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Abstract.
We report on frequency-resolved experimental measurements of the ultrasonic diffusivity in the MHz range. By means of robotics, we produced a highly monodisperse suspension made of soft metallic micro-beads randomly-dispersed in a water-based gel-matrix. The measured diffusion coefficient is shown to vary significantly with frequency by more than one order of magnitude that is probably due to the sharp scattering resonances of the metallic particles.

1. Introduction

Classical waves, such as light or sound, propagating in a random medium progressively lose their coherence and behave diffusively, similarly to classical particles [1]. When the diffusive regime is reached, energy spreads proportionally to the square root of time like a cloud of particles undergoing a random walk at a rate determined by the diffusion coefficient (or diffusivity). Various experimental measurements of ultrasonic diffusivity were reported in polycrystals [2], glass beads suspensions [3, 4], aluminum foams [5], concrete [6, 7, 8], steel rods [9, 10], etc. Recently, the achievement of highly monodisperse emulsions served to emphasize the strong impact of scattering resonances on the key transport parameters of classical waves in disordered media through ultrasonic experiments [11]. Through accurate measurements of both ballistic and diffusive transport over a wide range of frequencies, Tallon *et al.* showed that the group velocity might be large near sharp resonances, whereas the energy velocity (as well as the diffusion coefficient) is significantly slowed down by resonant scattering delay [12]. Moreover, the important influence of the relative refractive index of scattering particles on energy transport has been also brought to light. Actually, Tallon *et al.* found interestingly that the energy velocity that describes the transport of energy by the dominant diffusive waves is mainly governed by the sound speed within the scatterers, and can be either much slower or faster than any of the other wave velocities by investigating highly monodisperse resonant emulsions containing either "slow" oil droplets (sound speed c_1 less than c_0 of the surrounding fluid) or "fast" liquid metallic droplets ($c_1 > c_0$) [13]. If their concentration is too high, liquid droplets tend to coalesce [11] and monodisperse resonant emulsions might be no longer suitable for the study of the diffusive wave transport in concentrated systems. It will be thus interesting to use other

resonant (solid) particles to circumvent these problems. 'Rigid' scatterers, such as steel rods [10], were demonstrated to exhibit some acoustic resonances. These shape resonances might be enhanced by considering 'softer' particles, *i.e.*, with a low shear elastic modulus (and with a low shear wave speed) because they are more deformable.

2. Materials and methods

The arrangements of parallel steel rods immersed in water were used for a long time as "model systems" to study the diffusive transport of ultrasound in resonant multiple scattering media [14]. Viard and Derode measured the ultrasonic diffusivity that was shown to vary with frequency, sometimes by more than a factor 2, in these systems composed of rigid metallic scatterers [10]. Alternatively, the use of soft metallic inclusions may enhance the scattering resonances provided their acoustic absorption remains very low. Some metals are well-known to be very soft, such as lead, in such a way that shear waves propagate slowly in these soft homogeneous materials ($c_{\mathrm{T}} \approx 0.7$ mm.μs^{-1}). There is a wide range of lead-based alloys, some of which are "low melt". Here, we used a eutectic alloy which undergoes liquefaction at ≈ 47 °C, supplied by Indium Corp. (Indalloy F117, contains 44.7% Bi, 22.6% Pb, 19.1% In, 8.3% Sn, 5.3% Cd). Its density is $\rho = 9.16$ g.cm^{-3} and the phase velocities were measured precisely as $c_{\mathrm{L}} = 2.37$ mm.μs^{-1} and $c_{\mathrm{T}} = 1.03$ mm.μs^{-1} for longitudinal and transverse waves, respectively, by using Time-of-Flight techniques. The corresponding attenuation coefficients α_{L} and α_{T} are so low in this BiPbInSnCd material that we could not measure these two parameters in the MHz range.

The emergence of scattering resonances in random media requires for highly monodisperse systems [15, 16] because polydispersity tends to smooth the sharp scattering resonances [17]. To produce a large amount of highly calibrated micro-beads made of Indalloy F117 fusible at low temperature, we used a millifluidic device developed by Tadmouri *et al.* [18]. Here, we fabricated BiPbInSnCd micro-beads with a average radius $a = 232$ μm and with a size-dispersion of 5%. Then, these spherical particles were randomly dispersed in a water-based gel-matrix of which acoustic properties were measured to be similar to water ($\rho_0 = 1.005$ g.cm^{-3}, $c_0 = 1.505$ mm.μs^{-1} and $\alpha_0 = 0.0015$ f^2 Np.mm^{-1} at frequency f in MHz). The volume fraction of particles is 10%.

