ON THE SOUND PROPAGATION IN SILICA AEROGELS FILLED IN BY NORMAL AND SUPERFLUID HELIUM

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О РАСПРОСТРАНЕНИИ ЗВУКА В АЭРОГЕЛЯХ КРЕМНЕЗЕМА, НАПОЛНЕННЫХ НОРМАЛЬНЫМ И СВЕРХТЕКУЧИМ ГЕЛИЕМ

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Magnetic Resonance in Solids
Electronic Journal

Volume **6**, *No.* **1**,
pages **213-220**, **2004**

http://mrsej.ksu.ru

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Earlier obtained experimental data concerning sound propagation in silica aerogels filled in by liquid ^{4}He are theoretically analyzed for normal as well as superfluid phase. The simple phenomenological model is proposed for normal phase. The data for superfluid phase are described in a rather good manner by the hydrodynamic theory with taking into account the clamping of normal component by silica strands. It is shown also that at temperatures below 1 K one needs a new hydrodynamic theory in which momentum transfer between aerogel and phonons in liquid helium should be taken into account

Introduction

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 a genuine phase transition. The transition temperature in aerogel T_c has been suppressed with decreasing aerogel porosity.

Acoustic experiments are important to understand porous media filled with fluid. Use of liquid ^{4}He offers unique advantages due to the existence of the superfluid phase with more than one sound mode. For the bulk fluid, there are first sound in which the two fluid components move together and propagates as a compressional wave and second sound in which the two fluid components move in opposite direction and propagates as a temperature wave [3]. In the porous media where the normal component is clamped by its viscosity and only the superfluid component can move, the fourth sound propagates and can be used to determine the superfluid fraction.

The high-porosity aerogels are so soft that the aerogel matrix clamped with the normal fluid is caused to move by the pressure and temperature gradients unlike other porous media. This results in sound modes intermediate between first and fourth sound [4] and second-like mode [5]. McKenna et al. [5] calculated longitudinal sound velocity for the two modes using the 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.

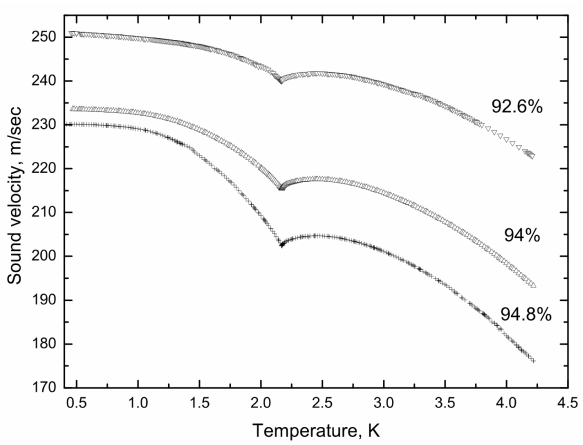

Fig. 1. Experimentally measured in [12] longitudinal ultrasound velocities in silica aerogels with various porosities filled in by liquid ^{4}He

Longitudinal and transverse ultrasound velocities have been measured in ^{4}He filled Vycor glass [6]. Warner and Beamish [7] have studied transverse sound (4 to 31 MHz) velocity and attenuation in alumina (Al_2O_3) ceramics with porosity changed from 44 to 92 %. 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 [8-11]. The alumina samples were made by differently (slip-casting, sinter and sol-gel process) and porosity changed from 44% to 92%. They could make measurement in both the low and high frequency regimes by changing the ultrasound frequency, temperature from normal to superfluid phase and using ceramics with various porosities. They argued that the Biot model [8-11] could quantitatively elucidate the velocity and attenuation in all cases.

