Resonant ultrasonic attenuation in emulsions

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Abstract. We report the achievement of scattering resonant emulsions devoted to the frequency-control of acoustic attenuation in the megahertz range. By means of robotics, we produced highly monodisperse, in both size and shape, fluorinated-oil droplet suspensions, providing experimental evidence of several Mie scattering resonances. Ultrasonic experiments performed in such complex media are compared, with an excellent quantitative agreement, to theoretical predictions derived within the framework of the independent scattering approximation.

1. Introduction

In disordered scattering media, the wave attenuation is basically due to i) the absorption within the surrounding media and ii) the wave scattering by the inclusions. To enhance the attenuation, an alternative is to exploit the scattering resonances of the inclusions, which depend on both their shape and their physical properties. In the audible domain ($f \leq 20$ kHz), many efforts have been devoted to the achievement of low-frequency resonators such as core-shell particles [1], or slip hollow spheres [2]. For higher frequencies ($f \geq 50$ kHz), the manufacture of such sophisticated objects is a difficult task due to the sub-millimetric scale required in the ultrasound domain. Despite their short lifetimes, the air-bubbles turned out to be an efficient way for ultrasonic insulation [3, 4]. The low-frequency resonators are not the only promising way to enhance acoustic attenuation: Mie scattering resonances could be a feasible alternative, provided a high sound speed contrast between the scatterers and the surrounding medium [5]. Although resonant scattering from spherical cavities in viscoelastic media has theoretically been considered for many decades [6], no experimental results have been reported about *resonant emulsions*, due to the coexistence of multiresonances and the difficulties associated with observing them at the high frequencies required [7]. Up to now, such random mixtures have only been considered in the long wavelength limit [8]. In this paper, we focus our attention on the resonant scattering regime occurring in emulsions.

2. Materials and methods

2.1. Sample fabrication

To observe Mie scattering resonances, we used a low absorption suspending gel that behaves almost like water in the megahertz range we investigated ($v_0 = 1490$ m.s^{-1}, $\rho_0 = 1$ g.mm^{-3} and $\alpha_0 = 10^{-4}$ MHz^{-2}.mm^{-1}). The yield stress of this water-based gel (~ 10 Pa) circumvents possible particle sedimentation but does not affect the wave propagation, *i.e.*, no shear wave can propagate into such fluid-like matrix. To enhance Mie scattering resonances, high compressibility and mass density contrasts between the two phases are required. As suggested in a recent numerical study [9], we used the fluorinated oil FC 40 (Fluorinert ®) possessing a rather low sound speed for a pure liquid ($v_1 \simeq 640$ m.s^{-1}) and a high mass density ($\rho_1 = 1.85$ g.mm^{-3}). As previously done for bubbly media [10], the emulsion was achieved thanks to robotics [11]. The fluorinated oil was continuously injected within the gel matrix by means of a moving syringe. Typically, 100 μm-radius droplets were obtained with large displacement velocities (~ 100 mm.s^{-1}) and low rates of oil-flow (~ 10 μL.min^{-1}). On one hand, this immiscible-oil-in-water emulsion is much more stable than a bubbly medium that makes its conservation longer. In addition, we obtained an excellent monodispersity (1%-polydispersity) as shown in Fig. 1. The volume fraction Φ_v of the fluorinated oil was estimated from the liquid quantity injected in the gel matrix. Then, the emulsion was optically characterized by the image analysis of about a hundred of droplets, of which radius measurements were made with a 1 μm-accuracy (see the particle size distribution in Fig.1). In this work, the mean radius of such emulsion made of fluorinated-oil droplets is $< a > = 104.5$ μm and the volume fraction is $\Phi_v = 0.5\%$. Note that the disorder arises from the emulsion transfert into a cell devoted to acoustic measurements

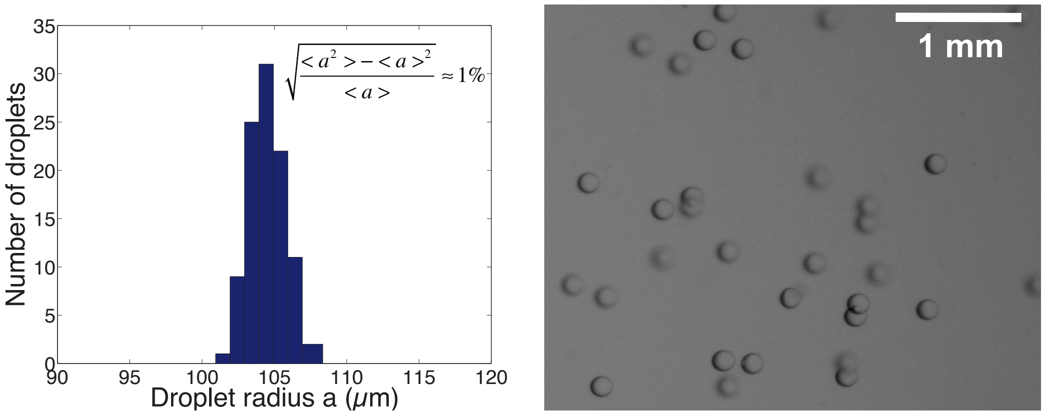

Figure 1. Discrete droplet-radius distribution measured by optical microscopy of an emulsion of fluorinated-oil droplets with $< a > = 104.5$ μm and $\Phi_v = 0.5\%$.

