very general and based on well-established conservation laws. Also, above the lambda transition the theory reduces to that of normal dissipative fluid mixtures, as is to be expected. Thermohydrodynamic analysis can be applied at various levels of approximation. Here we are concerned with hydrodynamic and diffusive flows of heat and mass which can be slowly varying in time, and neglect the effects of acceleration, viscosity, and boundary layers. In this approximation Khalatnikov has shown² that the effective conductivity is given by the expression

$$K_{\text{eff}} = K + \rho DR (k_T - k_T^*)^2 / c M_3, \quad (1)$$

where D is the isothermal mass diffusivity, K is the diffusive thermal conductivity, ρ is the density, R is the gas constant, k_T is the thermal diffusion ratio, k_T^* its effective value in the superfluid phase, and M_3 is the molar mass of ^3He . Khalatnikov has argued² that K , D , and

k_T^* are finite while $k_T \rightarrow 0$ as $c \rightarrow 0$, leading to the prediction $\lim_{c \rightarrow 0} (K_{\text{eff}}) \sim 1/c$. In Sec. II we rederive the results of KTFH in the limit that ∇c and ∇T are coupled and show that k_T is not an independent parameter but is actually determined by a constraint which has two solutions. It is immediately obvious that one solution explains the DZTM result, while the other leads to the prediction in conflict with it. The former solution predicts several new results, which we compare with existing data. Two of the predictions require new experimental tests. In Sec. III we describe an experiment that was performed to obtain the necessary data. The experiment also gives values of K_{eff} in reasonable agreement with those of DZTM near $c = 10^{-7}$. Our conclusions are summarized in Sec. IV. A preliminary report of some of this work has been published elsewhere.⁶

II. THEORY

This section is divided into three parts. In Sec. A, we solve KTFH for mixtures of any concentration and show that the theory is incomplete. As a result, the variable c and the normal fluid velocity v_n cannot satisfy all the conditions imposed. We propose a way to complete the theory. In Sec. B, we apply our analysis to the case of the dilute mixtures and show how the $1/c$ divergence of K_{eff} can be eliminated. In Sec. C, we discuss the case of pure ^4He .

A. Mixtures of any concentration

We begin by writing down the relevant equations⁷ from KTFH,

$$i = -\rho D(\nabla c + k_T \nabla T / T), \quad (2)$$

$$q = [k_T(\partial\mu/\partial c)_T - T(\partial\mu/\partial T)_c + \mu]i - K \nabla T, \quad (3)$$

$$Q = q + \rho S T v_n + \mu \rho c v_n, \quad (4)$$

$$\nabla J = -\rho \partial c / \partial t, \quad (5)$$

$$J = i + \rho c v_n, \quad (6)$$

$$\partial v_s / \partial t + \nabla(v_s^2 / 2 + \mu_4) = 0, \quad (7)$$

$$\delta \Pi_{ik} / \delta r_k = 0; \quad \Pi_{ik} = \rho_s v_{si} v_{sk} + \rho_n v_{ni} v_{nk}. \quad (8)$$

Here i and q are the diffusive mass and heat current densities, $\mu = \mu_3 - \mu_4$ is the chemical potential difference between ^3He and ^4He , Q is the applied heat current density, J is the total ^3He mass current density, ρ_n is the normal fluid density including both ^3He and ^4He components, ρ_s is the superfluid density, v_s is the superfluid velocity, and Π_{ik} is the momentum tensor. In addition, Khalatnikov showed that in steady state, $\nabla\mu_4 = 0$, leading to a coupling of the concentration gradient and the temperature gradient. This leads to the result^{2,8}

$$k_T^* \equiv -T \nabla c / \nabla T = T[(\partial\mu/\partial T)_c + S/c] / (\partial\mu/\partial c)_T, \quad (9)$$

where S is the entropy per gram. This relation can be interpreted as a balance between the osmotic pressure and the fountain pressure. The normal fluid therefore does not experience any net force, and does not accelerate. In general, for a time-dependent state, the coupling need not be maintained. If it is broken, then $\nabla\mu_4 = -c(\partial\mu/\partial c)_T[k_T^* \nabla T / T + \nabla c]$, in which we have neglected the pressure effect.² The evolution of v_s is then determined by Eq. (7). For small $\nabla\mu_4$, Eq. (7) reduces to a wave equation, which describes second sound modes.² This oscillatory state decays to a state of uniform μ_4 on the time scale of second sound damping τ_2 . If τ_2 is short compared to the time scale associated with the relaxation of ^3He concentration τ , then the coupling approximation is valid for the relaxation process. Otherwise, the oscillatory state would persist and decay slowly to the final state of a uniform mixture. DZTM get $\tau \sim 10^3$ sec which is many orders of magnitude greater than τ_2 for a sample of similar geometry.⁹ Therefore coupling is an extremely good approximation for the time scale of interest here.

To establish the central point of our argument, we now consider the fully coupled limit and explore some of the hydrodynamic implications. Coupling can be used to eliminate the variable ∇T from the above equations and to express all the unknowns, such as i , q , J , and v_n , in terms of ∇c . From Eq. (4), we obtain

$$v_n = E / G \nabla c + Q / \rho S T G, \quad (10)$$

where

$$E = P D (1 - k_T / k_T^*) / S T - K / \rho S k_T^*, \quad (11a)$$

$$P = k_T (\partial\mu/\partial c)_T - T (\partial\mu/\partial T)_c + \mu, \quad (11b)$$

and

$$G = 1 + \mu c / S T. \quad (11c)$$

We now apply the above analysis to the special case of one-dimensional flow in the absence of any applied heat flux ($Q = 0$). If Eqs. (2)–(9) are internally consistent, they should be applicable to any relevant situations, which certainly cover the case discussed here. Therefore if a logical solution for this special case does not exist, then an inconsistency exists.

