

³K. A. Shapiro and I. Rudnick, Phys. Rev. **137**, A1383 (1965).

⁴P. G. de Gennes, Phys. Lett. **44A**, 271 (1973).

⁵W. M. Saslow, Phys. Rev. Lett. **31**, 870 (1973).

⁶D. N. Paulson, R. T. Johnson, and J. C. Wheatley, Phys. Rev. Lett. **30**, 829 (1973), and unpublished work.

⁷T. A. Alvesalo, Yu. D. Anufriyev, H. K. Collan, O. V. Lounasmaa, and P. Wennerström, Phys. Rev. Lett. **30**, 962 (1973).

⁸S. J. Putterman, Phys. Rev. Lett. **30**, 1165 (1973).

⁹D. N. Paulson, R. T. Johnson, and J. C. Wheatley, Phys. Rev. Lett. **31**, 746 (1973).

¹⁰The second longitudinal resonance occurred at a frequency only 1–2% greater than $2f$.

¹¹The quantity $(f/C_1)^{4\text{He}, T=0}$ varied from 0.088 at zero pressure to 0.081 on extrapolating the pressure measurements to 34 bar. Values of C_1 for ^4He were ob-

tained from B. M. Abraham, Y. Eckstein, J. B. Ketterson, M. Kuchnir, and P. R. Roach, Phys. Rev. A **1**, 250 (1970). At zero pressure the corresponding index of refraction is 2.16, rather different from the value of 1.34 expected from the empirical factor used in Ref. 3; however, both the powder and the packing are quite different from those used in Ref. 3.

¹²R. A. Webb, T. J. Greytak, R. T. Johnson, and J. C. Wheatley, Phys. Rev. Lett. **30**, 210 (1973).

¹³M. Kriss and I. Rudnick, J. Low Temp. Phys. **3**, 339 (1970).

¹⁴A. J. Leggett, Phys. Rev. **140**, A1869 (1965).

¹⁵See J. C. Wheatley, Physica (Utrecht) **69**, 218 (1973), for a recent compilation.

¹⁶R. Balian and N. R. Werthamer, Phys. Rev. **131**, 1553 (1963).

¹⁷J. Seiden, C. R. Acad. Sci., Ser. B **276**, 905 (1973).

Mass Diffusivity of ^3He - ^4He Mixtures near the Superfluid Transition

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We report on measurements of relaxation times for the decay of isothermal concentration gradients in ^3He - ^4He mixtures over the temperature range $10^{-5} \lesssim |\epsilon| \equiv |T/T_\lambda - 1| \lesssim 10^{-2}$. The data yield a mass diffusivity D which diverges when T_λ is approached from higher temperatures. When fitted with $D \propto \epsilon^{-z}$, they give $z = 0.40, 0.36,$ and 0.33 for ^3He concentrations 0.100, 0.208, and 0.399, respectively. The estimated uncertainty for z is ± 0.06 . These results are consistent with theoretical predictions, but differ from experimental results by others.

Recent measurements¹ of mass diffusion in dilute ^3He - ^4He mixtures (^3He concentration $X < 1\%$) seem to imply that the mass diffusivity D remains finite when the superfluid transition temperature T_λ is approached from higher temperatures. This behavior would be in conflict with theoretical predictions.²⁻⁶ We wish to report on measurements of relaxation times for isothermal concentration gradients in ^3He - ^4He mixtures near T_λ . Our results are for molar ^3He concentrations $X = 0.100, 0.208,$ and 0.399 , and for $10^{-5} \lesssim |\epsilon| \equiv |T/T_\lambda - 1| \lesssim 10^{-2}$. Approaching T_λ from higher temperatures, they reveal a divergent D with critical exponents which, within the uncertainties of theory and experiment, agree with the predictions^{3,4} of dynamic scaling^{2,3} and of mode-mode-coupling calculations.⁶

