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The effective velocity and attenuation of longitudinal waves through random dispersions of rigid, tungsten-carbide beads in an elastic matrix made of epoxy resin in the range of beads volume fraction 2%–10% are determined experimentally. The multiple scattering model proposed by Luppé, Conoir, and Norris [J. Acoust. Soc. Am. 131(2), 1113–1120 (2012)], which fully takes into account the elastic nature of the matrix and the associated mode conversions, accurately describes the measurements. Theoretical calculations show that the rigid particles display a local, dipolar resonance which shares several features with Mimaert resonance of bubbly liquids and with the dipolar resonance of core-shell particles. Moreover, for the samples under study, the main cause of smoothing of the dipolar resonance of the scatterers and the associated variations of the effective mass density of the dispersions is elastic relaxation, i.e., the finite time required for the shear stresses associated to the translational motion of the scatterers to propagate through the matrix. It is shown that its influence is governed solely by the value of the particle to matrix mass density contrast.

I. INTRODUCTION

As proposed from the very beginning of research on acoustic metamaterials,¹ a successful strategy for obtaining negative effective density, compressibility, or refractive index is to exploit the intrinsic (so-called local) resonances of mechanical resonators having a subwavelength size and embedded in the propagation medium. Compared to the phononic crystals strategy, the strategy based on local resonances benefits from mainly two advantages. First, the targeted properties can a priori be obtained without any crystalline order of the embedded resonators, although order may play a significant role. Second, since at resonance the acoustic wavelength in the embedding medium can be large compared to the resonator size, depending on the resonator design and performances, the targeted properties can be obtained using extremely thin samples compared to wavelength. Reported efficient locally resonant metamaterials (LRM) involve mechanical resonators with various degrees of sophistication and assembled in one, two, or three dimensions: Helmholtz resonators,²–⁴ air bubbles in liquids,⁵ solid membranes, either loaded⁶ or unloaded,⁷ liquid films in foams,⁸ core-shell particles,¹ mass-spring systems,⁹ “slow” spheres,¹⁰ rod-spring resonators,¹¹ hollow tubes,¹² and various combinations of them.¹³,¹⁴ In the audible domain several applications of LRM have already been implemented, which involve handmade or machined, resonators with centimeter size. In the ultrasonic domain, several applications of LRM are also foreseen. Their practical implementation raises several issues such as the resonators miniaturization and assembly,¹⁵ or the detailed knowledge of elastic properties of the materials constituting the resonators at ultrasonic frequencies.¹⁶

We focus on the dense core-soft shell particles, which have been used in the first LRM¹ and have been extensively studied theoretically. This kind of resonator can be sketched as a mass-spring resonator where the oscillating mass is the rigid, dense core and the spring is the compliant shell deformed by the core displacement.¹⁷,¹⁸ The high density of the core ensures that the phase delay of the core motion with respect to the matrix motion above the resonance frequency has a noticeable impact on the effective density of the LRM.¹⁷,¹⁹ The softness of the shell results in a small resonance frequency, ensuring that the wavelength in the embedding medium is large compared to the resonator size. Although this kind of resonator has allowed to obtain negative effective density in the audible domain,¹ its complex structure and the large dissipation in the viscoelastic shell at ultrasonic frequencies, which dampens the resonance,¹⁹ makes them difficult to implement in the ultrasonic domain. As shown by Kinra et al. in the 1980s,²⁰,²¹ another kind of composite material actually displays a solid body translation resonance, i.e., a similar local resonance, namely, a dispersion of dense, hard particles in a less dense, elastic matrix. Given the prolific worldwide research on metamaterials, it is surprising that such a simple kind of LRM has, to our knowledge, never been studied by the metamaterials community.

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in particular, regarding its effective properties around its local resonance. More generally, as pointed in one of the last reviews on this kind of material, there is a lack of experimental data on the acoustic properties of dispersions of dense, rigid particles. In this article, we provide original, comprehensive, and accurate data on effective acoustic properties of random dispersions of dense, tungsten-carbide (TC) beads in an epoxy matrix around their local resonance frequency in the 0.1–2.5 MHz frequency range. From a theoretical point of view, while acoustic propagation in such dispersions at frequencies smaller than the local resonance frequency has been the subject of several works, the influence of the local resonance on acoustic propagation has actually been much less studied. In this article, we compare over the entire investigated frequency range our experimental data with several multiple scattering theories, in particular, with a recent model which takes into account the effect of longitudinal-transverse wave conversions on the scattering of waves.