We solve for J in terms of ∇c , substitute J into Eq. (5), and find that in the limit of small ∇c , Eq. (5) reduces to the diffusion equation

$$\partial c / \partial t = D_{\text{eff}} \nabla^2 c, \quad (12)$$

where $D_{\text{eff}} = D(1 - k_T / k_T^*) - cE / G$. For a particular experimental setup, the quantity $c(\mathbf{r}, t)$ can be obtained by solving Eq. (12) with the appropriate boundary conditions. Then $v_n(\mathbf{r}, t)$ can be calculated simply by substituting $c(\mathbf{r}, t)$ into Eq. (10). However, v_n obtained this way must also satisfy the equation of motion and the conservation of momentum [Eqs. (7) and (8)]. When $\nabla\mu_4 = 0$, Eqs. (7) and (8) can be used to show that for one-dimensional flow along the z axis (see the Appendix),

$$\partial v_n / \partial z = 0. \quad (13)$$

Substituting Eq. (10) into Eq. (13), we obtain $(E/G) \partial^2 c / \partial z^2 = 0$. It can be seen from Eq. (11a) that in KTFH the quantity E has a value controlled by the three free parameters D , k_T , and K which are determined by microscopic arguments. Since from Eq. (11c), G is, in general, finite, KTFH would force $\partial^2 c / \partial z^2$ to be zero, in contradiction with the requirements of Eq. (12). Thus it is clear that we cannot simultaneously accept a true coupling of ∇c and ∇T and maintain the independence of D , k_T , and K . Our proposal to escape from this dilemma is to suggest that, at least for superfluid mixtures, we must have $E = 0$. We then obtain the following relationship between the three formerly independent parameters:

$$D(1 - k_T / k_T^*) = T K / \rho k_T^* P. \quad (14)$$

Clearly this new relation is a direct result of coupling, since it reduces the number of degrees of freedom of the system. Formerly, the value of k_T could only be deduced from microscopic arguments. Now, with the help of Eq. (14) and measurements of D and K , it is possible to determine k_T from experiment even in a superfluid.

The constraint imposed by Eq. (14) changes the picture of KTFH significantly. To explore the changes we list the results of KTFH after we have eliminated the variable ∇T . We obtain

$$i = -\rho D_a \nabla c, \quad (15)$$

where

$$D_a = D(1 - k_T / k_T^*), \quad (16)$$

and

$$q = -\rho S T E \nabla c, \quad (17)$$

$$J = \rho(cE / G - D_a) \nabla c + cQ / S T G. \quad (18)$$

The quantity D_a can be interpreted as the apparent

diffusivity of a coupled mixture. Using the constraint $E=0$, we obtain

$$q=0, \quad (19)$$

$$v_n = Q/\rho STG, \quad (20)$$

$$J = -\rho D_a \nabla c + cQ/STG, \quad (21)$$

$$D_{\text{eff}} = D_a = D(1 - k_T/k_T^*). \quad (22)$$

These results imply that all the heat generated by a heater in a superfluid mixture is carried away by counterflow, just as in the case of a pure superfluid, in contrast to the previous result in Eq. (4). One interesting question is whether there is a finite counterflow in a mixture when the heater in a thermal conductivity cell is off. From Eq. (20) it can be seen that after coupling is established, there is no counterflow even when the mixture is not totally uniform. This is contrary to the previous result of KTFH in which v_n is proportional to ∇c . Equation (22) shows that the effective diffusivity and the apparent diffusivity become the same quantity in the coupled limit.

We can now easily obtain an expression for K_{eff} valid for any concentration from the steady-state condition, $J=0$, and Eqs. (9), (14), (21), and (22),

$$K_{\text{eff}} = (STG/cP)K. \quad (23)$$

This result shows that K_{eff} is proportional to K , which, in general, differs from Khalatnikov's prediction given by Eq. (1).

We note that the results given by Eqs. (14)–(23) can be obtained from energy considerations. Since no mechanism exists in the bulk of the fluid to convert diffusive flow energy into hydrodynamic flow energy, these must be conserved independently. Conservation of hydrodynamic flow energy implies that for one-dimensional flow v_n is not a function of position, as indicated by Eq. (13). In addition, the independent conservation of diffusive flow energies implies that ∇T in the bulk is the result of a balance of diffusive flow energies alone. The diffusive energy carried by the ^3He diffusion current cannot be converted into hydrodynamic energy. In order for the ^3He to diffuse, energy conservation forces the creation of a ∇T in a superfluid. A heat current density given by $K\nabla T$ is then carried by elementary excitations (rotons, photons, etc.), which balances the diffusive energy carried by ^3He mass diffusion given by Pi . We obtain the general relation $i = K\nabla T/P$ which explicitly shows that ∇T in a superfluid is caused by ^3He diffusion. Substituting this into Eq. (3), we obtain $q=0$, in agreement with Eq. (19). We have already shown that ∇T must be coupled to ∇c ; otherwise the normal fluid would accelerate. Eliminating ∇T in favor of ∇c , we obtain $i = -\rho D_a \nabla c$, where $D_a = TK/\rho P k_T^*$, in agreement with Eqs. (14), (15), and (16). Thus the apparent (or effective) mass diffusion in superfluid mixtures is a process in which ^3He atoms must get rid of the energy they carry, through thermal diffusion in the bulk, in order to diffuse. Since thermal diffusion acts as an additional bottleneck which limits ^3He mass diffusion, we have a general result $D_a < D$.

The relation between i and ∇T can also be obtained by solving Eqs. (9), (14), (15), and (16). Then, using Eq. (23), we rewrite it in terms of measurable quantities

$$i = (cK_{\text{eff}}/STG)\nabla T. \quad (24)$$

This expression is not affected by variations of v_n and Q .

We now consider relaxation phenomena in the coupled limit. We solve the diffusion equation [Eq. (12)] for one-dimensional flow and the boundary conditions $c(0,t)=c_0$ and $J(d,t)=0$, appropriate for a thermal conductivity cell with a large isothermal top tank similar to that shown in Fig. 4. The slowest decay mode when $Q=0$ is

$$c(z,t) = c_0 - A \sin(\pi z/2d) \exp(-t/\tau). \quad (25)$$

Here z is the distance from the top plate, d is the distance between the end plates of the thermal conductivity cell, c_0 is the concentration in the top tank, A is a constant determined by the initial conditions. The relaxation time is

$$\tau = 4d^2/\pi^2 D_{\text{eff}}. \quad (26)$$

Using Eqs. (14), (22), and (23) we obtain

$$\tau = (\rho SG k_T^*/cK_{\text{eff}})(2d/\pi)^2. \quad (27)$$

For a thermal conductivity cell with no top tank, similar to the one used by DZTM, the appropriate boundary conditions are $J(0,t)=0$ and $J(d,t)=0$. In this case, the slowest decay mode when $Q=0$ is

$$c(z,t) = c_0 + A \cos(\pi z/d) \exp(-t/\tau'), \quad (28)$$

where

$$\tau' = \tau/4. \quad (29)$$

When Q is not zero, Eq. (12) is replaced by $\partial c/\partial t = D_{\text{eff}}\nabla^2 c - (Q/\rho STG)\nabla c$, which is not purely diffusive, but relaxes exponentially, and can certainly be solved by interested readers.

