Sound velocity in helium-filled porous Vycor glass

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The velocity of longitudinal and transverse sound in helium-filled porous Vycor glass was measured and analyzed with the use of the Biot theory of sound propagation in fluid-filled porous media. From transverse wave velocities, the superfluid fraction was determined and, from longitudinal velocities, the elastic properties of the helium were determined.

The superfluid properties of ⁴He in Vycor (a porous silica glass with a network of interconnected pores about 30 Å in radius) have been studied in various experiments,¹⁻³ and we recently⁴ observed freezing of ⁴He in Vycor at pressures considerably above the bulk melting curve. Recently,⁵ there has been interest in ⁴He-filled porous glass as an example of a system to which the Biot theory⁶ of sound propagation in fluid-filled porous media can be applied. The unique properties of superfluid ⁴He permit the limiting (zero viscosity) features of the Biot theory to be explored in a manner not accessible with conventional fluids.

Our experimental method has been described elsewhere.⁴ The Vycor sample was 0.3 cm in diameter and 1.02 cm in length and was contained in a pressure cell with a volume of 2.4 cm³. From the shift in the superfluid transition temperature T_{λ} , obtained from both velocity and attenuation measurements,⁴ we estimated the average pore radius to be 34 Å.

The theory of sound propagation in a fluid-filled porous solid has been treated in a series of papers by Biot,⁶ and the case of superfluid ⁴He has been discussed by Johnson et al.⁵ The main result of the theory is that, at low frequencies, two types of waves can propagate, the usual longitudinal and transverse ones. At high frequencies, there is, in addition, a third wave in which the fluid moves relative to the solid matrix. This "slow wave," which has only recently been observed in a conventional system,⁷ is just the familiar fourth sound wave in superfluid helium. The relevant frequency is $\omega_c = 2\eta/\rho R^2$, where η and ρ are the fluid's viscosity and density and R is the pore size. At sound frequencies below ω_c , the viscous penetration depth $\delta = (2\eta/\rho\omega)^{1/2}$ is larger than the pore size and the fluid is viscously locked to

the solid matrix. At frequencies above ω_c , only a fluid layer of thickness $\delta < R$ is locked to the matrix and the rest of the fluid "decouples" from it. This decoupling allows the slow wave to propagate and, in addition, modifies the velocities of the transverse and the fast longitudinal waves.

Biot's complicated expressions simplify considerably when the pores contain helium whose elastic moduli are much smaller than those of Vycor. In this "rigid frame limit"⁶ the low-frequency $(\omega \ll \omega_c)$ expressions for the transverse (v_t) and longitudinal (v_l) sound velocities are

$$v_t^2 = \mu_V / (\rho_V + \phi \rho_{\text{He}}) \tag{1a}$$

and

$$v_I^2 = \frac{(K_V + 4\mu_V/3) + (1/\phi)(1 - K_V/K_s)^2 K_{\text{He}}}{\rho_V + \phi \rho_{\text{He}}} ,$$
(1b)

and the slow wave does not propagate. Here, ϕ is the porosity ($\phi \approx 0.28$), ρ is density, μ and K are shear and bulk elastic moduli, and the subscripts V, s, and He refer to empty Vycor, the solid glass which makes up the Vycor matrix, and ⁴He, respectively. There are two effects due to filling the pores with helium. First, the density is increased from ρ_V to $\rho_V + \phi \rho_{\text{He}}$. Secondly, the elastic moduli are changed. Since the liquid helium has no shear modulus, the transverse velocity is not affected but the effective modulus for compressional waves is increased by an amount proportional to the bulk modulus of helium, K_{He}.

In the high-frequency, rigid frame limit (ω $\gg \omega_c$), the velocities become

$$v_{t}^{2} = \mu_{V} / [\rho_{V} + (1 - 1/\alpha)\phi\rho_{He}] , \qquad (2a)$$

$$v_{t}^{2} = \{ (K_{V} + 4\mu_{V}/3) + [(1/\phi)(1 - K_{V}/K_{s})^{2} - (1/\alpha)(2 - \phi - K_{V}/K_{s})]K_{He} \} / [\rho_{V} + (1 - 1/\alpha)\phi\rho_{He}] , \qquad (2b)$$

$$v_{slow}^{2} = K_{He}/\alpha\rho_{He} = v_{4th \ sound} , \qquad (2c)$$

where α is the decoupling parameter (see below). The main difference from the low-frequency expressions for v_l and v_l is in the effective density which is reduced by an amount $\phi \rho_{\rm He}/\alpha$. This is the amount of helium which "decouples" from (i.e., is not viscously locked to) the Vycor. The decoupling parameter α depends only upon the geometry of the porous medium and is related⁵ to the index of refraction *n* for fourth sound by $\alpha = n^2$. There is also a change in the effective modulus for longitudinal sound.