2.2. Acoustic measurements

Ultrasonic measurements were carried out in a large water tank (80x40x30 cm^3) by multi-echo spectroscopy [12]. An ultrasound pulse was generated by a broadband pulser/receiver connected to a broadband longitudinal wave transducer, having a central frequency of 10 MHz. The samples were poured into a polystyrene rectangular cell of which the width $d = 15$ mm was large enough to separate the multiple echoes on the walls. By means of this reflexion technique, we first measured the complex-valued wavenumber $k_0 = k_0' + ik_0''$ of the pure gel-matrix ($k_0' = \frac{\omega}{v_0}$ and $k_0'' = \alpha_0$ with $\omega = 2\pi f$ standing for the angular frequency). Then, we measured the effective attenuation coefficient of the emulsion that exhibits several sharp peaks (see Fig. 2).

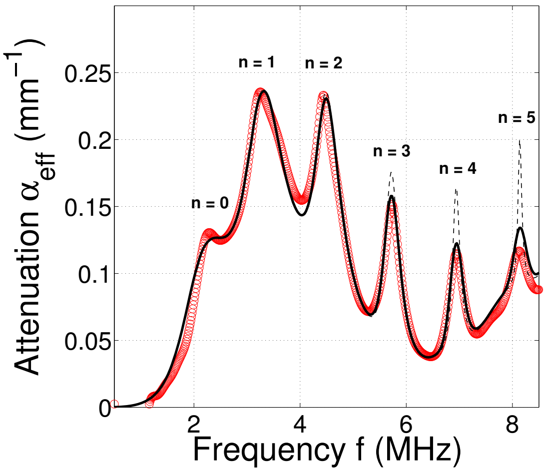

Figure 2. Acoustic attenuation measured in an emulsion made of fluorinated-oil droplets with $< a > = 104.5$ μm and $\Phi_v = 0.5\%$ (red open circles). Theoretical predictions calculated from the ISA model for a monodisperse emulsion (dashed line) and for a polydisperse emulsion (solid line) taking into account the particle size distribution measured by optical microscopy (Fig. 1).

3. Results and discussions

Since the volume fraction of oil droplets is quite low ($\Phi_v \leq 1\%$), we analyzed the experimental results within the framework of the independent scattering approximation (ISA), which is equivalent to the Foldy's model [13]. For polydisperse media [14], the effective wavenumber k_{eff} of the coherent pressure-wave propagating is given by:

$$k_{eff}^2 = \left(\frac{\omega}{v_{eff}} + \mathrm{i}\alpha_{eff}\right)^2 = k_0^2 + \int_a 4\pi\eta(a)\mathrm{d}a f_s(a) \tag{1}$$

where $\eta(a)$ is the number of droplets per unit volume whose radius is between a and $a + \mathrm{d}a$ ($\Phi_v = \int_a \frac{4}{3}\pi a^3\eta(a)\mathrm{d}a$) and $f_s = \frac{1}{\mathrm{i}k_0}\sum_{n=0}^{\infty}\{2n+1\}S_n(k_0 a)$ is the forward scattering function [15]. The complex-valued coefficients S_n, which depend on $k_0 a$ and also on the ratios $\frac{\rho_1}{\rho_0}$ and $\frac{v_1}{v_0}$, describe the scattering properties of a single droplet. The index n is associated with the nth mode, *e.g.*, n=0, n=1, n=2, n=3, n=4 and n=5 refer to the monopolar, dipolar, quadrupolar, hexapolar, octupolar and decapolar modes, respectively. The resonance frequencies of each mode fix the frequency-locations of the attenuation peaks[1]. As the absorption within the pure gel-matrix is very low ($\alpha_0 \simeq 10^{-2}$ mm^{-1} at 10 MHz), it could reasonably be neglected. In addition, Eq. 1 can be linearized since the volume fraction of droplets is quite low ($\Phi_v \leq 1\%$). Thus, the effective attenuation coefficient α_{eff} can be derived from the imaginary part of the effective wavenumber k_{eff} given in Eq. 1 as following:

$$\alpha_{eff} = \int_a \frac{-2\pi v_0^2\eta(a)\mathrm{d}a}{\omega^2}\mathrm{Re}\left[\sum_{n=0}^{\infty}\{2n+1\}S_n(k_0 a)\right] \tag{2}$$

From the droplet-radius distribution displayed in Fig. 1, we calculated the effective attenuation for the emulsion, revealing an good quantitative agreement with experiments, as

[1] The first-order resonances of the monopolar ($n = 0$), dipolar ($n = 1$), quadrupolar ($n = 2$), hexapolar ($n = 3$), octupolar ($n = 4$) and decapolar ($n = 5$) modes occur at 2.4, 3.3, 4.5, 5.7, 6.9 and 8.1 MHz, respectively, as shown in Fig. 2.

shown in Fig. 2. Moreover, a much better agreement is found by taking into account the particle size distribution measured by optical microscopy, rather than considering a monodisperse emulsion. The impact of polydispersity is particularly observable for high-order resonances as recently reported [14]. Thus, the ISA well-describes Mie scattering resonances occurring in dilute emulsions ($\Phi_v \leq 1\%$). As suggested by Richter *et al* [17], the high sensitivity to resonance peak frequency-positioning could make the *resonant regime* a useful tool for *particle sizing*, for which the long-wavelength regime is usually used [7].

4. Conclusion

In summary, we have presented clear experimental evidence about Mie scattering resonances occurring in dilute emulsions with high compressibility and mass density contrasts. This class of strongly scattering media might be used as absorbing materials provided the volume fraction of oil-droplets is appreciably increased ($\geq 10\%$). From a theoretical point of view, the interactions between oil-droplets should be taken into account *via* multiple-scattering theory.

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