A schematic diagram of our sample cell is shown in the inset of Fig. 1. Most of the mixture is contained in the 0.50-cm-long holes of 0.11 cm diameter in part A . These holes have a center-to-center spacing of 0.14 cm, and yield a 50% transparency. In addition there are two 0.10-cm-high spaces filled with sample, one each above and below the perforated center section of part A . These spaces each contain a capacitor to be used for high-precision measurements of the concentrations of the mixture at the ends of our sample cell. One plate of each capacitor is shown as part B or part B' . The other, shown as a dashed line, was a sheet of 0.012-cm-thick stainless steel, perforated by 0.012-cm-diam holes with a 0.025 cm center-to-center distance (37% transparency).⁷ The spacings between the capacitor

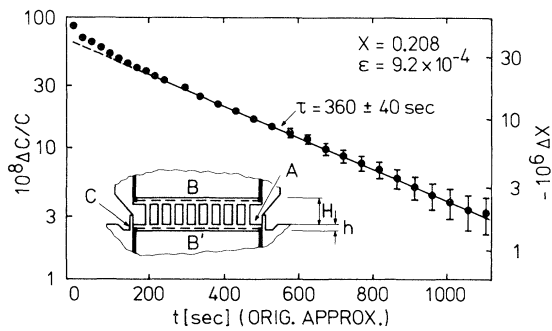


FIG. 1. Relaxation of the capacitance change $\Delta C/C$ after a steady-state heat current was turned off. After about 200 sec the temperature gradient had decayed, and the remaining $\Delta C/C$ corresponded to the concentration difference ΔX shown on the right-hand ordinate. The linearity of the data indicates the existence of a unique isothermal concentration relaxation time τ .

plates were 0.010 cm.

Isothermal conditions in the entire cell are guaranteed in the absence of deliberately induced heat currents because parts A , B , and B' are made of copper. However, it is possible to create a vertical temperature gradient between parts B' and A because the connecting piece C provides poorly conducting stainless-steel walls over a height h of 0.10 cm. A heat current flowing from B' to A creates a temperature gradient which is associated with a concentration gradient and a gravitationally stable density gradient over the height h ; but the remainder of the sample, of height $H=0.6$ cm between the bottom surfaces of parts A and B , will remain isothermal and will have a uniform concentration. When the heat current is turned off after a steady-state concentration gradient has been established, then the thermal gradient over the height h relaxes with a thermal relaxation time τ_T of about 30 sec. For $T < T_\lambda$, the pertinent mass diffusivity D_{eff} is an effective diffusivity which is infinite, and concentration gradients are always determined by the thermal gradients and the condition that the chemical potential gradients be zero.⁸ Therefore the measured mass relaxation time τ should in that case be the same as τ_T . However, for $T > T_\lambda$ the mass diffusivity D is finite. Although we shall see that D diverges near T_λ and therefore becomes larger than the thermal diffusivity D_T , the mass relaxation times τ for our system are larger than τ_T because mass diffusion must take place over a length $H+h=0.70$ cm whereas heat conduction through the helium proceeds only over a length $h=0.10$ cm. Since the relaxation times

vary as the square of the length, one has approximately $\tau/\tau_T=50$ if $D=D_T$. Although sufficiently near T_λ one expects $D \gg D_T$, one can estimate from the dynamic-scaling prediction to be discussed below, and we find from our measurements, that $\tau > \tau_T$ over the entire accessible range of ϵ , and that it is therefore possible to permit the temperature gradient to vanish, and then to study the decay of the remaining concentration gradient under the condition $\nabla T=0$.

In Fig. 1 we show a typical example of the observed concentration relaxation. The return of the capacitance change ΔC in the bottom condenser to the steady-state value $\Delta C=0$, a corresponding to $\nabla T=0$ and $\nabla X=0$, is shown on a logarithmic scale as a function of time after a heat current of $33 \text{ erg sec}^{-1} \text{ cm}^{-2}$ was turned off (a steady-state ∇X had of course first been established). After switching the heat current off, we observe a fast decay of the capacitance for about 200 sec, corresponding in part to the decay of the temperature gradient. From then on the decay of ΔX occurs under isothermal conditions, and the measured ΔC is proportional to ΔX . We estimate that possible systematic errors in the measured concentration changes from long-term drifts may be about 6×10^{-7} , corresponding to errors in the capacitance of 0.6×10^{-6} pF out of 53 pF, or about 1 part in 10^8 . Corresponding error bars are shown in the figure. The data for elapsed times greater than several thermal relaxation times (say $t \geq 200$ sec) can be fitted well by a straight line, implying the existence of a single dominant concentration relaxation time τ . The estimated systematic error yields an uncertainty in τ of about 10%.