Based on the above analysis, we obtain the following picture of the relaxation phenomena studied by DZTM. In a thermal conductivity measurement, when a heater is on, a constant v_n given by Eq. (20) flushes the ^3He atoms, which causes a depletion of ^3He at one end of the cell and a collection of ^3He at the other, creating a ∇c . A mass diffusion current given by Eq. (15) is then generated as a result of the ∇c . This mass diffusion current carries energy, which cannot be converted to hydrodynamic energy due to the absence of a microscopic conversion mechanism. Thus a ∇T is formed in a superfluid, and the energy carried by the mass diffusion current is returned by thermal diffusion. Counterflow transports away all the heat generated by the heater. At any instant in time if Q and v_n are changed abruptly, ∇c , ∇T , and i would not follow the abrupt change, but relax slowly to their new steady-state values.

Returning to the difficulties of treating D , K , and k_T as independent parameters, we substitute Eq. (28) into Eq. (10) and obtain

$$v_n = -(AE\pi/dG)\sin(\pi z/d) \exp(-t/\tau).$$

This unusual functional form for a hydrodynamic flow violates the conservation of momentum and the equation of motion, which requires v_n to be constant throughout the cell. Since A is determined by the initial conditions of the decay of concentration gradient, it is nonzero in general, and $E=0$ again appears to be the only way out of the problem.

Above the lambda transition temperature T_λ Behringer and Meyer¹⁰ have observed a gradual onset of coupling. As T_λ is approached, the concentration and the thermal relaxation times become almost identical. They have divided the transition region near T_λ into four regions: the complete-uncoupling, the weak-coupling, the intermediate-coupling, and the strong-coupling regions. Our analysis can be extended to cover the strong-coupling case above T_λ by simply setting v_n , v_s , and ρ_s to zero in Eqs. (4), (6), (7), and (8). The result in Eq. (14) remains unchanged. Since K , D , and k_T are all measurable quantities above T_λ , an opportunity exists to test the validity of Eq. (14). To do so, we define a quantity $F=[TK/\rho PDk_T^*+k_T/k_T^*]^{-1}$, where k_T^* for temperatures above T_λ is defined as its value just below T_λ . We rewrite Eq. (14) as $F=1$. Therefore F can be viewed as an indicator of the degree of coupling. If $F=1$, the system is perfectly coupled. This situation applies only to the superfluid case, where v_n is finite and $q=0$. Equations (3) and (4) then imply that ∇T does not depend on q or on Q . Above T_λ , $v_n=0$, and q is the applied heat flux. Thus there is always a component of ∇T given by $-q/K$ and, strictly speaking, the system is not truly coupled. However when the applied heat flux is small, many of the characteristics of coupling are observed¹⁰ near T_λ . Since our analysis indicates that $F=1$ corresponds to the perfectly coupled case, we expect values of F far from unity to apply to the noncoupled case, and values F close to unity to apply to the strong-coupling case. The physical meaning of F will become more apparent if one considers an experiment in which ∇c is kept constant, the external heat flux is turned off, and ∇T is measured as a function of $T-T_\lambda$. Using Eqs. (2) and (3), we find that above T_λ , $\nabla T = -(K/\rho PD + k_T/T)^{-1} \nabla c$. Intuitively, ∇T should have a small value far away from T_λ and rise to a value given by the coupling relation as T_λ is approached. One can easily show that F is just the fraction of the coupled ∇T reached. In this context, it is obvious that $F \rightarrow 1$ as coupling sets in.

To evaluate F , we first use the results of Sreedhar and Daunt, Lounasmaa, and Weinstock *et al.*¹¹ to determine $\mu(c, T)$. Since the zero-point energies of both isotopes are not well known, we have assumed that their difference is zero. From $\mu(c, T)$ we compute the values of P . Then we calculate F with no adjustable parameters, using the data for D , k_T , and K , from Gestrich *et al.*⁸ Figure 1 shows the dependence of F on the reduced temperature $\varepsilon = |1 - T/T_\lambda|$ for a mixture of $c=0.04$. Our analysis predicts that $F=1$ when the system is perfectly coupled. A value of 0.7 ± 0.2 is approached by this mixture near the transition. Considering that the mixture may not be strongly coupled until it is extremely close to T_λ , and the large uncertainty in the measurements, we feel that experiment and theory are in reasonable agreement.

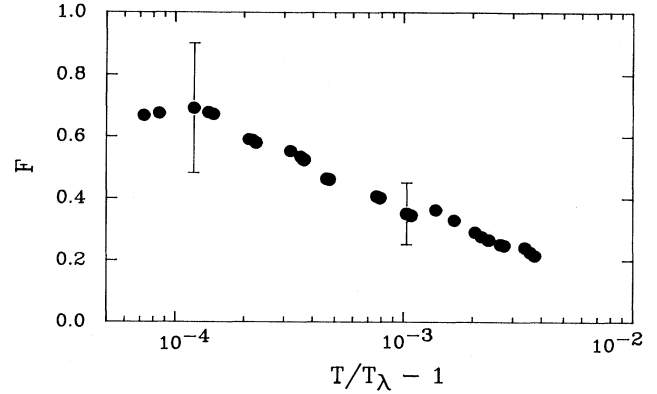


FIG. 1. A plot of the quantity $F=[TK/\rho PDk_T^*+k_T/k_T^*]^{-1}$ against the reduced temperature for a sample with $c=0.04$, using the data of Gestrich *et al* (Ref. 11).

We now turn our attention to the case of dilute mixtures where the discrepancies between KTFH and experiment are observed.

B. Dilute mixtures

In the dilute limit, the following approximations are valid:

$$\begin{aligned} G &\rightarrow 1, \\ k_T^* &\rightarrow M_3 S/R \sim 0.56, \\ \mu &\rightarrow (RT/M_3) \ln c, \\ P &\rightarrow (k_T/k_T^*)(ST/c). \end{aligned}$$

From Eq. (14) the relation between D , K , and k_T becomes

$$D(1 - k_T/k_T^*) = cK/\rho S k_T. \quad (30)$$

Substituting this to Eq. (1) we obtain

$$K_{\text{eff}} = K(k_T^*/k_T). \quad (31)$$

Equation (31) can also be obtained by using Eq. (23) and the dilute form for P .

