

NONLINEAR CONCENTRATION DEPENDENCE OF SOUND VELOCITY IN ^3He - ^4He SOLUTIONS*

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We present measurements of the change in the velocity of sound in liquid ^4He on the addition of small quantities of ^3He for three compositions and three frequencies. It is shown that the change is not strictly linear in ^3He concentration and that the nonlinearity may be explained to a large extent if corrections to the ^3He effective mass, which arise from the many-body interactions, are included.

In a previous communication,¹ it was shown that the temperature dependence of the attenuation in ^3He - ^4He solutions was in qualitative agreement with theory^{2,3} but the temperature dependence of the velocity was not. Since only velocity changes relative to the velocity at 35 mdeg K were measured, nothing could be said about the concentration dependence which is expected to be an order of magnitude greater. In this communication, we present measurements of the change in the velocity of sound as ^3He is added to pure ^4He , which yields the velocity of sound in a solution of known composition to a precision limited only by the knowledge of the velocity of sound in pure ^4He .

Khalatnikov,⁴ by using a generalization of the two-fluid model to include the effect of impurities, has shown that the velocity of sound in a solution, $c(X)$, is given (to first order in X) at zero

frequency by

$$[c(X)]^2 = \left(\frac{dP}{d\rho(X)} \right)_{X, T} \left[1 + \frac{\rho_s}{\rho_n} \left(\frac{d\rho}{dX} \frac{X}{\rho} \right)^2 \right], \quad (1)$$

where X is mole fraction of ^3He , P is the pressure, $\rho(X)$ is the concentration-dependent density; ρ_s and ρ_n are the superfluid and normal-fluid densities, respectively. The normal-fluid density is given (again to first order in X) by $\rho_n = \rho(m^*/m_4)X$, where m^* is the effective mass of ^3He and m_4 the mass of ^4He . If the molar volume of the solution is related to that of pure ^4He by the expression $V = V_4(1 + \alpha X)$, then it follows that the density (to terms linear in X) is given by

$$\rho(X) = \rho_4 \left[1 - \alpha X + \frac{m_3 - m_4}{m_4} X \right].$$

Substituting the expressions for $\rho(x)$ and for ρ_n into Eq. (1) gives the following expression for $c(X)$ in terms of experimentally measured parameters:

$$\frac{c(X) - c(0)}{c(0)} \equiv \beta X = \frac{X}{2} \left[\left(1 + \alpha - \frac{m_3}{m_4} \right) + \rho \frac{d\alpha}{d\rho} + \frac{m_4}{m^*} \left(1 + \alpha - \frac{m_3}{m_4} \right)^2 \right].$$

Equation (2) was originally derived by Baym³ by using a kinetic method; the above derivation is due to Massey.⁵ In order to estimate β , we substitute for α the value 0.28 from Kerr,⁶ for $\rho d\alpha/d\rho$ the value -1.4 from Boghosian and Meyer,⁷ and for m^* the value $2.5m_3$ from Sandiford and Fairbanks⁸ and obtain the value $\beta = -0.35$. Thus, one would expect the velocity of sound in a 1-mole% solution to decrease about 0.4% from that in pure ^4He .

Conventional transit-time measurements using pulse-echo techniques are not sufficiently sensitive to determine such small effects with precision; nor can the recently devised method of McSkimin⁹ be applied here because it requires a wide-bandwidth (loaded) transducer which greatly reduces the signal to noise.^{10,11} We have used for these measurements a modification of the phase-cancellation technique^{10,11} which permits

a resolution to a few parts in 10^6 . Both the change in phase and amplitude of the signal traversing the helium were monitored as an accurately known quantity of ^3He was slowly admitted to the sonic cell. From the change in phase, or the equivalent, the change in transit time, Δt , we deduce the velocity in the solution relative to ^4He , $[c(X, T, f)]^{-1} - [c(0, T, f)]^{-1} = \Delta t/d$, where d is the acoustic path length. Three precisely known quantities of ^3He were prepared for each frequency such that the compositions after each increment was added corresponded to nominal 1.3, 3.0, and 5.5 mole%. The ^3He was added to an accurately known amount of ^4He , and the change in velocity (transit time) and attenuation were measured for each addition. The procedure was to cool the ^4He to the lowest temperature (165 mdeg K), then record the change in

Table I. Observed transit time of a sound wave in three solutions of ^3He - ^4He at 165 mdeg K relative to the transit time in pure ^4He for three frequencies, and the absolute attenuation of the same solutions at 165 mdeg K.

f (MHz)	X (mole fraction)	$\Delta t/d$ (nsec/cm)	$\frac{c(X, 165, f) - c(0, 165, f)}{Xc(0, 165, f)}$	$\alpha(X, 165, f)$ (dB/cm)
14.935 402	0.0129 ₅	89.7 ₈	-0.164 ₈	0.8 ± 0.2
	0.0299 ₀	210.7 ₆	-0.167 ₁	0.6 ± 0.2
	0.0551 ₂	397.3 ₆	-0.170 ₁	1.4 ± 0.2
44.994 458	0.0126 ₈	85.6 ₉	-0.160 ₆	4.5 ± 0.2
	0.0300 ₀	207.5 ₀	-0.164 ₀	10.6 ± 0.2
	0.0549 ₇	390.3 ₅	-0.167 ₆	16.2 ± 0.2
75. 016 768	0.0127 ₇	85.2 ₀	-0.158 ₈	5.5 ± 0.2
	0.0297 ₈	203.7 ₀	-0.162 ₃	19.9 ± 0.2
	0.0552 ₆	390.3 ₈	-0.166 ₈	37.4 ± 0.2