In Fig. 2 we collect the measured concentration relaxation times τ over the range $\epsilon = \pm 3 \times 10^{-4}$ for $X=0.208$. Also shown are the thermal relaxation times τ_T which were measured separately with a thermometer. It is evident that $\tau = \tau_T$ below T_λ , as we expected from two-fluid hydrodynamics⁸ with the infinite effective mass diffusivity D_{eff} . However, for $T > T_\lambda$ we have $\tau > \tau_T$. The decrease in τ towards τ_T as T_λ is approached from higher temperatures, resulting from the divergence of D , is evident from the data.

In Fig. 3 we show τ for all three concentrations as a function of ϵ on logarithmic scales. The measurements extend over three decades in ϵ , from 10^{-5} to 10^{-2} . For each concentration they can be represented within their scatter by a straight line which corresponds to $\tau = \tau_0(X)\epsilon^z$. For our ϵ range we obtain $z = 0.40, 0.36, \text{ and } 0.33$

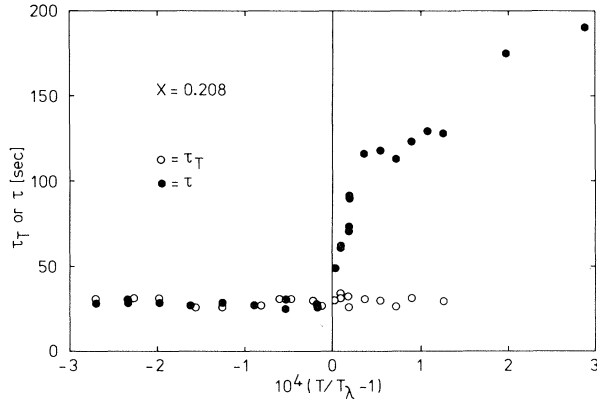


FIG. 2. Concentration (τ) and thermal (τ_T) relaxation times as a function of $T/T_\lambda - 1$ on linear scales. Whereas singularities in τ_T at T_λ are undetectable with our resolution, a rapid decrease of τ towards τ_T due to the diverging mass diffusivity is evident when T_γ is approached from higher T .

for $X = 0.100, 0.208, \text{ and } 0.399$, respectively. For all concentrations we estimate that random and systematic errors result in an uncertainty of ± 0.06 for z . At an ϵ of 10^{-4} our data yield $\tau = 215, 146, \text{ and } 105$ sec for $X = 0.100, 0.208, \text{ and } 0.399$, respectively.

The normal modes in mixtures of ^3He and ^4He have been examined by Griffin.⁹ They are combinations of mass diffusion and heat conduction with diffusivities D_A and D_B which are related⁹ to the mass diffusivity D and the thermal diffusivity D_T . On the basis of dynamic scaling^{2,3} it is predicted³ that one of D_A and D_B diverges asymptotically as $u_2 \xi$ when T_λ is approached from higher T . Here u_2 is the second-sound velocity below T_λ , and ξ is the correlation length for fluctuations in the order parameter. From experiment⁴ it is known that D_T remains finite at T_λ . For that case, a divergent D_A or D_B is possible only if D diverges.⁴ For a divergent D , one has $D \gg D_T$ sufficiently near T_λ , and then⁴

$$D_A = (1 + Z_2)D + Z_2(1 + Z_2)^{-1}D_T + O(D_T^2/D) + \dots \quad (1)$$

Here Z_2 is a parameter involving thermodynamic variables and transport coefficients which vanishes for $X = 0$.^{4,9} However, for $X > 0$ one has $Z_2 \sim \epsilon^{-\alpha}$, where α is the exponent which describes the singularity in the specific heat at constant chemical potential. Since α is near zero^{10,11} and probably negative,¹⁰ we do not expect the factor $1 + Z_2$ to result in an ϵ dependence of D which is very different from that of D_A ; but the effect of