In this limit, simple kinetic theory predicts that D approaches a constant determined by the scattering of ^3He atoms with the excitations of the host liquid.¹² Since, even in the absence of ^3He , thermal diffusive conduction can still be carried by the host excitations alone,¹¹ we also expect K to approach a constant. Whether the system contains superfluid or not, these two basic predictions of microscopic theory appear to be extremely well established. Equation (30) then implies that $k_T(1 - k_T/k_T^*)$ must approach zero in the dilute limit. Clearly, either $k_T \rightarrow 0$ or $k_T \rightarrow k_T^*$.

No explicit microscopic calculation for k_T in the low- c regime appears to exist. For an ordinary dilute mixture one can argue¹³ that as c approaches zero, both i and ∇c must approach zero, while ∇T can remain finite. Equation (2) then implies that $k_T \rightarrow 0$. In the superfluid phase this argument is not compelling since coupling requires

that ∇T must also approach zero with ∇c . Thus $k_T \rightarrow k_T^*$ is also an acceptable solution. It may be possible to decide on the correct solution for k_T based on general physical arguments, but so far we have not found a way to do this. Instead, we will compare the predictions from the two possibilities with existing experimental results. First, we can use Eqs. (14) and (23) to rewrite k_T in terms of measurable quantities

$$k_T = k_T^* (1 - cK_{\text{eff}} / \rho S D G k_T^*) . \quad (32)$$

For $c \sim 10^{-7}$, we estimate $D = 3 \times 10^{-5}$ cm²/sec from the data of Garwin and Reich,¹⁴ and $K_{\text{eff}} \sim 10^{-1}$ W/cm K from DZTM, leading to $k_T = (0.997 \pm 0.003) k_T^*$. Clearly this indicates that $k_T \rightarrow k_T^*$ is the correct solution. The largest uncertainty in this estimate is in the value of D .

Second, above T_λ it is possible to measure k_T directly. As the transition is approached, we expect the behavior of the mixture to tend towards that of the coupled superfluid case. The measurements of Gestrich *et al.*⁸ at $c = 0.04, 0.12, 0.30,$ and 0.48 all show that $k_T \sim k_T^*$ near the superfluid transition temperature, where ∇c and ∇T become coupled. Since the lambda transition is second order, we expect k_T to be roughly continuous across the transition. Thus experiment appears to indicate that the solution $k_T \rightarrow k_T^*$ is not just limited to dilute mixtures, but is a property of coupled mixtures of any concentration. This implies that the coefficient $K_{\text{eff}} / \rho S D G k_T^*$ in Eq. (32) is small. In Sec. II A we discussed the quantity $F = [TK / \rho P D k_T^* + k_T / k_T^*]^{-1}$, which approaches unity in the strong-coupling region near T_λ . Therefore the observation that $k_T \rightarrow k_T^*$ near T_λ would also imply that $TK / \rho P D k_T^*$ is small in the coupled limit. In the normal phase at a fixed temperature from T_λ , k_T must approach zero as $c \rightarrow 0$. But the measurements of Gestrich *et al.*⁸ suggest that at fixed c , $k_T \sim k_T^*$ as $\epsilon \rightarrow 0$. Thus for small c , it is necessary that k_T remains near zero until very close to T_λ . This behavior is observed in the data of Gestrich *et al.* The quantity $\lim_{c \rightarrow 0, \epsilon \rightarrow 0} k_T(c, \epsilon)$ is not well defined.

Third, it is already clear that the K_{eff} measurements of DZTM are not consistent with $k_T \rightarrow 0$, since this would predict that $K_{\text{eff}} \sim 1/c$. On the other hand if $k_T \rightarrow k_T^*$, then $K_{\text{eff}} \rightarrow K$, a constant, in approximate agreement with observation.

Fourth, the relaxation time measurements of DZTM present a serious difficulty with the $k_T \rightarrow 0$ solution. Since $\tau \sim 1/D_{\text{eff}}$ [Eq. (26)] and measurements of τ by DZTM show that, for small c , τ increases as $c \rightarrow 0$, the implication is that D_{eff} must decrease. If $k_T \rightarrow 0$, then Eqs. (22) and (30) can be used to obtain $D \sim D_{\text{eff}} [1 + cK / \rho S D k_T^* + O(c^2)]$. Thus D is a product of two terms, both of which are positive decreasing functions as $c \rightarrow 0$, implying that D must also decrease as $c \rightarrow 0$. From simple kinetic theory, the removal of impurities can only decrease the probability of collision in the dilute limit. Therefore D can only increase as $c \rightarrow 0$. On the other hand, if $k_T \rightarrow k_T^*$, then $D \sim D_{\text{eff}} [\rho S D k_T^* / cK]$. Therefore D is the product of decreasing function and an increasing function as $c \rightarrow 0$, and the relaxation data can-

not be used to determine whether D increases or decreases in this limit. But Eqs. (30) and (22) imply that $D_{\text{eff}} \rightarrow 0$. The increase of τ as $c \rightarrow 0$ is then explained by Eq. (26). Microscopically, we have already argued that the apparent (or effective) diffusion is a process in which ³He atoms must get rid of the energy they carry, through thermal diffusion in the bulk, in order to diffuse, forcing the creation of a ∇T in a superfluid. In dilute mixtures, the energy carried by ³He mass diffusion is $k_T S T i / k_T^* c$. If $k_T \rightarrow k_T^*$ then this energy diverges as c^{-1} for a fixed i . This large energy carried by ³He is the cause of the long relaxation time. The c^{-1} divergence originates from the logarithmic divergence² of μ_3 .

From the above discussion it can be seen that a number of different experimental results are in agreement with the possibility that k_T does not approach zero with c . Also, to our knowledge, there are no data in conflict with this solution. From the theoretical standpoint, the only relevant argument appears to be that of Landau and Lifshitz¹³ based on the normal fluid case, which is not compelling, for reasons stated earlier. In the remainder of this section we shall explore other implications of the new solution.