In [12] we have observed sound signal of 10 MHz longitudinal ultrasound with three different porosity aerogels from 0.5 K to 4.2 K, and measured the sound velocity and attenuation in order to study sound propagation in liquid ^{4}He filled aerogel system both in the normal and superfluid phase. The viscous penetration depth of liquid ^{4}He at 10 MHz is estimated to be longer than the typical SiO_2 strand distance. Then, 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 happens if the relation of sound velocities between aerogel and fluid is counterchanged. For reader' convenience the obtained in [12] experimental results are shown in Fig. 1. The present paper is devoted to the theoretical discussion obtained in [12] experimental results and to the comparison with the existing theoretical approaches to sound propagation in porous media filled in liquid ^{4}He.

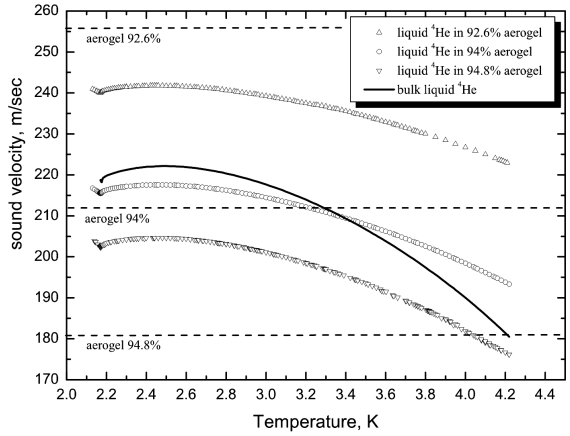

Fig. 2. The temperature dependence of the sound velocity in different aerogels filled in by liquid ^{4}He in normal phase. The similar dependence for bulk liquid ^{4}He is shown also as well as sound velocities in aerogels obtained from these experimental results (see text for details)

Normal phase

The temperature dependence of the sound velocity is similar to that of bulk for every aerogel (see Fig. 2, where experimental data as well as data for bulk liquid ^{4}He and the obtained aerogel sound velocities are shown, see discussion below). The absolute value greatly depends on the aerogel 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. Then, tortuosity

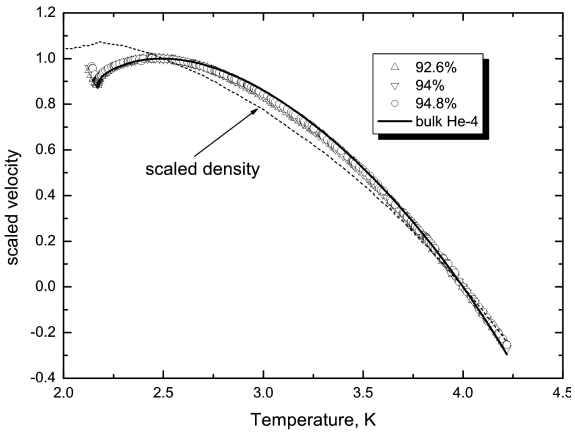

Fig. 3. The scaled temperature dependencies of sound velocities in aerogels filled in liquid ^{4}He and in bulk liquid ^{4}He. Also the scaled density of liquid ^{4}He is shown

cannot explain this behavior. The similarity of the temperature variation brings scaled behavior to mind. The scaled temperature variations in the normal phase for each aerogel and also bulk liquid coincide with each other as shown in Fig. 3. This means the temperature variation is determined mainly by bulk liquid.

It is useful to compare our data with the longitudinal sound velocity in ^{4}He filled Vycor [6]. In the normal phase, the sound velocity in Vycor system was almost constant, reflecting the constant sound velocity of Vycor glass. Contrary to aerogel case, Vycor glass determined the sound velocity of the composite system. In the normal phase, we can compare the sound propagation in aerogel with other porous media, which is usually explained by Biot's theory [8-11].

