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Porosity dependence of sound propagation in liquid ^{4}He filled aerogel

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Longitudinal sound wave propagation has been studied in an aerogel-liquid ^{4}He system for various porosities of aerogel. The superfluid transition was identified as the absorption peak, whose magnitude was suppressed by aerogel. The sound velocity was analyzed within a hydrodynamic theory in both normal and superfluid phases. The absorption peak due to phonon-roton interaction around 1 K was not observed even with the most porous aerogel. The low temperature sound velocity and attenuation show that direct collisions of phonons with aerogel strands plays an important role in the acoustic properties.

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Porous media filled with fluid have been intensively studied experimentally and theoretically because of their physical and technological importance. The effect of disordered pore structures on the properties of the fluid can be examined in these systems. There has been considerable interest in the behavior of superfluid ^{4}He in the presence of a random disorder induced by highly open porous media. Recent experiments on the superfluid transition of ^{4}He contained in porous media such as Aerogel, Xerogel and Vycor glass have revealed that the superfluid transition differs from that of bulk ^{4}He [1, 2]. The superfluid transition of ^{4}He in aerogel has been observed to be sharp [1, 2], and has suggested to manifest a genuine phase transition. The transition temperature in aerogel T_c has been suppressed with decreasing aerogel porosity.

Understanding the results of acoustic experiments is important when dealing with porous media. Use of liquid ^{4}He offers unique advantages due to the existence of the superfluid phase with more than one sound mode. The bulk fluid displays two propagating modes: first sound (compressional wave) and second sound (temperature wave)[3]. In a porous media where the normal component is clamped by its viscosity and only the superfluid component can move, fourth sound (relative motion of the superfluid and normal fluids) propagates and can be used to determine the superfluid fraction.

Longitudinal and transverse ultrasound velocity have been measured in ^{4}He filled Vycor glass[4]. Warner and Beamish[5] studied the transverse sound velocity and attenuation in alumina ceramics with various porosities. They argued that the experimental results in both the low and high frequency regimes for normal and superfluid phases can be quantitatively elucidated by the Biot model [6 – 9].

Silica aerogels are synthesized via a sol-gel process and hypercritical drying which enable production of tenuous solids with porosity ϕ as large as 99.8 % and unique acoustic properties. Silica aerogel is thought of as a network of nanoscale SiO_2 strands. The elastic moduli of aerogels are a few orders of magnitude smaller than that of bulk solids and the sound speed substantially depends on the porosity. Ultrasound measurements have shown sound speeds as low as 20 m/s for the highest porosity aerogel [10].

The high-porosity aerogels are so soft that the aerogel matrix and the clamped normal fluid moves as the results of pressure and temperature gradients, unlike other porous media. This results in sound modes intermediate between first and fourth sound [11] and a second-like mode [12]. McKenna et al. [12] calculated the longitudinal sound velocity for the two modes using modified two-fluid hydrodynamic equations in order to take aerogel motion into consideration. They also observed the propagation of both the fast (intermediate between first and fourth sound) and the slow (second-like sound) modes in ^{4}He in aerogel from 1.1 K to T_c. They found agreement of the model with the observed sound velocity within the experimental temperature range.

We have observed the 10 MHz longitudinal ultrasound signal with three different porosity (92.6, 94.0 and 94.8%) aerogels from 0.5 K to 4.2 K, and measured the sound velocity and attenuation in order to study sound propagation in the liquid ^{4}He filled aerogel system both

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in the normal and superfluid phases. Preliminary results have been published elsewhere[13]. The viscous penetration depth of liquid ^{4}He at 10 MHz (about 40 nm) is estimated to be longer than the typical SiO_2 strand distance (about 10 nm for 98% aerogel and much shorter for 95%); thus in the first approximation the normal fluid in these systems is expected to be completely locked to aerogel matrix by viscosity. The sound velocity of aerogel largely depends on the porosity so that we can obtain an aerogel whose sound velocity is larger or smaller than that of bulk fluid. It is interesting to see what would happen if the relation of sound velocities between aerogel and fluid is counterchanged.

Three aerogels were grown by a sol-gel process from tetramethoxysilane (TMOS) as a one step process. The porosities were determined using a standard dry weight method. We machined aerogels into cylinders (7 mm in diameter) which were enclosed in brass shells of 8.5 mm "outer" diameter and 3.0 mm length. The ends of the samples were polished flat and parallel. The sample cylinder was sandwiched between two $LiNbO_3$ transducers with springs. We used the same pair of transducers for every sample in order to easily compare the signal attenuation among aerogels. Aerogels were immersed in liquid ^{4}He at SVP. Temperatures were measured with a ruthenium oxide resistance thermometer and stabilized using a PI controller. The ultrasonic measurements were made using a standard pulse transmission and a phase sensitive detection technique.