Our article is organized as follows. In Sec. II, we present the acoustic properties of the beads and matrix and we detail the samples fabrication protocol. Then, we describe the acoustic measurements and signal analysis procedures. In Sec. III, we briefly present the ingredients of the multiple scattering theories, which we aim to confront to our measurements, and we thoroughly compare our experimental data to these theories. Finally, in Sec. IV, we provide several physical insights in the scatterer dynamics and in the acoustic propagation in these dispersions. We evidence the strong similarities of this kind of resonance with Minnaert’s resonance in bubbly liquids. We identify the causes of smoothing of the dipolar resonance and evaluate their impact on the behavior of the effective mass density of the dispersions.

II. EXPERIMENTS

A. Materials properties

The composite material under study is a suspension of dense, TC spherical particles (labeled by \( p \) in the following) with monodisperse diameter, randomly dispersed in a matrix made of epoxy resin.

We used spherical, submillimetric TC beads supplied by Marteau-Lemarié (Pantin, France) and having nominal density \( \rho_p = 14.95 \times 10^3 \) kg m\(^{-3} \) and Lamé constants \( \lambda_p = 2.09 \times 10^{11} \) Pa and \( \mu_p = 2.66 \times 10^{11} \) Pa. Two bead sizes have been used, 397 \( \mu \)m and 500 \( \mu \)m nominal diameter \( 2R \), with small nominal diameter dispersity (\( \pm 2.5 \) \( \mu \)m).

Epoxy resin, “2-Ton” model supplied by ITW Devcon (Danver, MA), has nominal density \( \rho = 1100 \) kg m\(^{-3} \). It has been chosen for its rather long curing time (30 min), which allows for (i) a quiet insertion of the beads, (ii) a reasonable heat release rate during curing, and (iii) time for enclosed air bubbles to rise and burst.

Even once cured, epoxy is well known to display a pronounced viscoelastic behavior in the ultrasonic frequency domain, as a consequence of its polymeric nature. In order to access the epoxy viscoelastic properties, frequency-dependent velocities and attenuations of both longitudinal (compression) and transverse (shear) waves in cured epoxy have been measured. The velocity \( c_L \) and attenuation \( \alpha_L \) of longitudinal waves in epoxy have been determined from transmission measurements pulses with 2.25 MHz central frequency at normal incidence through a 9.43 mm thick epoxy slab immersed in water using the same protocol as for the beads dispersion samples described below. The velocity \( c_T \) and attenuation \( \alpha_T \) of transverse waves in epoxy have been determined from transmission measurements of longitudinal pulses with 2.25 MHz central frequency through the slab and to exploit the longitudinal-transverse-longitudinal wave conversion at water epoxy interfaces, according to the method described in Ref.23. The measured variations of the velocities and attenuations of both longitudinal and transverse waves in epoxy as a function of frequency are plotted in Figs. 1(a) and 1(b), respectively.

The complex Lamé constants \( \lambda \) and \( \mu \) of epoxy are then determined from the relationships \( c_L = \sqrt{(\lambda + 2\mu)/\rho} \) and \( c_T = \sqrt{\mu/\rho} \). The frequency-dependence of the Lamé

