Ultrasound propagation in dense aerogels filled with liquid $^4$He

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Abstract. Longitudinal ultrasound propagation was studied in dense aerogels filled with liquid $^4$He. Sound velocity and attenuation were measured at the frequency of 6 MHz in both normal and superfluid phases. Pressure dependence of velocity and attenuation were also studied. Studied aerogels had porosities about 85%. They had two different types of structure, tangled strand structure and aggregated particles structure. The pore size distributions were narrow. Reduction of superfluid transition temperature mainly depended on not porosity but mean pore size. The structure of gel played an important role in sound velocity and attenuation.

1. Introduction

Liquid $^4$He confined into various nano-porous media, such as aerogel, Vycor glass etc., attracts attention in last decades [1-3]. $^4$He in nano-porous media is an ideal example of Bose system with disorder. A number of features have been revealed such as a suppression of superfluidity by disorder and the alteration of $P-T$ phase diagram of $^4$He confined to pores [1].

Acoustic property of superfluid helium in aerogel is of interest because it has a normal component that plays the role of viscous fluid and simultaneously a superfluid component with zero viscosity. The normal fluid in high-porous media like aerogel is locked to the substrate by viscosity and drags the substrate in response to liquid motion. So, one needs to consider the normal fluid motion taking into account a restoring force as well as addition of substrate mass [2, 3]. We have reported acoustic properties of liquid helium in aerogel using longitudinal ultrasound and the different behavior from rigid porous materials [4-6]. In the previous work, an anomalous attenuation maximum was observed in superfluid phase with 97% open aerogel at the frequency of 10 MHz [6]. In order to elucidate the acoustic properties in porous materials, the obvious way is to make measurements by varying the sound frequency and using materials that have different pore structure. In the present work, we have done acoustic experiment of liquid $^4$He in gels with different characteristic structures.

Silica aerogels are made by sol-gel process and dried in order to obtain a high porosity glass. Aerogels with controlled pore size and structure can be synthesized by our research group. Our gels have porosity about 85% that is smaller than previous works [2, 4-6]. They had two different types of structure, tangled strand structure and aggregated particles structure. The mechanical strength also differs so much among our aerogels. It is of great interest to understand the way that the characteristic structure of gels affects the nature of the wave propagation. We have measured sound velocity and attenuation at the frequency of 6 MHz and found that gel structure as well as porosity influenced sound velocity and attenuation. The suppression of superfluid transition was mainly determined by the mean pore size.

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2. Experiment

Three aerogels with 83.1, 85.2, and 87.2% porosities have been made by Kyoto University group. N₂ adsorption measurements indicated that the pore size distributions were narrow and the typical pore diameters are presented in table 1. The structure of gel was observed using SEM. It is confirmed that Gel A and B had tangled strand structure and Gel C had aggregated particles structure. Generally speaking aerogels are brittle and have low mechanical strength. However, gels with tangled strand structure have shown to have high mechanical strength and elasticity [7]. The gels were machined into cylinders of 7 mm in diameter and about 2.3 mm in length for the sound experiment.

There are two sound modes in superfluid ¹He in aerogel, a slow critical mode, and a fast one which resembles first sound. The fast mode was observed in the present study. The sound cell in which the aerogel was sandwiched between two LiNbO₃ transducers with resonance frequency of 6 MHz was cooled using a ³He cryostat. The ultrasonic measurements were made using a standard pulse transmission and a phase sensitive detection technique described elsewhere [4, 5]. Liquid ¹He was filled in the normal phase since it has small surface tension. No damage due to surface tension of liquid ¹He in aerogel was checked when we changed samples. The resolution of sound velocity was about 10⁻⁵. Absolute velocity was obtained by the flight-time of the signal, so that the absolute value was determined within about 2%. Relative attenuation change was observed with a resolution of about 0.01 dB.

| Porosity (%) | Typical pore size (nm) | Structure         |
|--------------|------------------------|-------------------|
| Gel A        | 83.1                   | 70.5              | tangled strand   |
| Gel B        | 85.2                   | 50.5              | tangled strand   |
| Gel C        | 87.2                   | 32.5              | aggregated particles |

3. Results and discussion

3.1. Superfluid transition temperature

The sharp critical attenuation peak and dip in sound velocity were observed at the superfluid transition in bulk liquid. Similar dip and attenuation peak are observed in our gels. Insets of figure 1 show these attenuation peaks at the transition. If superfluid transition temperature Tc of liquid ¹He is suppressed by impurity scattering due to SiO₂ of aerogel, suppression is considered to increase with decreasing porosity. However, the suppression of Tc in Gel C was about 12 mK and the largest among three gels. The suppression of Tc increased with decreasing pore size in this study. The suppression magnitude of Tc depended on pore size and not on porosity. When Tc is compared with the data relating Tc to pore size reported by Wada et al. [8], they are in reasonable agreement. Tc suppression from bulk had weak pressure dependence.