We were able to observe the sound signal throughout the temperature range from 0.5 K to 4.2 K. The transmitted sound signal through the aerogel in a vacuum, however, could not be observed because of the imperfect connection between the transducers and aerogel or large attenuation results from the aerogel.

Figure 1 and 2, respectively, show the sound velocity u and attenuation α for the three aerogels as a function of temperature; for comparison, those of bulk helium obtained in a different run were plotted as well. The superfluid transitions T_c were identified as a dip in velocity and by an absorption peak for each aerogel. Those in aerogels were as sharp as in bulk, which represented a homogeneous transition in aerogel. T_c in aerogels are listed in Table. The magnitude of T_c suppression and porosity dependence agree with specific heat measurement[1, 2].

The temperature dependence of the sound velocity is similar to that of bulk for each aerogel. The absolute value varies inversely to the porosity. In ordinary porous media, the sound velocity is modified by tortuosity and the acoustic index n decreases with increasing porosity. The absolute value varies in opposition to the porosity

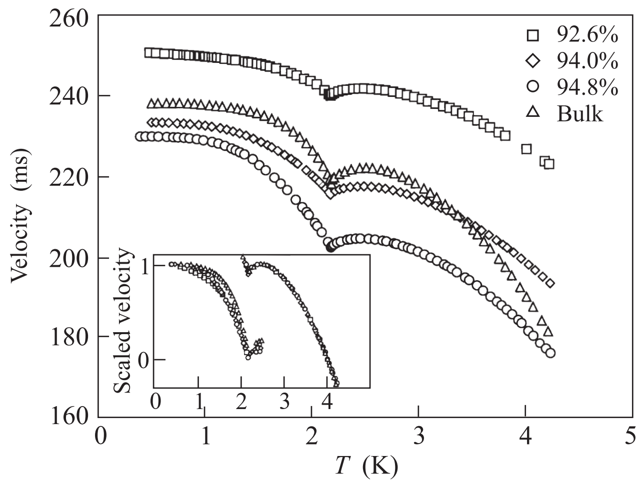

Fig.1. Sound velocity for various aerogels as a function of temperature. That of bulk helium is also shown for comparison. The inset shows the scaled sound velocity. Velocity is scaled between 2.5 K and 4.2 K, and 0.5 K and T_c, in the normal and the superfluid phase

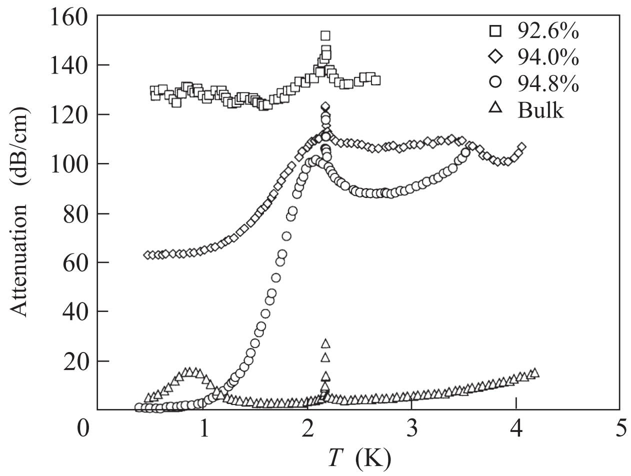

Fig.2. Attenuation of sound for various aerogels as a function of temperature. That of bulk helium is also shown for comparison. For clarity, that for 94% and 92.6% are shifted +20 dB/cm and +40 dB/cm, respectively. Those in the normal phase vary due to interference between the ultrasound signal and electrical feedthrough from the transducer and amplifier

dependence of the acoustic index; thus, tortuosity may not explain the behavior, but may still have an effect. The observed signal results from a compressional wave within liquid helium modified by aerogel.

The similarity of the temperature variation brings a scaled behavior to mind. Velocity for each aerogel and bulk liquid was scaled as $(u(T)-u(4.2\,\mathrm{K}))/(u(2.5\,\mathrm{K})-u(4.2\,\mathrm{K}))$ and $(u(T)-u(T_c))/(u(0.5\,\mathrm{K})-u(T_c))$ in normal and superfluid phase, respectively. The scaled temperature variations in the normal phase for each aerogel and for bulk liquid coincide with each other as shown in the inset of Fig.1. This means the temperature variation is determined mainly by the bulk liquid. However, that in the superfluid phase depends on the porosity. We will discuss this behavior in detail below.