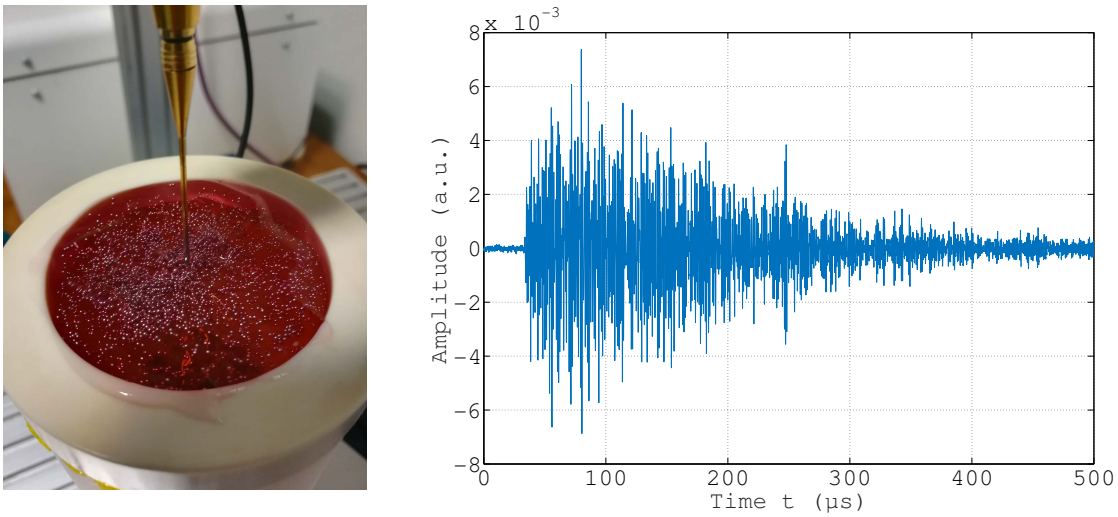

Figure 1. (left) Picture of the ultrasonic setup used to measure the time- and position-resolved transport of multiply scattered acoustic waves inside the suspension with a needle hydrophone. (right) Typical transmitted *coda* for a transverse distance ρ of 40 mm, at a depth $z = 15$ mm within a suspension of BiPbInSnCd micro-beads whose volume fraction is 10%.

The experimental setup used to probe the propagation of the multiply scattered waves was the same as that used by Tallon *et al.* [11, 12, 13]. Briefly, the incoming wave was generated by a focusing transducer (Olympus V395), excited with a short Gaussian pulse centered at 1.8 MHz, and was further confined to a well localized pointlike source by an acoustic diaphragm, which was placed on one side of a circular slab containing the sample. In order to measure the total time-dependent transmitted field in the regime where multiple scattering dominates, we used a small needle hydrophone with a diameter of 500 μm Precision Acoustics NH 0500). The hydrophone was immersed in the suspension as shown in Fig.1 and, because of its small size, was capable of resolving the spatial variation of the acoustic wave field inside the sample. The hydrophone detector was attached to motorized linear stages, allowing the diffuse field to be probed at any point (ρ, θ, z) within the circular sample (where $\rho = z = 0$ mm denotes the midpoint of the source at the input face of the sample). In this paper, all measurements were done at a depth $z = 15$ mm. Typical time dependence of a transmitted field at a transverse position $\rho = 40$ mm within the slab is shown in Fig. 1. This long *coda* signal is characteristic of multiple scattering contributions and the slow decrease in the amplitude of this signal with time is a signature of low absorption, due to very low intrinsic acoustic losses in the materials employed to fabricate the sample.

To characterize the diffusive transport of energy by the multiply scattered waves, the average field (averaged over all transverse positions ρ at $z = 15$ mm) was first subtracted from the total transmitted field in order to suppress any residual ballistic component and obtain only the multiply scattered field (the *coda*). It is worth noting that the transmitted time-dependent field at each position of the hydrophone was also filtered numerically with a narrow-band Gaussian filter ($\Delta f = 50$ kHz). The envelopes of the time-dependent transmitted fields were then calculated and squared to determine a quantity proportional to the energy density U at the position of the hydrophone z inside the sample for different radial positions ρ ranging from 0 mm to 40 mm (with a step of 5 mm), as shown in Fig. 2. For a given radial position ρ, the intensity profiles were averaged over 600 different angular positions θ (from 0 to $2\pi/600$).