In a series of papers [8-11], Biot proposed a phenomenological theory of acoustic propagation in porous, fluid filled, macroscopically homogeneous and isotropic media. The parameters of that theory are the tortuosity α, the porosity ϕ (fluid volume fraction), the density of liquid ρ_f and solid ρ_{sol}, the bulk modulus of fluid K_f, the bulk modulus of solid K_s, the bulk modulus and the shear modulus of the skeletal frame K_b and N. At frequencies high enough that the viscous skin depth is much smaller than the pore size Biot argued that there are two (fast and slow) longitudinal modes in the composite system:

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$$v_{fast,slow}^2 = \frac{1}{2\left(\rho_{11}\rho_{22}-\rho_{12}^2\right)}\left[P\rho_{22}+R\rho_{11}-2\rho_{12}Q \pm - \right.$$
$$\left. \pm\sqrt{\left(P\rho_{22}+R\rho_{11}-2\rho_{12}Q\right)^2-4\left(\rho_{11}\rho_{22}-\rho_{12}^2\right)\left(PR-Q^2\right)}\right] \quad (1)$$

where coefficients P,Q,R can be related to above parameters:

$$P = \frac{(1-\phi)\left[1-\phi-K_b/K_s\right]K_s+\phi\left(K_s/K_f\right)K_b}{1-\phi-K_b/K_f+\phi\left(K_s/K_f\right)}+\frac{4}{3}N \quad (2)$$

$$Q = \frac{\left[1-\phi-K_b/K_s\right]\phi K_s}{1-\phi-K_b/K_s+\phi K_s/K_f}, \quad (3)$$

$$R = \frac{\phi^2 K_s}{1-\phi-K_b/K_s+\phi K_s/K_f}, \quad (4)$$

and the density terms are:

$$\rho_{11}+\rho_{12} = (1-\phi)\rho_{sol}, \quad (5)$$

$$\rho_{22}+\rho_{12} = \phi\rho_f, \quad (6)$$

$$\rho_{12} = (1-\alpha)\phi\rho_f. \quad (7)$$

In [13] 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 [14,15] 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

$$v_l^2 = \frac{\left(K_b+4N/3\right)+\left(1/\phi\right)\left(1-K_b/K_s\right)^2 K_f}{\rho_{scel}+\phi\rho_f} \quad (8)$$

(ρ_{scel} is the density of solid skeletal frame, below this value will be changed by the density of aerogel) and the slow wave doesn't propagate. In the case of aerogel the bulk modulus of SiO_2 is much greater than the modulus of aerogel $K_s \gg K_b$, and the expression (8) can be simplified.

To explain the data obtained in [12] for the normal phase we can apply the Biot's theory in low frequency limit since the viscous penetration depth is larger than the mean separation of SiO_2. So the mechanical properties of aerogel and the

complex system can be evaluated by fitting formula (8) to the experimental data for every aerogel. But we should mention here, 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 propose some phenomenological model for sound propagation in the normal phase for the composite system in order to estimate the sound velocity in the aerogel. Actually there are three "channels" for the sound propagation in the system under consideration and they work in parallel way. The first channel is determined only by the elastic properties of liquid ^{4}He and corresponds to the case when the sound waves propagate only in the liquid confined between SiO_2 strands. The second channel corresponds to sound propagation through the interconnecting net of SiO_2 strands and is determined by the elastic properties of aerogel. Finally the third channel can be represented as the "mixed" channel when there are continuous transformations of the sound waves from waves in liquid ^{4}He to the waves in SiO_2 strands and back and this channel is determined by the elastic properties of liquid ^{4}He as well as of bulk SiO_2. Keeping mind the analogy with the consequent series of spring the bulk modulus for this “mixed” channel can be write down as:

$$K_{mix} = \frac{\phi K_{\mathrm{He}} \cdot K_{\mathrm{SiO_2}}}{\phi K_{\mathrm{He}} + K_{\mathrm{SiO_2}}}, \tag{9}$$

where K_{He} and $K_{\mathrm{SiO_2}}$ stand for bulk modulus of liquid ^{4}He and silica strands. We note here that usual relationship between bulk modulus K and sound velocity u for an elastic media with density ρ