It is useful to compare it with the longitudinal sound velocity in ^{4}He filled Vycor [4]. In the normal phase, the sound velocity in the Vycor system was almost constant, reflecting the constant sound velocity of Vycor glass. Contrary to the aerogel case, Vycor glass dominated the sound velocity of the composite system.

In a series of papers [6–9], Biot proposed a phenomenological theory of acoustic propagation in porous, fluid filled, macroscopically homogeneous and isotropic media. In [14] Johnson applied the Biot's theory for the superfluid ^{4}He in pores below 1.1 K when the normal component can be neglected. In the case of silica aerogel filled in liquid ^{4}He the parameters of that theory appear to be the tortuosity α, the aerogel porosity ϕ(fluid volume fraction), the density of liquid ^{4}He ρ_{He} and solid $\rho_{\mathrm{SiO_2}}$, the bulk modulus of helium K_{He}, the bulk modulus of silica $K_{\mathrm{SiO_2}}$, the bulk modulus and the shear modulus of the aerogel structure itself K_a and N_a. At frequencies high enough that the viscous penetration depth is much smaller than the pore size Biot argued that there are two (fast and slow) longitudinal modes in the composite system. The corresponding expressions for them can be found in [14]. In [15, 16] this theory has been generalized in the case of superfluid-saturated porous media, when all damping processes are neglected.

In the low frequency limit the expression for fast longitudinal sound velocity is given by:

$$u^2_{fast} = \frac{(K_a + 4N_a/3) + (1/\phi)(1 - K_a/K_{\mathrm{SiO_2}})^2 K_{\mathrm{He}}}{\rho_a + \phi\rho_{\mathrm{He}}}, \tag{1}$$

In the normal phase the relationship between the viscous penetration depth and pore size (in ours case the mean separation of SiO_2 strands) is questionable because of complicated disorder structure of aerogel and temperature dependence of liquid ^{4}He viscosity. That is why we have attempted to apply Biot's theory in both limit cases. The observed signal in the present study corresponds to the fast mode. In high frequency limit the corresponding mechanical properties of aerogel and the complex system were evaluated by fitting the experimental data for each aerogel. We could fit the temperature dependence with this theory only if the coupling constant was nearly zero or even negative for each aerogel. As shown by Johnson [14], n is given as square root of the coupling constant so that this becomes nearly zero. According to this, n as evaluated by the Biot theory in high frequency limit had no physical meaning. We should note here that the Biot theory is applicable mainly to a situation in which sound propagation is mainly determined by a solid and fluid provides a small perturbation to the system and works well in ^{4}He filled Vycor [4] and alumina ceramics [5]. The equation which represents the sound velocity [14] has no analytical solution in the case that sound velocity of solid and liquid is close. The aerogel-liquid ^{4}He system is really this case. We concluded that high frequency limit of Biot's theory is not applicable to the aerogel-liquid ^{4}He case.

In the low frequency limit the mechanical properties of aerogel and the complex system can be evaluated by fitting formula (1) to the experimental data for every aerogel. But that the structure of silica aerogel is still under studying and in contrast to usual solids there is no simple relationship between the bulk modulus and the shear modulus. Hence the mentioned fitting procedure includes so many parameters in order to get aerogel sound velocity. That is why we present only a very simple phenomenological model for sound propagation in the normal phase, a more complicated theory has been published in [17]. On the assumption that two different elastic media (the fluid and aerogel) are in parallel, the bulk modulus of the composite medium K_m can be estimated to be $\phi K_{\mathrm{He}} + K_a$ using that of the fluid K_{He} and aerogel K_a. The total density is expressed using the density of aerogel ρ_a, that of liquid helium ρ_{He} and ϕ as $\rho_a + \phi\rho_{\mathrm{He}}$. Then, the sound velocity, u is expressed as

$$u^2 = \frac{\phi K_a + K_{\mathrm{He}}}{\rho_a + \phi\rho_{\mathrm{He}}} = \frac{u_a^2\rho_a + \phi u^2_{\mathrm{He}}\rho_{\mathrm{He}}}{\rho_a + \phi\rho_{\mathrm{He}}}, \tag{2}$$

where u_a, u_{He} is the sound velocity of aerogel and that of liquid helium, respectively.

Equation (2) has been used to estimate the sound velocity of aerogel by fitting the temperature dependence of the observed sound velocity in the normal phase. The aerogel sound velocities are assumed to be constant with temperature, considering other experiments [18, 19]. The best fitting is obtained with the aerogel sound velocities which are listed in Table. These values are consistent with the sound velocity obtained by Gross et al. [10]. We will use these values to analyze the sound mode in the superfluid phase.