For liquid ⁴He above the superfluid transition, $\eta \approx 3 \times 10^{-5}$ poise and $\rho_{\text{He}} \approx 0.16 \text{ g/cm}^3$, so $\omega_c/2\pi \approx 500 \text{ MHz}$ for the 34-Å Vycor pores. Thus at our measuring frequencies of 10 to 20 MHz, we are always in the low-frequency limit [Eq. (1)] above the superfluid transition temperature. Below T_{λ} , however, the viscosity of the superfluid fraction is zero and so $\omega_c = 0$. Thus superfluid ⁴He in Vycor represents the high-frequency limit [Eq. (2)] of the Biot theory.

Here, we first measured v_i and v_i in empty Vycor at 4.2 K. We then measured the velocity changes (also at 4.2 K) as we filled the Vycor with liquid ⁴He at saturated vapor pressure, and as we pressurized the liquid ⁴He up to 140 bars. The results are shown in Fig. 1(a).

The decrease in $v_t(p)$ as the ⁴He pressure in-



FIG. 1. Liquid ⁴He-filled Vycor at 4.2 K. Pressure dependence of (a) normalized longitudinal and transverse sound velocities and (b) change in effective modulus due to the ⁴He. [The solid line is the calculated change in bulk modulus using Eq. (4) and the pressure-dependent modulus of bulk ⁴He.]

creases is simply due to the increase in ρ_{He} with pressure. Using the manufacturer's quoted value of $\phi = 0.28$, the density ρ_{He} can be obtained from the empty and full values of v_t , using Eq. (1a). The densities thus derived from the data of Fig. 1(a) range from 0.16 \pm 0.01 g/cm³ at 1 bar up to 0.22 \pm 0.01 g/cm³ at 140 bars. At low pressures the densities are somewhat larger than the bulk ⁴He densities (e.g., 0.125 g/cm³ at 1 bar and 4.2 K). This increased average ⁴He density in Vycor has been previously observed⁸ and was attributed to the presence of one or two atomic layers of high-density ⁴He on the pore surfaces.

Were it not for the additional modulus term

$$(1/\phi)(1-K_V/K_s)^2 K_{\rm He}$$

in Eq. (2b), the values of $v_t^2(p)/v_t^2$ (empty) and $v_l^2(p)/v_l^2$ (empty) would be the same. From Fig. 1(a), they are clearly different at high pressures. This difference we attribute to the helium bulk modulus which increases rapidly with pressure. The difference shown in Fig. 1(a) between v_l and v_t can be expressed as a change in effective modulus ΔK for v_l , i.e.,

$$v_{l}^{2}(p) = [(K_{V} + 4\mu_{V}/3) + \Delta K(p)]/(\rho_{V} + \phi\rho_{He}) ,$$
(3)

where the (Biot) expression (1b) predicts

$$\Delta K(p) = (1/\phi)(1 - K_V/K_s)^2 K_{\text{He}}(p) \quad . \tag{4}$$

Figure 1 (b) shows the values of ΔK derived from the velocity data compared to the Biot prediction (solid curve) calculated from Eq. (4). We used for K_{He} the bulk ⁴He modulus taken, e.g., from Wilks⁹ and $K_V/K_s \approx 0.33$ from the measured sound velocities in porous and fused (solid) Vycor. The differences at low pressures may reflect a ⁴He modulus in Vycor larger than the bulk value, similar to the increased density discussed above.

One manifestation of the transition from normal to superfluid is the appearance of the Biot slow wave (fourth sound) below T_{λ} as has been discussed by Johnson *et al.*⁵ However, we found that, in addition, v_t and v_l show the decoupling predicted by the Biot theory, a result not previously observed.