$$K = \rho u^2 \tag{10}$$

and much more higher values of sound velocity and density of silica in comparison with liquid ^{4}He give us the following simplification for K_{mix}:

$$K_{mix} = \frac{\phi\rho_{\mathrm{He}} C_{\mathrm{He}}^2 \cdot \rho_{\mathrm{SiO_2}} C_{\mathrm{SiO_2}}^2}{\phi\rho_{\mathrm{He}} C_{\mathrm{He}}^2 + \rho_{\mathrm{SiO_2}} C_{\mathrm{SiO_2}}^2} \approx \rho_{\mathrm{He}} C_{\mathrm{He}}^2, \tag{11}$$

The relative weight of the third channel α with respect to the relative weight of the first and the second channel can be considered as the fitting parameter and might depend on temperature. The temperature dependence of this parameter follows from the temperature dependence of the liquid ^{4}He density (the “effectiveness” of the sound transformation on the boundary liquid ^{4}He - SiO_2 surface is determined by the densities difference). The bulk modulus of the composite medium can be calculated with the bulk modulus of the fluid (K_{He}) and that of aerogel (K_a) and the “mixed” channel and assuming three different elastic media in parallel, resulting in

$$K = (1-\alpha)(\phi K_{\mathrm{He}} + K_a) + \alpha K_{mix}. \tag{12}$$

The total density is expressed using 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

$$u^2 = \frac{\rho_{\mathrm{He}} u_{\mathrm{He}}^2 (\phi + \alpha(1-\phi)) + \rho_a u_a^2 (1-\alpha)}{\phi\rho_{\mathrm{He}} + \rho_a}. \tag{13}$$

Note that this equation looks a rather similar to (8). In the simplified form (at $\alpha = 0$) the given equation has been used in [12] to obtain aerogel sound velocity u_a from the experimental data under assumption that the aerogel sound velocities are constant with temperature, considering other experiments [16,17]. The obtained aerogel sound velocities for three aerogels listed below are consistent with the density dependence of aerogel sound velocity obtained by Gross [18]:

Porosity	u_a, m/sec
92.6%	256
94%	212
94.8%	182

We will use these values to analyze the sound mode in the superfluid phase.

Strictly speaking there are two fitting parameters in Eq. (13): u_a and $\alpha(T)$. The simplified case ($\alpha = 0$) can be considered as the first approximation. After that as far as the temperature dependence of u is determined mainly by liquid helium (see Fig. 3) we can neglect by temperature dependence of $\alpha(T)$ and get the main term in the expansion of $\alpha(T)$ with respect to T. So, in principle, the values aerogel sound velocity and parameter α can be found with the desired accuracy.

Superfluid phase

The propagation of sound in superfluid phase is usually described by linearizing the two-fluid hydrodynamic equations. Following Atkins [19] we can write down the continuity equation:

$$\frac{\partial\rho}{\partial t} = -\nabla\cdot\mathbf{j} = -\nabla\left(\rho_n \mathbf{v}_n + \rho_s \mathbf{v}_s\right) \tag{14}$$

and the equation for the entropy conservation

$$\frac{\partial(\rho\sigma)}{\partial t}+\nabla\cdot(\rho\sigma\mathbf{v}_n)=0. \tag{15}$$

In equation of motion we omit terms quadratic in the velocities

$$\rho_n\frac{\partial\mathbf{v}_n}{\partial t}=-\frac{\rho_n}{\rho}\nabla p-\rho_s\sigma\nabla T \tag{16}$$

$$\rho_s\frac{\partial\mathbf{v}_s}{\partial t}=-\frac{\rho_s}{\rho}\nabla p+\rho_s\sigma\nabla T. \tag{17}$$