For numerical calculations in the superfluid phase, we use two-fluid hydrodynamic equations which take into account the ability of aerogel to move. These equations introduced for this case by McKenna et al. [12] and were investigated partially in [20]. Because of numerous

The porosity ϕ, superfluid transition temperature T_c, and sound velocity of aerogel u_a evaluated from the data in the normal phase

No.	ϕ (%)	T_c (K)	u_a (m/sec)
1	92.6	2.165	256
2	94.0	2.168	212
3	94.8	2.168	181

mathematical errors in the last paper the system of two-fluid hydrodynamic equations for the sound propagation in the liquid ^{4}He filled silica aerogel has been rederived in present study. In the notations of [20] it looks as:

$$p'\left[-\frac{u^2}{u_1^2}+\frac{\rho_s\rho_{na}+\rho_n^2}{\rho\rho_{na}}\right]+T'\left[-u^2\frac{\partial\rho}{\partial T}-\frac{\sigma\rho_s\rho_a}{\rho_{na}}\right]+ \\ +p_a'\frac{\rho_n}{\rho_{na}}=0, \quad (3)$$

$$p'\left[-u^2\frac{1}{\rho}\frac{\partial\rho}{\partial T}-\frac{\sigma\rho_s\rho_a}{\rho_{na}}\right]+ \\ +T'\left[-\frac{u^2}{u_2^2}\frac{\sigma^2\rho\rho_s}{\rho_n}+\frac{\sigma^2\rho_s(\rho+\rho_a)}{\rho_{na}}\right]+ \\ +p_a'\frac{\sigma\rho_s}{\rho_{na}}=0, \quad (4)$$

$$p'\frac{\rho_n\rho_a}{\rho\rho_{na}}+T'\frac{\sigma\rho_s\rho_a}{\rho_{na}}+p_a'[\frac{\rho_a}{\rho_{na}}-\frac{u^2}{u_a^2}]=0. \quad (5)$$

By neglecting $\partial\rho/\partial T$ the secular equation for this system is reduced to the equation from [12]. Excluding p_a' from (5) we arrive to two-equation system similar to the obtained in [20] from which the sound conversion phenomena in superfluid ^{4}He in aerogel can be investigated. The numerical calculations of (3)-(5) give the sound velocity of the two modes – the fast and the slow ones. The velocities of two sound modes calculated for 92.6 and 94.8% as above are shown in Fig.3. The solid and dotted line corresponds to fast and slow mode, respectively. The sound velocity of these modes converges to that of bulk helium and aerogel, since there is neither normal component nor viscous coupling between the two media at low temperatures. The slow mode velocity goes to zero at T_c. It is clearly shown that the experimentally observed sound mode corresponds to the fast mode; these agree well between 1 K and T_c for all aerogels. However, the discrepancy becomes significant below 1 K. The calculated fast mode converges to the greatest sound velocity in the composite system (for 94.0 and 94.8% aerogel – to bulk liquid velocity, for 92.6% aerogel – to aerogel sound velocity). On the other hand, the experimentally observed sound velocities

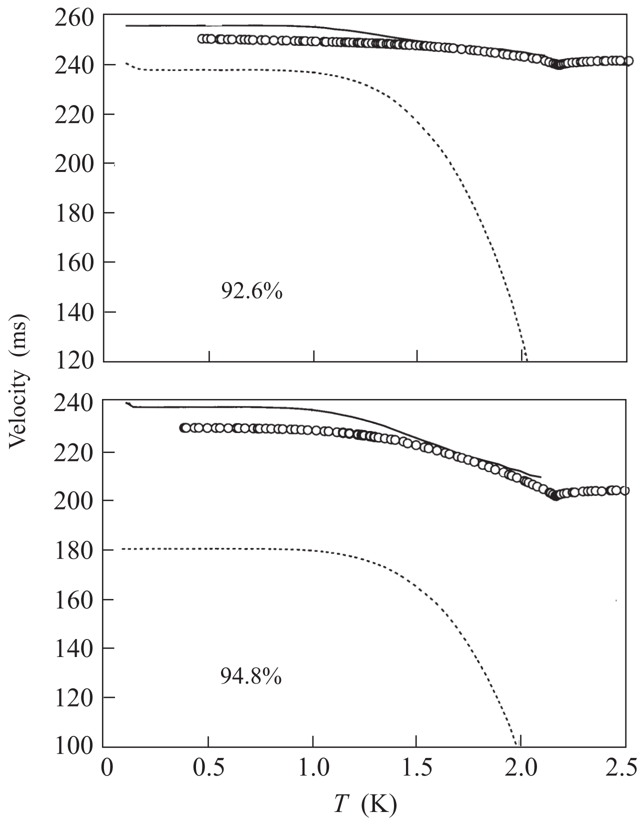

Fig.3. Comparison between the observed sound velocity and that of fast mode calculated by the hydrodynamic equations in the superfluid phase. Circles represent experimental points and lines are theoretically calculated