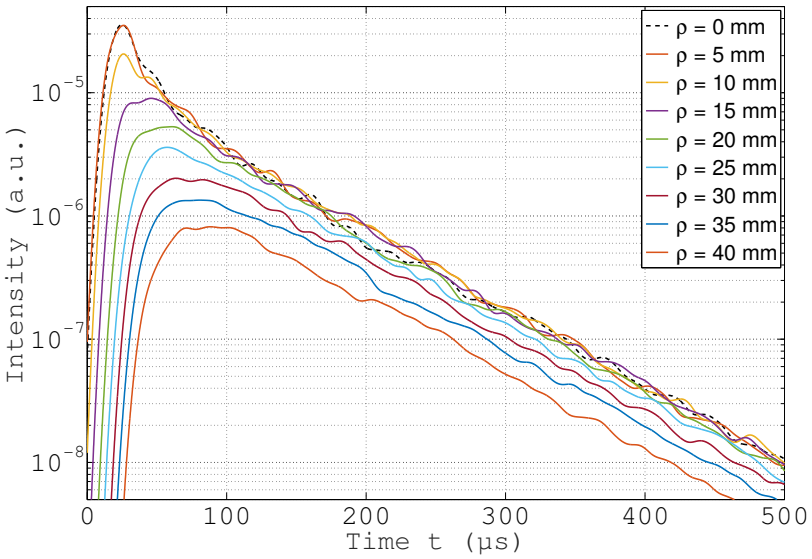

Figure 2. Intensity for different distances ρ, at a depth $z = 15$ mm within the suspension of BiPbInSnCd micro-beads, after numerical filtering of transmitted *codas* at $f = 1.0$ MHz.

3. Results and discussions

If the propagation distance is large enough [19], the ensemble-averaged energy density obeys the diffusion equation with characteristic diffusion coefficient (or diffusivity) D. From the solution of the diffusion equation for a point source incident on an infinite slab of thickness L, the evolution of the energy density as a function of time and position is known to have the form $U(z,\rho,t) = U(z,0,t)\exp[-\rho^2/(4Dt)]$, where $U(z,0,t)$ is a relatively complicated function that includes the effects of boundary conditions and absorption [3]. For a given value of z, the ratio $R(\rho,t) = U(\rho,t)/U(0,t)$ is a Gaussian that describes the transverse growth of the diffusive halo whose width is denoted w. The angular averaged energy density profiles shown in Fig. 3 clearly show the smooth Gaussian shape of the ensemble averaged diffuse halo at three different times. The fits of Gaussians to these averaged spatial profiles (solid curves) then enable the width squared of the diffusive halo $w^2(t)$ to be measured over time.

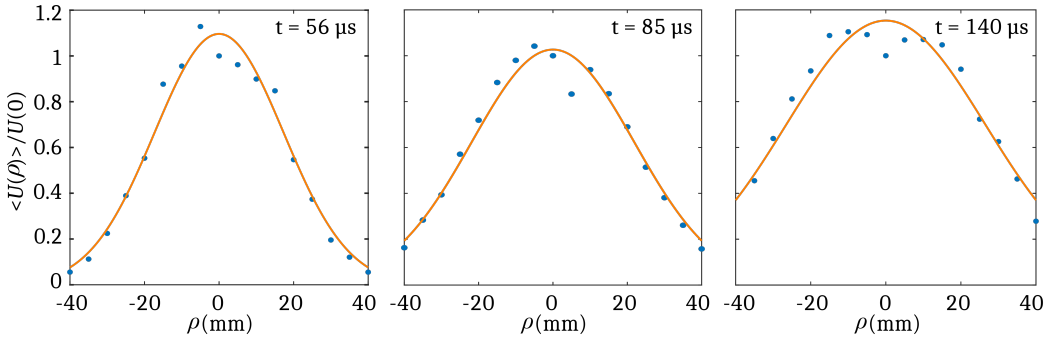

Figure 3. Angular-averaged spatial profiles of the energy density normalized by the energy density at $\rho = 0$ mm. Solid curves are fits of Gaussian expressions, *i.e.*, $\propto \exp[-\rho^2/w^2(t)]$, shown here at three different times t. These data are for a filtering frequency $f = 1.0$ MHz.