We now return to Khalatnikov's original result [Eq. (1)] for K_{eff} . This can be reduced to the simple form $K_{\text{eff}} \sim [(k_T^* / k_T) + O(c)] K \sim K + O(c)$, where the term of order c contains contributions from assuming $G \sim 1$, $k_T \sim k_T^*$, and using the dilute form of μ . A microscopic theory of the concentration dependence of K has been given by Khalatnikov and Zharkov.¹² In their analysis, K is expressed in terms of the diffusive resistivity R_4 of pure ⁴He, enhanced by a term due to scattering by the ³He, initially of order c . For $T > 1.2$ K, the phonon contribution is small; R_4 originates primarily from roton-rotor scattering.¹² We now group all the contributions of order c together to obtain $K_{\text{eff}}^{-1} = R_4 + A c + O(c^2)$ where R_4 and A are temperature-dependent parameters. Figure 2 shows a two-parameter fit to the data of DZTM

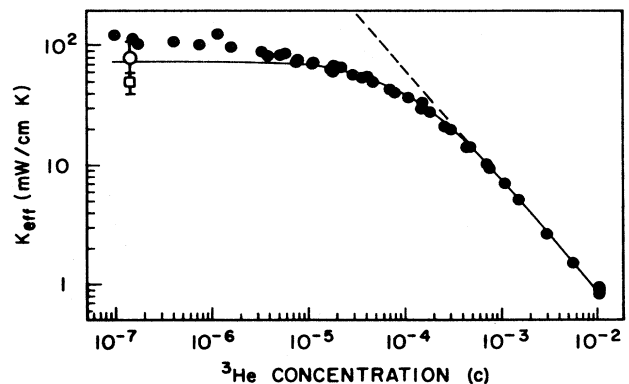


FIG. 2. Comparison of the measurements of DZTM with our results near $c = 1.5 \times 10^{-7}$. \circ , our mass diffusion current measurement; \square , our relaxation time measurement; \bullet , data of DZTM; —, the fit of our analysis to the data of DZTM for $c > 10^{-5}$ where their measurements are in the linear regime; ---, the $1/c$ divergence predicted by Khalatnikov's hydrodynamic theory.

represented by the solid line and yielding the values $R_4=0.0133$ cm K/mW and $A=115$ cm K/mW, for $\epsilon \sim 10^{-3}$. Since the data for $c < 10^{-5}$ were obtained by extrapolation to zero power, their accuracy is lower, and we excluded them from the fit. The curve obtained from the fit is shown over the entire concentration range. Excellent agreement is obtained over the range of the fit ($c > 10^{-5}$), where the data are more reliable. At lower concentrations, the data deviate slightly from the function, possibly due to residual nonlinearities in the extrapolation scheme. It is interesting to note that the curve is in good agreement with our measurements of K_{eff} at $c \sim 10^{-7}$ obtained from the experiment described in Sec. III C. These measurements have the advantage that they were performed in the linear regime without extrapolation. Improved measurements over the whole low- c region would lead to a valuable test of our prediction for $K_{\text{eff}}(c)$.

C. Pure ^4He

The above analysis seems to contradict the well-known fact that the measured thermal conductivity of pure superfluid helium is essentially infinite. To explain this, we point out that our analysis is valid only for small values of $\Delta c/c$ across a mixture. On the other hand, measurements in the nominally pure superfluid always violate this condition. In the derivation of Eqs. (2) and (3), it is assumed that the state of the mixture is only slightly perturbed from uniformity,^{2,13} requiring that

$$\nabla c \ll c/d, \quad (33)$$

where d is the cell gap. DZTM have shown that this linear regime is associated with an exceedingly small power density as $c \rightarrow 0$. Rewriting Eq. (33) we obtain $\Delta T \ll -cT/k_T^*$ and $Q \ll cK_{\text{eff}}T/dk_T^*$ for steady-state conditions under a constant applied heat flux. To stay within the linear regime at small c , it is clear that the power must be kept small and the resolution of the thermometer used in the measurement must be high enough to resolve the reduced signal. Any measurements with a finite applied power will be in the nonlinear regime when c is small enough. Also, Eq. (27) indicates that the relaxation time diverges as c^{-1} . For our thermal conductivity cell with a 2-mm gap (Fig. 4), the relaxation time was 4.5 days for $c=1.5 \times 10^{-7}$. The same cell would have a relaxation time of 1.8 yr for a sample with $c=10^{-9}$. Therefore experiments in the linear regime in a very dilute mixture are technically difficult at present. DZTM have shown that in the nonlinear regime K_{eff} increases with applied power, and Behringer¹⁵ has analyzed the system. A complete treatment within the framework of our analysis is beyond the scope of the present work. However a qualitative argument can be made for the case of a thermal conductivity cell with a large isothermal top tank. In this system an upper limit on the concentration difference between the end plate is c , when all the ^3He is flushed to the top tank. Since coupling is a force balance, it is reasonable to assume that it holds even in the nonlinear regime. We then obtain the maximum temperature difference between the end plates, $\Delta T_m = -cT/k_T^*$. For

large powers ΔT_m is a constant, leading to an extremely large apparent value for K_{eff} given by $K_{\text{eff}}=Q_a dk_T^*/cT$. This K_{eff} diverges as c is reduced to zero, but Eq. (33) is violated. It can also be seen that for normal well-grade laboratory helium with $c \sim 10^{-7}$, ΔT_m is an extremely small number, undetectable in all but the most sophisticated experiments.

We note that a finite diffusive conductivity in the absence of counterflow has been observed in pure superfluid ^4He below 0.6 K, where there is not enough normal fluid to support counterflow.¹⁶ Above 0.6 K, counterflow causes the thermal conductivity to appear infinite, but an underlying finite diffusive conductivity, of course, still exists. On the basis of our analysis, the value of R_4^{-1} obtained from the data of DZTM is the first direct measurement of this diffusive conductivity near T_λ .

Measurements of the second sound damping coefficient should also give information on the diffusive conductivity of pure ^4He . Figure 3 shows the results for K obtained from second sound damping¹⁷ and the K_{eff} data Ptukha⁵ at $c=1.04 \times 10^{-4}$. From Fig. 2 we do not expect R_4^{-1} to differ from the measurements of K_{eff} at $c \sim 10^{-4}$ by more than a factor of 2. Also measurements of DZTM indicate that the function $K_{\text{eff}}(c)$ is only weakly temperature dependent above 1.7 K. Therefore, to first order we expect Ptukha's measurement of K_{eff} to be a rough representation of R_4^{-1} . Between 0.6 and 1 K, the two sets of data in Fig. 2 fall in the same region of the graph. Above 1 K a large discrepancy exists. Very close to T_λ , K_{eff} does not diverge³ while the second sound damping constant D_2 appears to be divergent.⁹ While there is not yet enough data to allow a complete separation of K from D_2 near T_λ , a clear discrepancy already exists in the temperature range $1.2 \text{ K} < T < 2.0 \text{ K}$. This may indicate that there is yet another property of the superfluid not included in our analysis or in KTFH.