at low temperature are lower than the calculated values. The porosity dependence of the velocity could not result from the tortuosity as in the case of normal phase. Then, the coupling between liquid and aerogel should be considered apart from viscosity of the normal fluid. We compared mean free path of phonons and rotons and that determined geometrically by aerogel strands. The geometrically limited mean free path becomes shorter than that of phonons and rotons below 1 K. Acoustic phonons are thought to be scattered by aerogel strands and to give rise to the momentum transfer between aerogel and phonons. This means that the simple hydrodynamic theory is not applicable to this temperature range because there is no mechanism of momentum transfer due to there being no viscous fluid. A new theory is necessary in which momentum transfer between aerogel and phonon should be taken into account as in the case of the liquid ^{3}He-aerogel system [21]. We applied the simple idea used in the normal phase to sound velocity at low temperatures because the ultrasound wavelength was long enough to regard the micro structure of aerogel as homogenous and there was only one fluid component. The calculated sound velocities using (2) and aerogel sound velocities listed in Table are slower than experimental ones for all aerogels; this may result from weaker coupling compared to the normal phase. A detailed coupling mechanism seems necessary to fit the sound velocity at low temperatures.

The attenuations in the normal phase were so large that the attenuation variations are experimental artifacts resulting from inadvertent interference between the ultrasound signal and electrical feedthrough from the transducer and amplifier. There is no substantial porosity dependence in the normal phase. At T_c, a sharp absorption peak was observed for each aerogel as was observed in the bulk liquid. In the specific heat measurement [1, 2], two distinct singularities of specific heat were observed. The higher temperature singularities were verified as coincident with the bulk liquid singularities. In our experiment, the absorption peak which corresponds to the T_λ was not observed for every aerogel. In fact, the temperature resolution of our experiment was not as good as the specific heat measurements [1, 2], but was adequate to distinguish between the peaks at T_c and T_λ. In the sound experiment, the attenuation due to the bulk liquid may be obscured by the large attenuation of the aerogel.

The observed constant attenuation in the superfluid phase below 1 K strongly depends on the porosity (or in other words on the density of SiO_2 strands that act as scattering centers) and there is no temperature dependent contribution from phonons and rotons. This behavior qualitatively agrees with the geometrically limited mean free path picture.

The absorption peak around 1 K which is observed in bulk liquid was not observed in the aerogel system. In the bulk liquid, the absorption peak is due to the phonon-roton interaction and the peak appears at the temperature that corresponds to $\omega\tau \sim 1$, where ω is the angular frequency of sound and τ is the relaxation time. The absence of the attenuation peak is considered to be explained as follows. The phonon mean free path increases with decreasing temperature and finally exceeds the geometrically limited mean free path in the aerogel. The relaxation time is limited by the aerogel and cannot satisfy the relation $\omega\tau \sim 1$, so that the attenuation peak is not observed. Similar peak suppression has been observed in the ^{4}He-^{3}He mixture [22], where the existence of ^{3}He excitations means that there will be more collisions, or in other words τ is smaller than that in pure ^{4}He. The peak of the attenuation occurs at lower temperatures compared to pure ^{4}He. Because of the lower temperature, the number of phonons and rotons is lower and the value of the peak attenuation is reduced compared to that of bulk ^{4}He. The same scenario may work in the aerogel-^{4}He system.

In conclusion, we have studied the low temperature acoustic properties of a liquid ^{4}He filled aerogel system for aerogels of various porosities and observed a compressional wave in both the normal and the superfluid phase using 10 MHz ultrasound. It has been found that sound velocity and attenuation are strongly influenced by aerogel. The scaling behavior has been found in the normal phase. In the superfluid phase, the two sound modes are calculated from the hydrodynamic model and the observed sound mode has been shown to correspond to the fast mode. The attenuation peak due to the phonon-roton interaction has not been observed in the present system. The geometrical limited phonon mean free path by aerogel strand seems to play an important role in the acoustic properties at low temperature. More detailed theoretical consideration is in progress [17].

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