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Ultrasonic investigation of phonon localization in a disordered three-dimensional "mesoglass"

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Abstract. One of the long standing questions in phonon physics has been whether or not the Anderson localization of acoustic phonons can be demonstrated unambiguously in disordered materials. In this paper, this question is addressed by reporting signatures of the localization of ultrasonic waves in a "mesoglass" made from a disordered three-dimensional network of aluminum beads. In the upper part of the intermediate frequency regime, which extends over the range of frequencies where the acoustic phonon wavelength is comparable with the sizes of the pores and beads, the intensity distributions of the speckle patterns due to strong multiple scattering show clear departures from Rayleigh statistics, with a variance that increases with frequency. This intensity distribution can be fitted with a stretched exponential, consistent with recent predictions for localization. In this frequency range, the time-of-flight profile of the transmitted intensity exhibits a non-exponential decay, which may be construed as a slowing down of the phonon diffusion coefficient with propagation time. These results are interpreted using recent theoretical predictions based on the self-consistent theory of the dynamics of localization, showing that our experimental data are consistent with the localization of acoustic waves in this mesoglass, and further elucidating their behaviour.

1. Introduction

For the last 20 years, there has been continuing interest in the possibility that acoustic phonons may become localized due to very strong scattering in sufficiently disordered materials. In atomic glasses, this question has been investigated in the context of the universal thermal conductivity plateau near 10 K, and it is now generally accepted on the basis of numerical simulations that the majority of the phonon modes in glasses are diffusive, with only a relatively small number of localized modes near the high frequency edge of the acoustic spectrum [1, 2]. Mesoscopic materials, which have internal structures at length scales between atomic dimensions and bulk, form another important class of disordered systems in which very strong scattering may occur, with potentially dramatic changes to phonon transport. Examples of such materials include packed powders, aerogels, sintered particle networks and foams [3-7]. The range of internal length scales in such mesostructures can vary enormously, from nanometers to millimeters, so that access to the intermediate frequency regime, in which strong scattering is expected, can be tuned to facilitate measurements of the phonon properties.

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As a result, previous Brillouin and ultrasonic experiments have shown that the Ioffe-Regel criterion for localization, namely that the product of wave vector k and scattering mean free path l_s be less than 1, can be realized in both aerogels and sintered bead networks [4, 6]. However, this condition alone does not appear to be sufficient to ensure that the modes are localized [1, 6].

Since the Anderson localization of acoustic phonons is essentially a wave phenomenon, ultrasonic experiments are well suited to look for experimental evidence for such behaviour. In this paper, we capitalize on the ability of ultrasonic measurements to study both the anomalous fluctuations and time dependent transmission of the intensity to investigate these localization signatures. These measurements show unambiguous evidence that the acoustic phonon modes of a mesoscopic glassy system are localized near the upper end of the intermediate frequency regime.

2. Basic properties of the mesoglass samples

Strongly scattering samples were made from a random loose packing of aluminum beads, which were lightly fused together while preserving their spherical shape using a braising technique. We refer to this system as a “mesoglass”, in which the disordered three-dimensional network of beads, joined by elastically weak necks, forms a glassy material made from mesoscopic particles rather than atoms. The beads were monodisperse, with a radius of 2.05 mm. The radius was chosen to be approximately 40 times larger than the glass bead radius in the sintered networks studied in [6], allowing the upper end of the intermediate frequency regime to be investigated in the low MHz ultrasonic frequency range. The samples were waterproofed with thin plastic walls to enable ultrasonic transmission experiments to be performed in a water tank, using a planar immersion transducer to generate broadband input pulses and a miniature hydrophone to detect the transmitted wave field in a single speckle. The hydrophone was scanned, typically in a grid of 55x55 positions that were a wavelength apart, to measure the dynamic near-field speckle pattern of the multiply scattered waves.

By averaging the transmitted field over all speckles, it was possible to extract the coherent component from the total wave field, and hence to measure the longitudinal phase and group velocities, as well as the scattering mean free path [8]. These measurements showed that very strong scattering is observed over the entire intermediate frequency regime (~ 0.2 – 3 MHz), with kl_s varying from approximately 1 to 2.5. In addition, band gaps due to weakly coupled resonances of the beads were observed, as first reported by Turner and Weaver [9]. In the first gap, which occurs near 0.5 MHz in our samples, there are essentially no modes, and transport through the sample proceeds by tunneling [10]. In this paper, we focus on the phonon behaviour outside and above the first band gap.