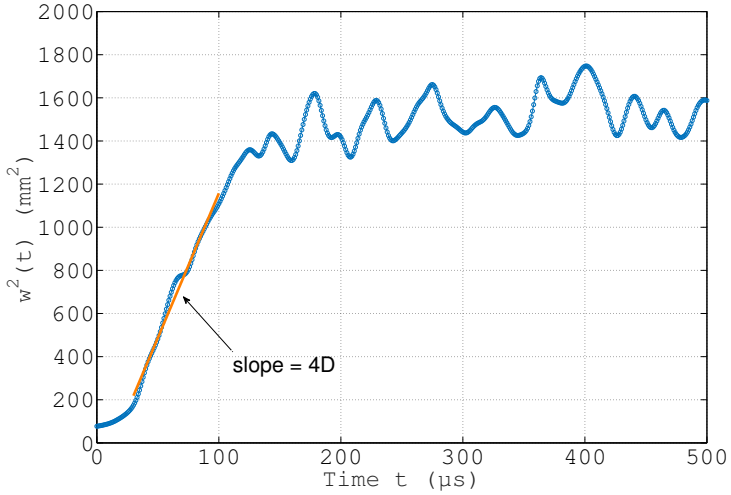

Figure 4. Time dependence of the width squared $w^2(t)$ of the diffusive halo deduced from the ratio $< U(\rho) > /U(0)$ measured and fitted at different times t. The solid line curve represents the linear fit $w^2(t) = 4Dt$ used to extract the diffusion coefficient D from these measurements. These data are for a filtering frequency $f = 1.0$ MHz.

The procedure for accurately measuring D was to capitalize on the linear dependence of the width squared of the diffuse halo on time, $w^2(t) = 4Dt$, and to perform a linear least squares fit of $w^2(t)$ versus t so as to determine D from the slope. Fig.4 shows typical results obtained at 1.0 MHz. The fit (solid line) was performed for a range of time for which $w^2(t)$ is proportional to the time t and over which numerical simulations of diffusion in a slab show that lateral walls of the cell have no effect on the halo evolution [11]. By repeating this procedure for measuring D from $w^2(t)$ at different filtering frequencies, we obtain the experimental values of the diffusion coefficient D shown as a function of frequency in Fig. 5.

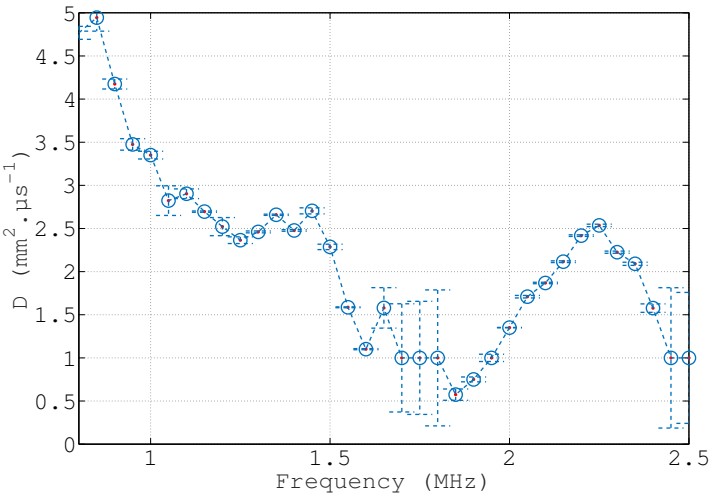

Figure 5. Measured diffusion coefficient (or diffusivity) D versus frequency.

As previously observed in strongly scattering resonant media such as monodisperse emulsions of fluorinated oil droplets [12, 13], the diffusion coefficient D is shown to varies strongly with frequency, from 5 mm^2.μs^{-1} at 0.85 MHz down to 0.5 mm^2.μs^{-1} at 1.85 MHz. Moreover, a clear dip is observed near the latter frequency, that should correspond to a sharp scattering resonance of the BiPbInSnCd micro-beads. For such a low value of D, the wave diffusion is slowed down significantly and might lead to a sub-diffusive wave transport regime [20]

4. Conclusion

We have reported on original measurements of ultrasonic diffusivity in a resonant multiple scattering suspension of soft metallic micro-beads. This coefficient of diffusion is shown to vary significantly with frequency by more than one order of magnitude reaching very low values for some frequencies. This slow down of the wave diffusion is probably due to the micro-beads scattering resonances whose the nature (mode/order) might be identified by looking at the modal coefficients of these spherical particles. Otherwise, complex wave phenomena such as Anderson localization [21] might occur in these strongly scattering resonant suspensions, provided the micro-beads concentration is high enough, since the wave diffusion might be stopped in the vicinity of strong scattering resonances. Unlike *all-fluid* resonant emulsions in which the coalescence of droplets becomes a major issue at high droplet concentrations [11], resonant suspensions made of solid scatterers might be "model systems" to complement previous works on the localization of ultrasound in three-dimensional media [22].

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