III. EXPERIMENT

Our analysis leads to four predictions: (1) the transport coefficients are constrained by Eq. (14) when ∇c and ∇T are coupled; (2) in the superfluid phase, K_{eff} ap-

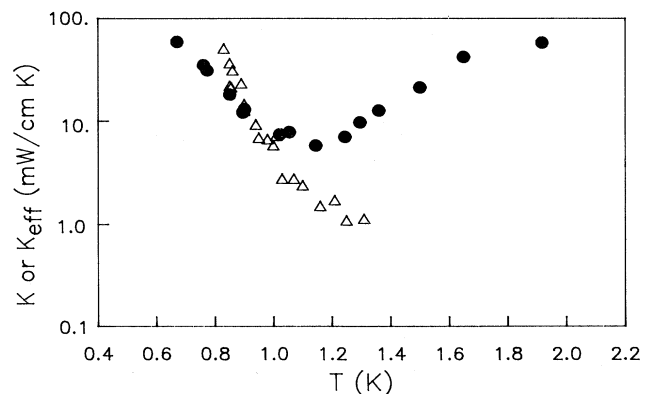


FIG. 3. Comparison of K_{eff} and K . ●, measurements of K_{eff} by Ptukha (Ref. 5) at $c=1.04 \times 10^{-4}$. Δ, measurements of K by Zinoveva (Ref. 17) using second sound damping in pure ^4He .

proaches a constant as c is reduced to zero; (3) τ approaches infinity as c is reduced to zero [Eq. (27)]; and (4) since diffusive energies are conserved independently, the temperature gradient in a superfluid mixture is caused by the diffusion of ^3He atoms, and ∇T is related to i by Eq. (24). We have already compared the first two predictions with experiment. The data of DZTM suggest that τ' may approach a constant at low c , in conflict with prediction (3). However it is clear from their work that the measurements were made in the nonlinear regime, in violation of Eq. (33). To measure τ in the linear regime and to observe the effect predicted by Eq. (24), we have performed a preliminary experiment described below.

We constructed an apparatus incorporating two very high-resolution thermometers (HRT's),¹⁸ capable of resolving temperature changes of 3×10^{-10} K. The HRT's were attached to the copper end plugs of a cylindrical sample cell with a stainless steel wall 0.012-cm thick. The end plugs were pressed into the wall section with an estimated mean gap of no more than 0.005 cm. A second helium container with a valve was connected to the first by a copper tube of 0.08 cm internal diameter as shown in Fig. 4, and the whole assembly was mounted in a vacuum can. The main and secondary chambers had cross sections of 1.2 and 2.5 cm², and vertical heights of 2 and 4 mm, respectively. Apart from the stainless-steel spacer, the apparatus was constructed from high conductivity annealed copper with large cross sections. Two samples of helium were studied: no. 1, containing a ^3He concentration¹⁹ of 1.5×10^{-7} , and no. 2, with a concentration of 1×10^{-9} . These samples almost filled the tanks, leaving a gas space of about 5% of the volume of the upper tank.

Our measurements primarily consist of observations of small temperature differences ΔT established across the

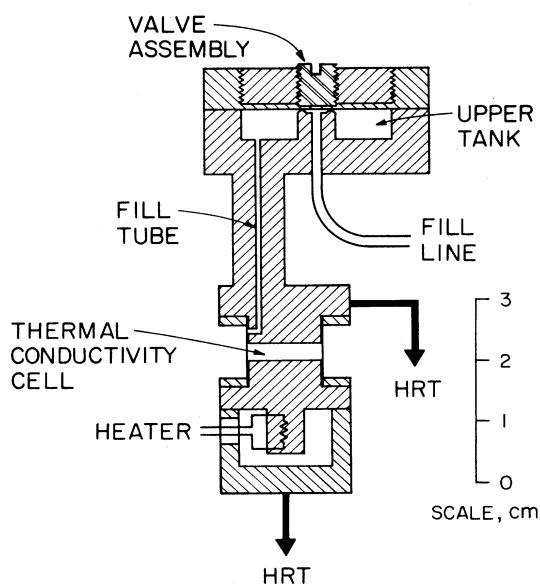


FIG. 4. Cross section of the apparatus showing the geometry of the two helium tanks and the connecting tube.

samples under various conditions near T_λ . Data were collected in two modes: continuous and discrete. In the former, the temperature of the sample was allowed to drift at a rate of about 10^{-10} deg/sec and the ΔT across the lower cell was recorded. Various power levels were applied to the lower plate as desired. In the latter, the temperature of the top plate was servocontrolled until equilibrium was established, and the resulting ΔT was recorded. The servoloop was then opened and the sample allowed to move to a new temperature. At the start of the experiment, we accurately cross calibrated the two HRT's below T_λ over a range of about 200 μdeg , neglecting the very small offset due to the background parasitic power input of 4×10^{-10} W and the Kapitza boundary resistance.

The main measurements commenced with the ^3He evenly distributed throughout the samples. Below the transition, when power levels of up to 5×10^{-9} W were applied to the bottom plate (avoiding convection), small, almost temperature-independent offsets were observed. Above the transition, much larger offsets occurred, due to the finite thermal conductivity of the fluid. Similar results were obtained for both samples, and the results for sample no. 2 with only the parasitic power input are shown by the crosses in Fig. 5(a).

On the other hand, with sample no. 1, if the ^3He was first flushed into the top tank, quite different results were obtained. Values of the temperature differences across the sample with zero applied power obtained after flushing are shown by the triangles in Fig. 5(a). Above the transition the results were unchanged, but well below, a large additional temperature differential was seen. We assume this region is the well-coupled region, where our analysis is applicable. To generate the observed offset with a heat flux, a power input to the bottom of the sample of about 10^{-7} W would be required, yet observations above the transition always gave values corresponding to the background power of 4×10^{-10} W mentioned above. In the region just below the transition temperature, the offset is attenuated, possibly due to nonlinear effects near T_λ . Outside this region the anomalous offsets are temperature independent to the accuracy of our measurements.