3. Statistical approach to phonon localization

The approach to localization is characterized by large fluctuations in all transmission quantities, reflecting the underlying changes due to interference effects in the speckle pattern [11]. As a result, the distribution of intensity in the speckle pattern is different to the usual Rayleigh statistics for diffuse modes, for which the probability that the intensity in a speckle spot, normalized by its average, has a value $s_{ab} = I_{ab}/\langle I_{ab}\rangle$ decreases exponentially with s_{ab}, i.e. $P(s_{ab}) = \exp(-s_{ab})$. Here a and b represent incident and outgoing modes, respectively. A comparison of the speckle statistics for diffuse and localized waves is shown in figure 1. Figure 1(a) shows that Rayleigh statistics are obeyed in the *lower* part of the intermediate frequency regime (e.g., for the glass bead sinters considered in [6]), indicating that these modes are diffusive despite very strong scattering ($kl_s \sim 1$). By contrast, near the *upper* end of the intermediate frequency regime, strong departures from Rayleigh statistics are found for our aluminium bead samples, as shown in figure 1(b). The tail ($s_{ab} > 10$) of the intensity distribution in figure 1(b) was found to exhibit stretched exponential behavior, $P(s_{ab}) \sim \exp[-2(g' s_{ab})^{0.5}]$, with a value of the effective dimensionless conductance $g' = 0.84$, which is consistent with the theoretical prediction for localization that $g' < 1$ [11]. The data were also compared with predictions for the full intensity distribution given by Nieuewenhuizen and van Rossum [12], confirming that $g' < 1$ and indicating that the acoustic phonon modes are localized at frequencies near 2.4 MHz for this system.

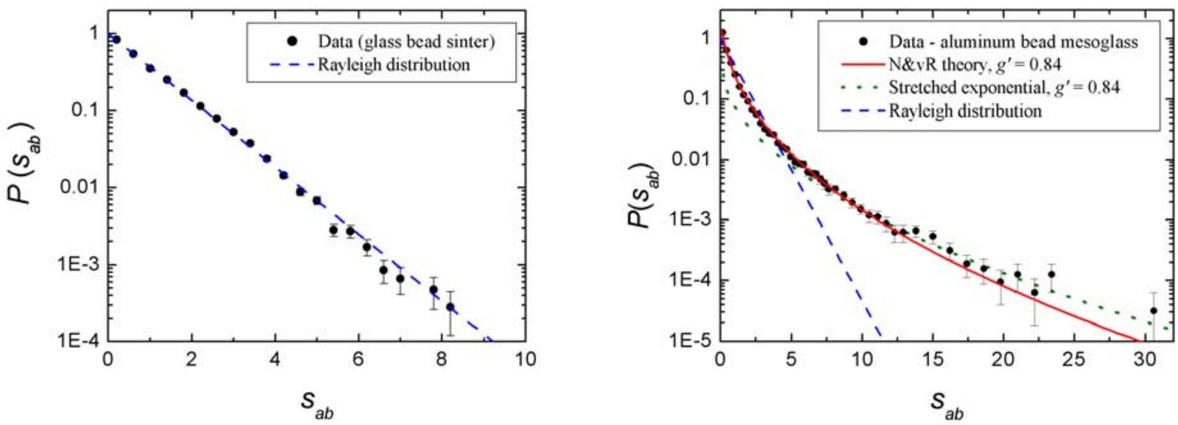

Figure 1. Speckle intensity distributions for mesoglasses in the lower (a) and upper (b) parts of the intermediate frequency regime. In (b), the frequency is 2.4 MHz, while in (a) the effective frequency (scaled by the difference in particle size) is 40 times lower.

The large values of s_{ab} that lead to non-Rayleigh statistics also cause the variance of the normalized transmitted intensity to be large, with var(s_{ab}) exceeding the diffuse value of 1 and approaching 4 near the highest frequencies investigated (figure 2). Chabanov *et al.* have proposed that localization is achieved when the var(s_{ab}) = $1 + 4/(3g') > 7/3$, corresponding to the localization condition $g' < 1$ [11]. In figure 2 the frequency dependence of var(s_{ab}) and g' (from $P(s_{ab})$) is compared, with the threshold values for localization indicated by dashed lines. These data indicate that the mobility edge occurs near 2.1 MHz for this aluminum bead mesoglass.