From Eqs. (1) and (2), the effect of going from $T > T_{\lambda}$ ($\omega \ll \omega_c$) to $T \ll T_{\lambda}$ ($\omega \gg \omega_c = 0$) is to decouple a fraction $1/\alpha$ of the helium density from the Vycor and thus to increase v_t and v_t . At saturated vapor pressure, the modulus effect in Eqs. (1b) and (2b) can be neglected. Figure 2 shows the normalized v_t and v_t as functions of temperature at saturated vapor pressure. The increase in the velocities due to decoupling is clearly shown below the superfluid transition temperature of 1.94 K. Also shown are the velocities in empty Vycor which are much less temperature dependent in this range.



FIG. 2. Normalized longitudinal and transverse sound velocities in empty Vycor (uppermost sets of points) and Vycor filled with ⁴He at saturated vapor pressure (lower sets of points). Note the break in vertical axis.

From the differences between the velocities just above T_{λ} and at the lowest temperature reached $(\approx 0.3 \text{ K})$, we found that a fraction ≈ 0.20 of the helium decoupled (cf. Fig. 2). The decoupling fraction has also been measured in torsional oscillator experiments (at frequencies in the low kHz range)^{1,2} and found to be about 0.16, somewhat smaller than our value. Bishop et al.¹ found that, at saturated vapor pressure, a fraction 0.34 of the ⁴He, corresponding to about two atomic layers, did not become superfluid and hence could not decouple. If we assume that in our experiment the same fraction 0.34 of the helium did not become superfluid, then the decoupling fraction $1/\alpha = 0.20/0.66 = 0.30$, and we get a value of 3.3 for the parameter α . As pointed out by Johnson et al.,⁵ this can be used to predict the fourth sound index of refraction for Vycor as $n = \sqrt{\alpha} \approx 1.8$. Although most reports on fourth sound experiments present only velocities normalized by the limiting lowtemperature velocity, we can estimate very roughly the index of refraction from the results of Gregory and Lim^{10} as $n \approx 2.1$.

The variation with temperature of the superfluid fraction ρ_s/ρ_{He} can also be obtained from the data of Fig. 2. Since the superfluid fraction is always in the high-frequency (decoupled) regime, while the normal component is viscously locked to the Vycor frame, the variation of v_t and v_l with temperature is the same as that of ρ_s .

In bulk ⁴He near the λ transition, the superfluid fraction varies as $(1 - T/T_{\lambda})^{\xi}$ where $\xi = \frac{2}{3}$. Figure 3 shows ρ_s/ρ_{He} (calculated from our velocity data) vs



FIG. 3. Superfluid density $\rho_s(T)/\rho_s(0)$ for ⁴He in Vycor (at saturated vapor pressure) from longitudinal and transverse sound velocities plotted on a log-log scale vs reduced temperature $1 - T/T_{\lambda}$, for $T_{\lambda} = 1.94$ K. The straight lines represent least-squares fits to the data and have slopes of 0.67 for the longitudinal waves and 0.69 for the transverse waves.

 $1 - T/T_{\lambda}$. Least-squares fits of the form $\rho_s/\rho_{\text{He}} = A(1 - T/T_{\lambda})^{\xi}$ give essentially the same exponent ξ for longitudinal and transverse measurements. (From the longitudinal data of Fig. 3, $\xi = 0.67$, and from the transverse data $\xi = 0.69$.) Fourth sound³ and torsional oscillator¹ experiments showed the same termperature dependence of ρ_s in Vycor with a similar exponent $\xi = 0.65 \pm 0.03$. This further demonstrates the usefulness of sound velocity measurements in studying superfluidity in restricted geometries.

In conclusion, we have measured longitudinal and transverse sound velocity behavior in liquid ⁴He-filled porous Vycor, as a function of temperature and pressure. We compared our results with Biot's theory of sound propagation in fluid-filled media and found this theory to apply well to the present case. In particular: (1) Biot's expression for the effective elastic moduli gave good agreement with the measured pressure dependence of the longitudinal sound velocity. (2) The transition from the low- to high-frequency limits of the Biot theory was achieved through the superfluid transition, and the resulting decoupling effect was found also to be consistent with the Biot theory. (3) From the temperature dependence of the sound velocities, the superfluid fraction in Vycor was found to have a temperature dependence (near T_{λ}) of the form $\rho_s(T) \propto (1 - T/T_{\lambda})^{\xi}$. The exponent ξ was essentially equal to the bulk value $\xi = \frac{2}{3}$.

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