The temperature differential was found to decay approximately exponentially with a time constant of about 4.5 days. Measurements taken approximately 48 h after the first set are shown by the solid circles in Fig. 5(a). The relaxation time we observe is many times longer than that expected from the DZTM data, about 10^3 sec. However, the geometry of our cell is somewhat more complicated, making the interpretation of our result less direct. It is possible that the constriction between the top and the bottom tanks somehow limits the flow and significantly alters the time constant. However, we note that the entire upper assembly up to the top plate of the sample cell is constructed from high conductivity annealed copper which is estimated to have a conductivity 200 times larger than K_{eff} . Therefore, to a good approximation, we can treat the entire upper assembly as isothermal. Coupling then ensures that this assembly has a uniform concentration right up to the immediate vicinity

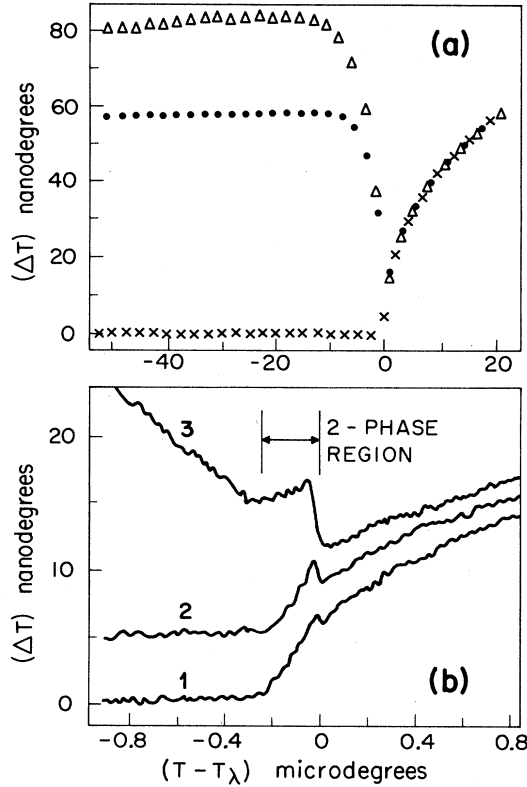


FIG. 5. Temperature differences observed across the cell near the lambda point. (a) \times , sample no. 2, $c=10^{-9}$, without heat flush; \triangle , sample no. 1, $c=1.5 \times 10^{-7}$, 137 h after heat flush; \bullet , sample no. 1, 185 h after heat flush. (b) The region very close to the transition. 1, sample no. 2, 500 h after heat flush; 2, sample no. 1, 670 h after heat flush; 3, sample no. 1, 240 h after heat flush. Curves 2 and 3 have been slightly offset vertically for clarity. The region over which helium I and II coexist in the lower tank is indicated.

of the top plate. Under isothermal conditions ^3He is transported entirely by counterflow, giving rise to a finite v_n in the constricted area between the two tanks as well as near the surface of the top plate. This is equivalent to assuming that the incoming ^3He spreads rapidly across the face of the top plate, under essentially isothermal conditions, before diffusing across the gap. We note that when the constriction in the isothermal tank is small enough, viscous drag on the normal fluid flow would affect the time constant. When this happens, coupling would no longer be a good approximation, since in this limit the hydrodynamic pressure gradient created by the drag would balance the osmotic pressure created by Δc . To be sure that we were not in this limit, we checked that the coupling relation applied by flushing all the ^3He to the top tank and measuring the initial ΔT . Our results agreed with the coupling prediction to within 30%. We then used the observed time constant and Eq. (27) to obtain $K_{\text{eff}} = 50 \pm 10$ mW/cm K, reasonably consistent with the DZTM data. This result is shown in Fig. 2.

To flush the ^3He into the top tank we applied a power of $\sim 10^{-3}$ W to the bottom plate. To verify that the flushing had occurred, we monitored the change in the relative concentration of ^3He in the two tanks by observing the relative shift in their transition temperatures with time, $\delta T_\lambda / \delta t$. Heat-capacity measurements were used to locate the transitions in each tank, allowing us to observe relative changes of c of about 3×10^{-8} . These measurements indicated that at least 80% of the ^3He in the lower tank initially was moved to the upper tank. After the heater is turned off, the ^3He begins to return to the lower tank, and in this process sets up the observed anomalous temperature gradient. A quantitative relation between ∇T and i is given by Eq. (24). The total ^3He mass current was estimated from the time dependence of the heat-capacity data. Based on our analysis, when the heater is off, $v_n = 0$, and the total ^3He mass current is purely diffusive. Under these conditions, it can easily be shown that the mass diffusion current i is related to $\delta T_\lambda / \delta t$ by the expression

$$i \sim \rho d (\delta c / \delta T_\lambda)_p (\delta T_\lambda / \delta t), \quad (34)$$

where the inverse slope of the lambda line $(\delta c / \delta T_\lambda)_p = 0.535 \text{ K}^{-1}$. Using Eqs. (24) and (34), the observed $\delta T_\lambda / \delta t$ and the ΔT well below T_λ , we compute the value $K_{\text{eff}} = 80 \pm 30$ mW/cm K for $c = 1.5 \times 10^{-7}$. This result is also shown in Fig. 2. Since the heater was off, the heat input to the sample was zero, so negligible temperature drop occurs at the boundaries of the sample due to Kapitza resistance. This circumstance greatly improves the reliability of our results for K_{eff} over previous experiments, in which the correction for the temperature drop across the boundaries was a dominant factor in the low- c regime. Figure 2 also shows that three methods of measuring K_{eff} are in reasonable agreement with the prediction based on our analysis. Also the internal consistency of the results gives additional support for the validity of our time constant measurement described above.

Near T_λ we observed two anomalies as shown in Fig. 5(b): a steplike feature observed near the temperature where the first layer of superfluid appears in the two-phase region, and a gradual rise in ΔT in the superfluid region just below the two-phase region. Comparison of curve no. 3 and curve no. 2 indicates that the gradual rise in ΔT in the superfluid phase exists only where ∇c is large. No gradual rise is observed in curve no. 2 where ∇c is small. This leads us to postulate that this feature is related to nonlinear effects near T_λ . Behringer and Meyer¹⁰ observed a strong-coupling region above T_λ . In this region, we expect ∇T to rise to the coupled value as T_λ is approached, leading us to interpret the steplike feature as the onset of coupling above T_λ . If this interpretation is correct, the location of the sharp anomaly would indicate that the samples are uncoupled until 7×10^{-8} K from T_λ , implying that the strong-coupling region above T_λ is extremely small for dilute samples.