Adding last two equations together we have

$$\frac{\partial\mathbf{j}}{\partial t}=-\nabla p, \tag{18}$$

where further second quadratic terms such as $\mathbf{v}_s(\partial\rho_s/\partial t)$ have been ignored. In combination, (14) and (18) yield the first linearizing hydrodynamic equation

$$\frac{\partial^2\rho}{\partial t^2}=\Delta p. \tag{19a}$$

To obtain the second one can eliminate ∇p between (17) and (18)

$$\rho_n\frac{\partial}{\partial t}(\mathbf{v}_n-\mathbf{v}_s)=-\rho\sigma\nabla T \tag{20}$$

which confirms that a temperature gradient produces relative motion between two fluids. Finally combining (14), (15) and (20) gives the second of the two equations we need:

$$\frac{\partial^2\sigma}{\partial t^2}=\frac{\rho_s}{\rho_n}\sigma^2\Delta T. \tag{19b}$$

After substituting $p=p_0+p'$ and $T=T_0+T'$ (where p_0,T_0 are the equilibrium values and p',T' are the perturbations due to sound wave) we get

$$\frac{\partial\rho}{\partial p}\frac{\partial^2 p'}{\partial t^2}-\Delta p'+\frac{\partial\rho}{\partial T}\frac{\partial^2 T'}{\partial t^2}=0 \tag{21a}$$

$$\frac{\partial\sigma}{\partial p}\frac{\partial^2 p'}{\partial t^2}+\frac{\partial\sigma}{\partial T}\frac{\partial^2 T'}{\partial t^2}-\frac{\sigma^2\rho_s}{\rho_n}\Delta T'=0. \tag{21b}$$

If we look for plane-wave solutions of these equations, p' and T' are proportional to $\exp(-i\omega(t-x/u))$, where $u$is the velocity in the x-direction. So we can rewrite equations (21a) and (21b) as:

$$\left(\frac{\partial\rho}{\partial p}u^2-1\right)p'+\frac{\partial\rho}{\partial T}u^2T'=0 \tag{22a}$$

$$\frac{\partial\sigma}{\partial p}u^2p'+\left(\frac{\partial\sigma}{\partial T}u^2-\frac{\sigma^2\rho_s}{\rho_n}\right)T'=0. \tag{22b}$$

Neglecting the anomalous small for superfluid helium thermal expansion coefficient $\partial\rho/\partial t$ we obtain two velocities for sound propagation from the secular equation of system (22):

$$u_1=\sqrt{\left(\frac{\partial p}{\partial\rho}\right)_\sigma}\quad\text{(the first sound)}, \tag{23}$$

$$u_2=\sqrt{\frac{\sigma^2\rho_s}{\rho_n\left(\frac{\partial\sigma}{\partial T}\right)}}\quad\text{(the second sound)}. \tag{24}$$

It is known that for liquid ^{4}He inside a porous media where the normal component is immobilized and only the superfluid is in motion the so-called fourth sound can be observed [20] and its velocity is given by:

$$u_4^2=\frac{\rho_s}{\rho}u_1^2+\frac{\rho_n}{\rho}u_2^2. \tag{25}$$

It is assumed here that the porous media is absolute rigid and doesn't take place in the oscillate motion of liquid. The silica aerogel represents an "easy carried along" media, so the normal component locked by its viscosity silica strands and aerogel matrix move together with a velocity $\mathbf{v}_n$. The modified hydrodynamic equations were introduced for this case by

McKenna *et al.* [5] and were investigated partially in [21]. Because of numerous mathematical errors in the last paper we solve here the system of hydrodynamic equations for the sound propagation in silica aerogel filled in by liquid ^{4}He. The starting hydrodynamic equations are:

$$\frac{\partial \rho}{\partial t} + \nabla\left(\rho_n \mathbf{v}_n + \rho_s \mathbf{v}_s\right) = 0, \tag{26}$$

$$\frac{\partial\left(\rho\sigma\right)}{\partial t} + \nabla\cdot\left(\rho\sigma\mathbf{v}_n\right) = 0, \tag{27}$$

$$\rho_s \frac{\partial \mathbf{v}_s}{\partial t} = -\frac{\rho_s}{\rho}\nabla p + \rho_s \sigma \nabla T, \tag{28}$$

$$\rho_{na} \frac{\partial \mathbf{v}_n}{\partial t} = -\frac{\rho_n}{\rho}\nabla p - \nabla p_a - \rho_s \sigma \nabla T, \tag{29}$$

$$\frac{\partial \rho_a}{\partial t} + \nabla\left(\rho_a \mathbf{v}_n\right) = 0. \tag{30}$$