phase and amplitude as the cell was warmed to ca. 440 mdeg K at which temperature the first increment of ^3He was added. The following day, after the apparatus had warmed to 4.2°K, the cell was cooled again to 165 mdeg K and the transit time and attenuation for the solution measured as a function of temperature up to ca. 440 mdeg K before adding the next increment of ^3He . The temperature dependence of the velocity for the nominal 5.5% solutions agreed with the published measurements¹ to within ±0.2 nsec, and also the check point for the previous day's addition agreed within ±0.2 nsec. We thus have an experimental verification of the homogeneity of solution and of the reproducibility of measurement. After three additions, the cell was warmed to 77°K, a mass (volume) balance was obtained on the gas, and the cell was then thoroughly evacuated for the next experiment. A sonic cell which had been used for other measurements¹¹⁻¹³ was modified by adding a fill line. It should be pointed out that the cell reservoir (35 cc) was large enough to contain the total solution (ca. 900 cc gas NTP) at less than one atmosphere at 4.2°K. The cerium (III) magnesium nitrate thermometer was thermally and mechanically anchored to the cell.^{12,13}

The data are summarized in Table I. All entries are given to the limits of precision although the observed delay times (column 3) are accurate only to ±1.5 nsec. If the velocity shift were strictly linear in concentration and temperature and frequency independent, then all entries in column 4 would be identical. However, m^* , α , and $\rho d\alpha/d\rho$ can be concentration and temperature dependent. Moreover, as shown in Ref. 1, there is a correction of order Xv_F^2 which does not vanish as T and f approach zero. It is, nonetheless, clear from the entries in Table I that theory³ is in accord with experiment to first order,

but the precision of these experiments is such as to require a more detailed theory for sonic propagation through the solutions.

Figure 1 shows the change in transit time per unit path length ($\Delta t/d$) relative to pure ^4He for the three concentrations studied at 165 mdeg K for the frequency of 75 MHz. The size of the points corresponds to ±2.5 nsec, ten times the actual precision. It is clear from the graph, as was also evident from the table, that the velocity change is not strictly linear with composition. The dashed line in the figure was drawn using an estimate of the limiting slope as both X and T approach zero; the solid line was calculated from Eq. (2) using a concentration-dependent effective mass¹⁴ m^* , and $\rho d\alpha/d\rho = -1.01$. The value -1.01 for $\rho d\alpha/d\rho$ was obtained from the limiting slope and the most recent values for¹⁴ $m^*(X=0) = 2.36m_3$ and for¹⁵ $\alpha = 0.284$; an extrapolation to zero frequency yields the value -1.03. It is clear from

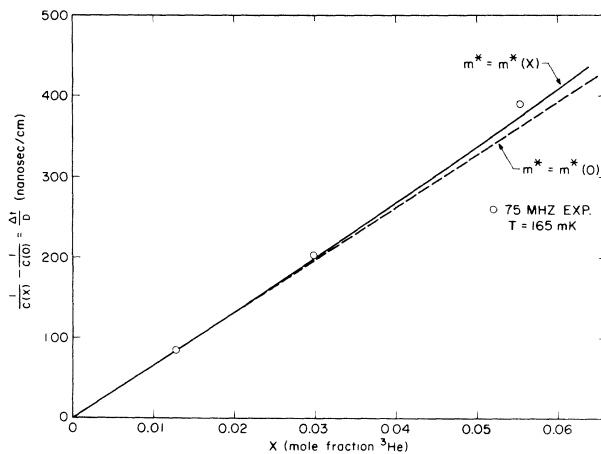


FIG. 1. $\Delta t/d$ vs X at 75 MHz. Circles are experimental points. The dashed line is the limiting slope at zero concentration; the solid line shows the effect of a concentration-dependent effective mass m^* (Ref. 14).

the figure that the use of concentration-dependent effective mass brings the theoretical calculation into better agreement with experiment.

There is, however, still a disagreement which is outside the experimental error; it is possible that agreement would be improved if one could correct the velocity change to zero temperature. Here, then, is the reason for displaying the 75-MHz data, the smaller temperature dependence for the velocity at this frequency.

Column 5 allows us to renormalize our previous attenuation measurements (Ref. 1, Fig. 2). According to the present experiment one should subtract 1.5, 0.3, and 1.0 dB/cm from the 15-, 45-, and 75-MHz data, respectively.¹ These small corrections verify the observation by Harding and Wilks¹⁸ (which was the basis for our previous normalization) that the attenuation in the solutions is the same as in pure ⁴He at 1.9°K. We may use the data of column 3 (interpolated to 5.527 mole%) to normalize our previous measurements¹ of the temperature dependence of the sound velocity relative to that of pure ⁴He at absolute zero. If to the delay time of each point in Fig. 1, Ref. 1 one adds 376.3, 371.7, and 372.5 nsec/cm for 15, 45, and 75 MHz, respectively, then the ordinate becomes $[c(X, T, f)]^{-1} - [c(0, T, f)]^{-1}$ to within our experimental error of ± 1.5 nsec/cm. A larger error is quoted for these values since they rely on the measurement for ⁴He. With the filling tube, we observed oscillations which made the phase balance uncertain to that amount.

We conclude by noting the large difference between our value of $\rho d\alpha/d\rho$, -1.01 , and the direct measurements of Boghosian and Meyer¹⁷ who find -1.4 . The discrepancy between the two is puzzling since the theory for the change in sound velocity rests only on the two-fluid model. Either number is within the estimated error of the

calculated value of Massey and Woo.¹⁸

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