IV. CONCLUSION

Our analysis of the hydrodynamics of superfluid mixtures indicates that Khalatnikov's original solution is in-

complete. Difficulties arise from treating D , K , and k_T as independent parameters. We have obtained a relation between them by accepting the coupling between ∇T and ∇c as an additional constraint. This relation enables us to derive a new solution which eliminates the divergent behavior of K_{eff} as a function of c , and restores agreement with the measurements. This solution implies that the quantity measured in a superfluid thermal conductivity experiment is actually the background diffusive thermal conductivity that would appear in the absence of counterflow.

We have not included critical effects due to fluctuations near T_λ , so we cannot predict the temperature dependence of the transport coefficients in this region. However, we deal with a very general aspect of the hydrodynamics, namely the reduction of one degree of freedom due to the constraint applied by coupling. It appears unlikely that the growth of fluctuation effects near the transition would affect our arguments. We can use our results together with those of Gestrich *et al.*⁸ to show that, in the superfluid phase, coupling applies for $\epsilon > 10^{-6}$, so there appears to be no reason to expect our analysis to fail in this region. Assuming there is no problem, we can then use the results of DZTM over the range $10^{-6} < c < 10^{-2}$ to show that K is constant, to within about 10%, over four decades of ϵ . For comparison, the thermal conductivity above the transition changes by a factor of about 30 over a similar range of ϵ . Also, for $c = 0.12$, measurements²⁰ of K indicate that there is no divergence at T_λ , but a continuous evolution into K_{eff} at the transition. It is important to determine whether the same behavior exists for mixtures with $c < 10^{-5}$. Our analysis of the data of DZTM suggests that for $c < 10^{-5}$ thermal conductivity measurements in dilute mixtures essentially probe a property of pure ^4He . If we take this point of view, then any measurements of K , above or below T_λ , would not be affected significantly by a very low concentration of ^3He impurity provided that the power density used is small enough. But dynamic renormalization-group theory²¹ predicts that in pure ^4He K diverges at T_λ . It is not clear how this can be reconciled with our analysis and the measurements of K_{eff} below T_λ .

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APPENDIX

In this appendix we show that $\partial v_n / \partial z = 0$, when $\nabla \mu_4 = 0$, i.e., when the temperature and concentration gradients are coupled. This result can be expected. By observing that when there is no external force acting on a fluid element ($\nabla \mu_4 = 0$), we can see that it does not ac-

celerate, which leads to a v_n that is not a function of position or time. A formal mathematical proof is presented below. The derivation applies to both static and dynamic cases with the following restrictions: (1) effects of viscosity and pressure are neglected and (2) the time scales are long compared to the damping of second sound.

For one-dimensional flow along the z axis, setting $\nabla \mu_4 = 0$, Eq. (7) becomes

$$\partial v_s / \partial t = -v_s \partial v_s / \partial z . \quad (35)$$

Using the method of separation of variables, we write

$$v_s = g(t)f(z) , \quad (36)$$

where g and f are unknown functions to be determined. Substituting Eq. (36) into Eq. (35), we obtain

$$-(1/g^2) \partial g / \partial t = \partial f / \partial z = \alpha , \quad (37)$$

where α is an integration constant independent of both z and t .

Solving Eq. (37) we obtain $f(z) = \alpha z + \beta$, $g(t) = 1/(\alpha t + \gamma)$, and

$$v_s(z, t) = (\alpha z + \beta) / (\alpha t + \gamma) , \quad (38)$$

where β and γ are integration constants independent of both z and t .

As t approaches infinity, Eq. (38) shows that v_s must approach zero. But it is commonly known that when a constant heat flux is applied, there is a constant v_s even as t approaches infinity. Since we have let the applied heat flux be a free variable in this calculation, Eq. (38) must also apply to the case of a constant heat flux. This implies that the integration constant α must be zero. Equation (38) then reduces to $v_s = \beta/\gamma$. Since both β and γ are independent of z and t , we obtain

$$\partial v_s / \partial z = 0 \quad (39)$$

and

$$\partial v_s / \partial t = 0 . \quad (40)$$

A possibility exists that α is a function of the applied heat flux Q in such a way that $\alpha(Q) = 0$, when $Q \neq 0$, and $\alpha(Q) = \alpha_0$, when $Q = 0$. Thus it seems that the appropriate functional form of $v_s(z, t)$, for the case $Q = 0$, should be given by Eq. (38). But Eq. (10) asserts that v_s must be a smooth function of Q . This would force α_0 to be zero as well.

In the problem considered here, there is no net mass flow in the laboratory frame, leading to

$$\rho_s v_s + \rho_n v_n = 0 . \quad (41)$$

Differentiating Eq. (41) and using Eq. (39), we obtain

$$v_s \partial \rho_s / \partial z + \rho_n \partial v_n / \partial z + v_n \partial \rho_n / \partial z = 0 , \quad (42)$$

For a small amplitude disturbance (when $Q = 0$) v_s, v_n and the derivatives of ρ_s, ρ_n are assumed to be small, leading to $\partial v_n / \partial z = 0$. Thus the result $\partial v_n / \partial z = 0$ can be obtained without using Eq. (8). However, the use of Eq. (8) allows the derivation to be extended to the case in

which v_s, v_n are not small, i.e., when $Q \neq 0$. This can be shown by writing Eq. (8) as

$$\partial(\rho_n v_n^2 + \rho_s v_s^2) / \partial z = 0. \quad (43)$$

Using Eq. (41) we obtain

$$\partial\{[\rho_n(\rho_s/\rho_n)^2 + \rho_s]v_s^2\} / \partial z = 0. \quad (44)$$

Using Eq. (39) we obtain

$$\partial[\rho_n(\rho_s/\rho_n)^2 + \rho_s] / \partial z = 0. \quad (45)$$

Since the total density ρ is a constant (neglecting pressure effects) and $\rho = \rho_n + \rho_s$, substituting this into Eq. (45) we obtain $\partial\rho_n / \partial z = 0$ and $\partial\rho_s / \partial z = 0$. The result $\partial v_n / \partial z = 0$ for all v_s, v_n can then be obtained by substituting into Eq. (42).

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