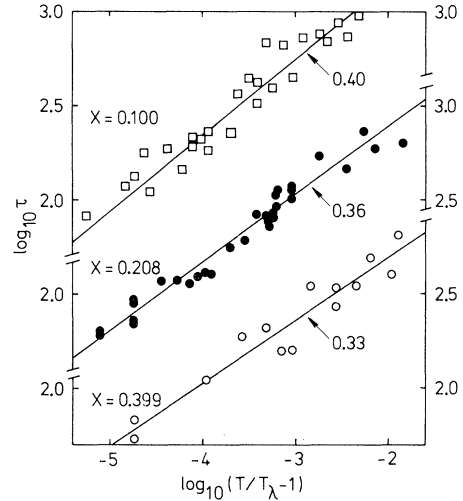


FIG. 3. Concentration relaxation times τ as a function of $T/T_\lambda - 1$ on logarithmic scales. The indicated slopes (0.40, 0.36, 0.33) are equal to the effective exponents z of the mass diffusivity.

Z_2 upon the amplitude of D can be estimated only when an additional transport coefficient becomes known. In any event, for $\alpha \lesssim 0$ Eq. (1) shows that a divergent D implies $D_A \sim D$. The other diffusivity, D_B , can be shown to remain finite if D diverges and D_T is finite.⁴ It follows that the dynamic-scaling prediction pertains to the mass diffusivity, and

$$D \sim u_2 \xi. \quad (2)$$

A similar prediction follows from mode-mode-coupling calculations.⁶ For our mass relaxation time we have

$$\tau = L^2/D \sim 1/u_2 \xi.$$

Here L is a characteristic length of our sample cell over which the diffusion takes place, and is on the order of 0.7 cm.

If one assumes a pure power law for ξ with a universal exponent of 0.67,^{12,13} and if one uses the measured u_2 ,^{13,14} one obtains an effective exponent $z \approx 0.27$. This prediction is lower than the measured values given above. However, it is not known whether a pure power law with the asymptotic exponent yields the appropriate correlation length for Eq. (2). Thus, one might calculate ξ from the relation^{2,3} $\xi \propto 1/\rho_s$, using the measured ρ_s .^{13,14} This, and the measured u_2 ,^{13,14} gives effective exponents for D , which vary from 0.31 to 0.41 as X changes from 0.1 to 0.4. The trend with X of this prediction is opposite to that observed, and we regard it as unlikely that the ex-

perimental errors are large enough to permit this behavior. As a third alternative, however, we recognize that there are appreciable ϵ -dependent contributions to u_2 which do not affect the asymptotic behavior, and it is not clear whether these are considered correctly by dynamic scaling. If the prediction Eq. (2) pertains only to the asymptotically dominant terms, one might use $u_2 \propto \epsilon^{0.33}$ and $\xi \propto \epsilon^{-0.67}$ for all X and predict a concentration-independent $z \approx 0.34$. This value falls within the permitted range of the experimental result. Nonetheless, we remark that the measured z for $X=0.1$ tends to be higher than any of the predicted values. There has been a tendency also for measurements^{15,16} of other transport properties to yield exponents which are larger than the predictions.

One might expect also to obtain the approximate amplitude of D from dynamic scaling by setting numerical constants equal to unity and treating the asymptotic proportionality $D_A \sim u_2 \xi$ as an equality. However, the large latitude in the choice for ξ and u_2 , and the unknown size of Z_2 in Eq. (1), permit at best an estimate for the order of magnitude of D . For $X=0$ and at $\epsilon=10^{-4}$ one obtains $D \approx u_2 \xi \approx 1.2 \times 10^{-3}$ cm²/sec if one uses $\xi = \xi_0 \epsilon^{-0.67}$, $\xi_0 = 2 \times 10^{-8}$ cm, and the measured u_2 .¹² With a characteristic length for our geometry of 0.7 cm, this yields $\tau \approx 400$ sec at $\epsilon=10^{-4}$ and $X=0$. This is consistent with the experimental values of $\tau(X)$ quoted above and shown in Fig. 3.