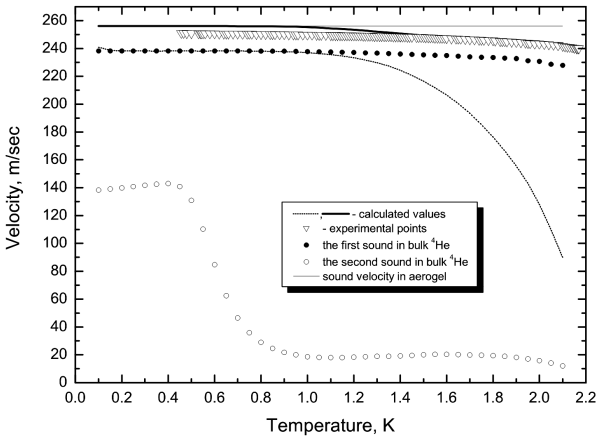

Fig. 4. The calculated sound velocities for two modes from equations (31) for 92.6% aerogel filled in by superfluid ^{4}He and experimental points from The temperatures dependencies of the first and second sound for bulk ^{4}He are shown also

These differ from the bulk superfluid ^{4}He equations (14)-(17) by the replacement $\rho_n \to \rho_n + \rho_a = \rho_{na}$ on the left-hand side of Eq. (29) and the additional restoring force p_a due to the aerogel. Performing the same calculations as above and introducing the perturbation p_a' due to sound plane-wave we obtain finally:

$$p'\left[-\frac{u^2}{u_1^2} + \frac{\rho_s\rho_{na} + \rho_n^2}{\rho\rho_{na}}\right] + T'\left[-u^2\frac{\partial\rho}{\partial T} - \frac{\sigma\rho_s\rho_a}{\rho_{na}}\right] + p_a'\frac{\rho_n}{\rho_{na}} = 0, \tag{31a}$$

$$p'\left[-u^2\frac{1}{\rho}\frac{\partial\rho}{\partial T} - \frac{\sigma\rho_s\rho_a}{\rho\rho_{na}}\right] + T'\left[-\frac{u^2}{u_2^2}\frac{\sigma^2\rho\rho_s}{\rho_n} + \frac{\sigma^2\rho_s(\rho+\rho_a)}{\rho_{na}}\right] + p_a'\frac{\sigma\rho_s}{\rho_{na}} = 0, \tag{31b}$$

$$p'\frac{\rho_n\rho_a}{\rho\rho_{na}} + T'\frac{\sigma\rho_s\rho_a}{\rho_{na}} + p_a'\left[\frac{\rho_a}{\rho_{na}} - \frac{u^2}{u_a^2}\right] = 0. \tag{31c}$$

By neglecting $\partial\rho/\partial t$ the secular equation for the system (31) is reduced to the equation from [5]. Excluding p_a' from (31c) we arrive to two-equation system similar to the obtained in [21] from which the sound conversion phenomena in superfluid ^{4}He in aerogel can be investigated. The numerical calculations of (31) give the sound velocity of the two modes – the fast and the slow ones (solid and dotted lines in Fig. 4-6, respectively). The temperature dependence between about