3.2. Sound velocity

Temperature variations of sound velocity in gels of this study are similar to that in more than 90% porous aerogels [4]. Sound velocity had a dip at the superfluid transition in all samples. Figure 2 shows the ultrasound velocity of liquid ¹He in the studied aerogels at 2.5 and 0.8 K as a function of pressure. For comparison, those of bulk helium obtained in a different run were plotted as well. Velocity increase with pressure is ascribed to the velocity increase of the liquid helium. Velocity difference among gels reflects the influence of mechanical properties of aerogels. Bulk SiO₂ has much higher speed of sound than liquid ¹He. Sound velocity is considered to decrease with increasing porosity as shown in figure 1. So, porosity dependence of sound velocity looks reasonable at a glance. Sound velocity of Gel A and B reasonably agrees with that of dilute aerogels, considering porosity difference of sound velocity [5,9]. However, sound velocity in Gel C was almost the same as that in 97% porous aerogel obtained in former study [5], even though density of SiO₂ in Gel C is four times larger than that in 97% porous aerogel. It is necessary to consider other reasons of low sound velocity in Gel C. We observed that mechanical properties of aerogel depend on gel structure [7]. Gels with tangled strand structure have higher mechanical strength than those with aggregated particle structure.
due to structure difference. Gel structure is considered as one of the origins of low sound velocity in Gel C.

3.3. Sound attenuation

Sound attenuation has a sharp peak at the superfluid transition temperature for all aerogels. We have observed large broad attenuation peak in superfluid phase for Gel B as shown in figure 1. Temperature variation of the large attenuation peak is similar to that observed in 97% porous aerogel [6]. However, no such large attenuation peak in superfluid phase was observed for Gel A and C. In the case of Gel A and C, only sharp peaks at $T_c$ were observed.

Large attenuation peaks were observed only in 85% (Gel B) and 97% porous aerogels. It was clear that porosity does not play crucial role in this phenomenon. From this point of view, aerogels structure and their mechanical strength seems to be important to explain the presence or absence of the large attenuation peak in superfluid phase.

We discuss coupling between gel and liquid $^4$He. Normal component of liquid $^4$He is coupled to aerogel by viscosity. Viscous penetration depth within which fluid viscously couples to the solid wall is estimated to be close to typical pore size of our gels at our experimental sound frequency. The pore size of Gel A is thought to be close to that of 97% porous aerogel because the suppression of superfluid transition was close. However, no large attenuation peak was observed in the superfluid phase for Gel A, though the coupling between aerogel and liquid is seems to be similar in both gels. In comparison between Gel A, B and 97% porous gel, sound velocity of Gel A and B is much higher than that of 97% porous aerogel. Sound velocity of Gel C is close to that of 97% porous aerogel as mentioned in the former section. So, mechanical properties of Gel C and 97% porous aerogel are thought to be similar from the point of view of elastic constant. However, no large attenuation peak was observed in superfluid phase for Gel C. From these results, anomalous attenuation in superfluid phase could not be attributed to only pore size or sound velocity of aerogel. Aerogel structure is considered to play an important role in sound propagation.

In order to explain the existence of the large attenuation peak below $T_c$, a theoretical model of 2D-3D roton transformations was proposed [10]. The calculated temperature dependence of sound attenuation shows a clear peak what supports the idea about roton transformations. The possibility of the sound modes conversion between first, second sound in superfluid and aerogel sound mode was discussed in this composite system [7]. However, a full explanation hasn’t be given yet. It is necessary to do experiments with various aerogels that have controlled pore size, structure, and mechanical strength. Frequency dependence should be studied too.

![Figure 1. Sound attenuation of liquid $^4$He in 85.2% open aerogel (Gel B) at 5, 15 and 25 bar. Insets show sharp attenuation peaks at the superfluid transition at 5 and 25 bar.](image)
Figure 2. Sound velocity of liquid $^4$He in three aerogels in normal phase (left) and superfluid phase (right). Those of bulk liquid are also shown as open circles for comparison.

4. Summary
Longitudinal ultrasound propagation was studied in about 85% porous aerogels filled with liquid $^4$He. Sound velocity and attenuation were measured at the frequency of 6 MHz in both normal and superfluid phases. Superfluid suppression mainly depended on not porosity but mean pore size. Large broad attenuation peak in superfluid phase was observed for 85% aerogel. This peak was similar to that was observed for 97% open aerogel. The peak could not be attributed to only pore size or sound velocity. Gel structure seems to play important role in sound attenuation.

5. References
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Acknowledgments
Financial support was provided by a Grant-in-Aid for Education, Culture, Sports, Science and Technology of Japan.