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¹G. M. Drabkin, V. A. Noskin, V. A. Trunov, A. F.

Shchebetov, and A. Z. Yagud, Zh. Tekh. Fiz. 42, 180 (1972) [Sov. Phys. Tech. Phys. 17, 142 (1972)]; G. M. Drabkin, V. A. Noskin, and A. Z. Yagud, Pis'ma Zh. Eksp. Teor. Fiz. 15, 504 (1972) [JETP Lett. 15, 357 (1972)]; G. M. Drabkin, V. A. Noskin, E. G. Tarovik, and A. Z. Yagud, Phys. Lett. 43A, 83 (1973).

²R. A. Ferrell, N. Ményhard, H. Schmidt, F. Schwabl, and P. Szépfalussy, Phys. Rev. Lett. 18, 891 (1967), and Phys. Lett. 24A, 493 (1967), and Ann. Phys. (New York) 47, 565 (1968).

³B. I. Halperin and P. C. Hohenberg, Phys. Rev. Lett. 19, 700 (1967), and Phys. Rev. 177, 952 (1969).

⁴G. Ahlers, Phys. Rev. Lett. 24, 1333 (1970).

⁵M. A. Eggington and A. J. Leggett, J. Low Temp. Phys. 5, 275 (1971).

⁶K. Kawasaki and J. D. Gunton, Phys. Rev. Lett. 29, 1661 (1972); M. K. Grover and J. Swift, J. Low Temp. Phys. 11, 751 (1973).

⁷Buckbee-Mears Co., St. Paul, Minn.

⁸For a discussion of the hydrodynamics of superfluid ³He-⁴He mixtures, see I. M. Khalatnikov, *Introduction to the Theory of Superfluidity* (Benjamin, New York, 1965), Part IV.

⁹A. Griffin, Can. J. Phys. 47, 429 (1969).

¹⁰G. Ahlers, Phys. Rev. A 8, 530 (1973).

¹¹F. M. Gasparini and M. R. Moldover, Phys. Rev. Lett. 23, 749 (1969); F. M. Gasparini, Ph. D. thesis, University of Minnesota, 1970 (unpublished).

¹²D. S. Greywall and G. Ahlers, Phys. Rev. A 7, 2145 (1973).

¹³W. C. Thomlinson, G. G. Ihas, and F. Pobell, Phys. Rev. Lett. 31, 1284 (1973).

¹⁴G. Ahlers and D. S. Greywall, Phys. Rev. Lett. 29, 849 (1972), and to be published; G. Terui and A. Ikushima, Phys. Lett. 43A, 255 (1973).

¹⁵J. A. Tyson, Phys. Rev. Lett. 21, 1235 (1968).

¹⁶G. Ahlers, in *Proceedings of the Twelfth International Conference on Low Temperature Physics, Kyoto, 1970*, edited by E. Kanda (Academic Press of Japan, Tokyo, 1971), p. 21.

Third Sound and the Healing Length of He II in Films as Thin as 2.1 Atomic Layers*

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Measurements of the velocity of third sound on films as thin as 2.1 atomic layers yield the healing length of superfluid He II down to temperatures of 0.1 K. It is argued that these films are two-dimensional superfluids.

The role of dimensionality in determining the properties of ordered phenomena such as magnetism, superconductivity, and superfluidity is a subject of continued and current interest. Of particular concern is the question of whether coherent phenomena observed in three-dimensional

configuration space persist in two-dimensional space.¹ He II is distinguished by the occurrence of extremely thin uniform films (measured in angstroms or atomic layers) and it is fair to regard them as two-dimensional if such a concept has any meaning for actual systems. There is a