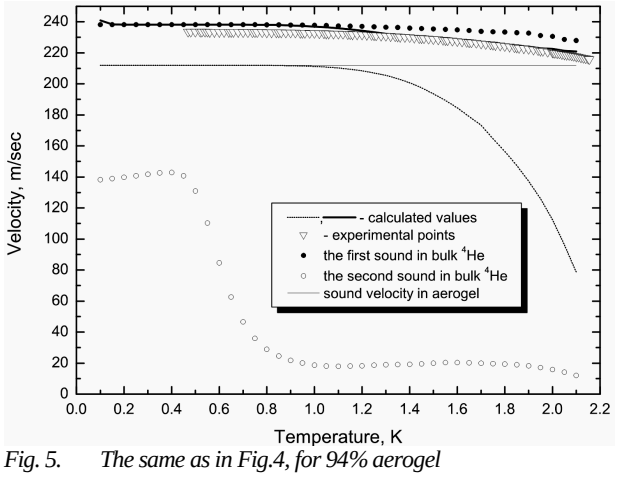

Fig. 5. The same as in Fig.4, for 94% aerogel

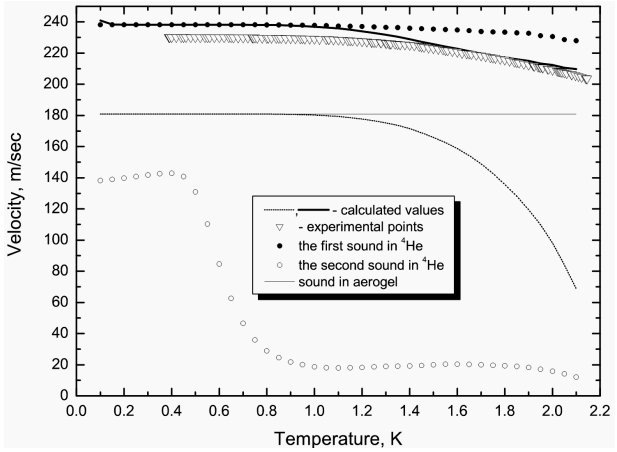

Fig. 6. The same as in Fig.4, for 94.8% aerogel

1 K and T_c is elucidated from the calculation using the aerogel sound velocity estimated from the analysis in the normal phase. The sound velocities of these modes converge to that of bulk helium and aerogel, since there is neither normal component nor viscous coupling in between at low temperatures. The slow mode velocity goes to zero at T_c. It is clearly shown that the experimentally observed

sound mode in [12] corresponds to the fast mode. They agree well between 1 K and T_c for all aerogels. However, the discrepancy becomes significant below 1 K. The calculated fast mode converged to the bulk liquid sound velocity in the case of 94.0 and 94.8 % aerogel since aerogel sound velocity is slower than liquid. The sound velocity of the fast mode for 92.6 % aerogel becomes higher than bulk liquid at low

temperatures which results from the higher sound velocity of aerogel than bulk liquid. On the other hand, the experimentally observed sound velocities at low temperature are lower than the calculated values. The porosity dependence of the velocity can not be determined by 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 the mean free path of phonon and roton and that determined geometrically by aerogel strands. The geometrically limited mean free path becomes shorter than that by phonon and roton below 1 K. Acoustic phonons are thought to be scattered by aerogel strands and 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 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 liquid ^{3}He -aerogel system [22].

Conclusions

In conclusion, we have studied theoretically the low temperature acoustic properties of liquid ^{4}He filled aerogel system for various porosity aerogels and interpreted the observed in [12] compressional wave in both the normal and the superfluid phase using 10 MHz ultrasound. It has been found that sound velocity is strongly influenced by aerogel. The temperature dependence of the sound velocity is similar to that of bulk liquid but the absolute value is varied by aerogel. The scaling behavior has been shown in the normal phase. The simple phenomenological model is proposed for 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 temperature variation between 1 K and T_c is explained by this model.

Acknowledgement

This work was partially support by the grants from “Universities of Russia”. The authors would like to express their appreciation to Prof. H. Suzuki and Dr. S. Abe for numerous useful discussions.

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