4. Time-dependent transmission

Additional evidence for a crossover from diffusive to localized waves was obtained by measuring the time-dependent ensemble-averaged transmitted intensity $I(t)$ in pulsed experiments. Here t is the propagation time. In the lower part of the intermediate frequency range (~0.25 MHz), the ensemble-averaged transmitted intensity $I(t)$ was determined at different frequencies and found to have an exponential decay at long times, $I(t) \sim \exp(-t/\tau_D)$, which was very well explained by diffusion theory. By fitting the predictions of the diffusion approximation to the measured $I(t)$, following the procedures outlined in [6, 13], the diffusion coefficient D was measured and found to be independent of frequency. This result is consistent with measurements of the diffusion coefficient in very porous glass bead networks throughout the lower part of the intermediate frequency regime [6], and confirms that ultrasound propagates diffusively at these lower frequencies.

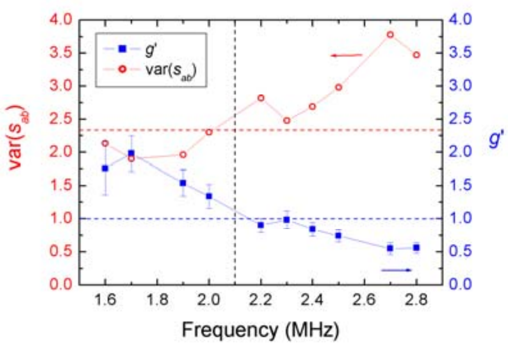

Figure 2. Frequency dependence of var(s_{ab}) and g'.

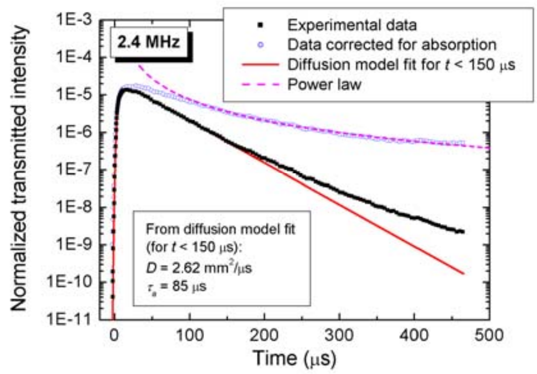

Figure 3. Time dependence of the average intensity $I(t)$.

In the upper part of the intermediate frequency range, the scattering of ultrasonic waves became stronger, and different behaviour for the time dependence of $I(t)$ was observed. In contrast to the diffusive regime, the tail of $I(t)$ has a non-exponential decay, which is not consistent with predictions based on diffusion theory (figure 3). It is convenient to describe the deviations from pure exponential decay as if they arise from a diffusive process with a time-dependent diffusion coefficient $D(t)$ that becomes smaller as the propagation time increases. If one assumes that the diffusion approximation is able to correctly describe the behaviour at early propagation times (< 150 μs) where the decay of $I(t)$ still appears exponential, and fits diffusion theory to estimate the absorption time τ_a, then one can correct the data for absorption by multiplying $I(t)$ by $\exp(t/\tau_a)$. The corrected $I(t)$ is shown by the open symbols in figure 3 and exhibits power law behaviour at long times. This behaviour is consistent with recent predictions based on the self consistent theory of the dynamics of localization that $I(t) \propto t^{1+s}$ (with $s \approx 0.85$) and $D(t) \propto 1/t$ at times $t >> t_D$ in the localization regime [14]. A more quantitative description of the time dependence of the transmission is given by fitting the data with predictions of the self consistent theory over the entire range of propagation times. An excellent fit is obtained, enabling the localization length ξ to be determined. This fit gives a value of ξ that is comparable to the sample thickness of 14.5 mm and provides convincing evidence for the dynamic localization of the acoustic phonons in this three dimensional system. We have also exploited the use of a quasi- point source and a sub-wavelength-sized detector, along with corresponding theoretical calculations for this geometry, to probe the transverse structure of the transmitted intensity for localized acoustic waves. We find that the transverse spreading of the intensity profile is dramatically inhibited by localization, giving new information of the properties of localized acoustic phonons in three-dimensions.

5. Conclusions

We have used ultrasonic experiments, in conjunction with recent theoretical developments, to demonstrate the Anderson localization of acoustic phonons in a three-dimensional mesoglass made from braised aluminum beads. We find, in the upper part of the intermediate frequency regime, that the time-dependent transmitted intensity decreases non-exponentially with time, consistent with theoretical predictions for localization. In the same frequency range, we also find that the normalized intensity distribution $P(s_{ab})$ exhibits non-Rayleigh statistics and can be fitted with a stretched exponential distribution. These results show that ultrasonic experiments provide a powerful way of probing acoustic phonon localization, opening up new possibilities for exploring the behaviour of localized waves in strongly disordered three-dimensional materials.

Acknowledgements

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