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

K. Matsumoto$^1$, Y. Matsuyama$^1$, D. A. Tayurskii$^{1,2}$, and K. Tajiri$^3$

$^1$Department of Physics, Kanazawa University, Kakuma-machi, Kanazawa 920-1192, Japan
$^2$Physics Department, Kazan State University, Kremlevskaya str., 18, Kazan, 420008, Russia
$^3$Ceramics Research Institute, National Institute of Advanced Industrial Science and Technology, Nagoya 463-8560, Japan

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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 (a compressional wave) and second sound (a 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, 7, 8, 9].

Silica aerogels are synthesized via a sol-gel process and hypercritical drying which enable production of tenuous solids with porosity $\phi$ 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-sound-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-sound-like) 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 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 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 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. Sound experiment with aerogel has an advantage to investigate what happens when the relation of sound velocities between fluid and porous media is changed.

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 diameter and 3.0 mm length. The ends of the samples were polished flat and parallel. The sample cylinder was machined into cylinders (7 mm in diameter) which were enclosed in brass shells of 8.5 mm diameter and 3.0 mm length. The ends of the sample in order to easily compare the signal attenuation among aerogels. Aerogels were immersed in liquid 4He 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 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 from the aerogel itself.

Figure 1 and 2 respectively, shows the sound velocity $c$ and attenuation $\alpha$ 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 is 2.165, 2.168, and 2.168 K for 92.6, 94.0, and 94.8 % porosity, respectively. 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 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 $(c(T) - c(4.2K))/(c(2.5K) - c(4.2K))$ and $(c(T) - c(T_c))/(c(0.5K) - c(T_c))$ 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 4He 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, 7, 8, 9], Biot proposed a phenomenological theory of acoustic propagation in porous, fluid filled, macroscopically homogeneous and isotropic media. We applied Biot’s theory in the low frequency limit since the viscous penetration depth is larger than the mean separation of SiO$_2$ strands. Biot argued that there are two (fast and slow) modes in the composite system. The observed signal in the present study corresponds to the fast mode. 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,
ated by the Biot theory had no physical meaning. The Biot theory is applicable 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 and alumina ceramics. The equation which represents the sound velocity 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 Biot’s theory is not applicable to the aerogel-liquid $^4$He case.

Here we present only a very simple phenomenological model for sound propagation in the normal phase, a more complicated theory will be published elsewhere. 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 $\phi K_{He} + K_a$ using that of the fluid $K_{He}$ and aerogel $K_a$. The total density is expressed using the density of aerogel $\rho_a$, that of liquid helium $\rho_{He}$ and $\phi$ as $\rho_a + \phi \rho_{He}$. Then, the sound velocity, $c$ is expressed as

$$c^2 = \frac{\phi K_a + K_{He}}{\rho_a + \phi \rho_{He}} = \frac{c_a^2 \rho_a + \phi c_{He}^2 \rho_{He}}{\rho_a + \phi \rho_{He}},$$

(1)

where $c_a$, $c_{He}$ is the sound velocity of aerogel and that of liquid helium, respectively.

Equation (1) 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. The best fitting is obtained with the aerogel sound velocity, 256, 212, and 181 m/s for 92.6, 94.0 and 94.8 % porosity, respectively. These values are consistent with the sound velocity obtained by Gross et al. 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. The velocities of three sound modes calculated for 92.6 and 94.8 % as above are shown in Fig. 3. The solid, broken and dotted line corresponds to fast, intermediate and slow mode, respectively. The sound velocity of these modes converges to that of bulk helium and aerogel to get more detailed pictures about this mode as well as two others. 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 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 1K. 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. 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 microstructure of aerogel as homogenous and there was only one fluid component. The calculated sound velocities using Eq. (1) and aerogel sound velocities obtained by fitting in the normal phase 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.

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 equations points and lines are theoretically calculated.
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 feed-through 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, 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 $T_\lambda$ was not observed for every aerogel. In fact, the temperature resolution of our experiment was not as good as the specific heat measurements, but was adequate to distinguish between the peaks at $T_c$ and $T_\lambda$. 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 general tendency of our results (attenuation is large in the normal phase and decreases with temperature in the superfluid phase) is similar to those of alumina ceramics.

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 $\omega$ is the angular frequency of sound and $\tau$ is the relaxation time. The absence of the attenuation peak can be qualitatively 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, where the existence of $^3$He excitations means that there will be more collisions, or in other words $\tau$ 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 apply 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 superfluid phases 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 three 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.

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