![Fig. 1](./fig1.png)
Table I. Best fit values [based on Eq. (1)] of the parameters defining Lamé coefficients of epoxy resin.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>λ</th>
<th>μ</th>
</tr>
</thead>
<tbody>
<tr>
<td>C₀ (×10⁹ Pa)</td>
<td>3.6</td>
<td>1.05</td>
</tr>
<tr>
<td>C₁ (×10⁹ Pa)</td>
<td>0.1</td>
<td>0.2</td>
</tr>
<tr>
<td>τ₁ (×10⁻⁹ s)</td>
<td>5000</td>
<td>5000</td>
</tr>
<tr>
<td>C₂ (×10⁹ Pa)</td>
<td>0.3</td>
<td>0.15</td>
</tr>
<tr>
<td>τ₂ (×10⁻⁹ s)</td>
<td>500</td>
<td>500</td>
</tr>
<tr>
<td>C₃ (×10⁹ Pa)</td>
<td>0.25</td>
<td>0.1</td>
</tr>
<tr>
<td>τ₃ (×10⁻⁹ s)</td>
<td>50</td>
<td>14</td>
</tr>
<tr>
<td>C₄ (×10⁹ Pa)</td>
<td>0.3</td>
<td>0.15</td>
</tr>
<tr>
<td>τ₄ (×10⁻⁹ s)</td>
<td>8</td>
<td>50</td>
</tr>
</tbody>
</table>

Constants, which result from the relaxation of the polymers, can be accurately described using the classical Zener model of viscoelastic materials. 24,25 As shown in Figs. 1(a) and 1(b), satisfactory fits to the experimental data could be obtained using only four relaxation times τₙ, n = 1, 4, which are related to the Lamé constants according to the following expressions: 26

\[ C(\omega) = C₀ + \sum_{n=1}^{4} C_n \frac{(\omega \tau_n)^2 - i\omega \tau_n}{1 + (\omega \tau_n)^2}, \]  

where C stands for λ or μ, C₀ is its low frequency value, and Cₙ, n = 1, 4 are weighting coefficients associated to relaxation times τₙ. The values of the relaxation times and associated coefficients corresponding to these best fits are given in Table I. Noticeably, the values of relaxation times are in good agreement with values reported in Ref. 24. For accuracy and simplicity purposes, these analytical approximations of the Lamé constants of epoxy are used in the following instead of their measured values.

### B. Samples preparation

Despite the large viscosity of uncured epoxy, the beads density is so large that sedimentation occurs during the curing, even with more rapidly curing resins. Consequently, three-dimensional, homogeneous samples could not be obtained by simply stirring the epoxy-beads mixture before solidification. Thus, beads dispersions having volume fractions ranging from φ = 2% to 10% were manufactured layer by layer according to the following protocol.

The lengthy spreading of the viscous resin poured at each step, competing with its solidification and the impossibility to steer the resin without enclosing bubbles, imposed a minimum resin height of 1 mm for each layer for a full and homogeneous coverage of the sample surface.

Cylindrical, 55 mm diameter Petri dishes were chosen as molds. Once the sample thickness e and the beads volume fraction φ had been chosen, the corresponding mass of beads was weighted and the beads set was split into N subsets, N = e – 1 (with e expressed in millimeters) being the number of layers which are expected to constitute the sample. In a horizontal Petri dish previously covered with a mold release wax (reference 66 333MR-1000P from Finish Care Products, South El Monte, CA), a reproducibly h = 1 mm thick layer of resin was poured. A subset of beads was randomly dispersed 20 min after, and then curing was achieved in 10 min. This sequence was repeated N times. Finally, a layer of epoxy was poured and cured, which allowed the sample top surface to be planar, and then the sample was removed from its mold. h is smaller than the average distance between nearest neighbors in a random dispersion which is expressed as \(0.9R\phi^{1/3}\) (Ref. 27) and varies from 650 μm (respectively, 820 μm) to 300 μm (respectively, 380 μm) for 397 μm (respectively, 500 μm) diameter beads in the investigated range of volume fractions. Consequently, the resulting beads distribution displays some anisotropy, whose importance increases with volume fraction. Noticeably, h is smaller than the smallest wavelength in resin \(λ_{min} = 1\) mm, met for \(f = 2.5\) MHz, which allows us to ignore phononic effects. The characteristic features of all the samples produced for this study are given in Table II.

### C. Acoustic measurements

The acoustic properties of the samples were determined by performing underwater longitudinal wave transmission measurements at normal incidence. The samples, fixed on a tapped panel having an air gap inside, were immersed in a water tank. Three pairs of Olympus (Tokyo, Japan) identical piezoelectric transducers having different central frequencies \(f_i\) and diameters (see Table III) were used, which allowed us to reliably cover the frequency range [0.1, 2.5] MHz. The sample was placed in the far field of the emitter [distance slightly larger than \((D/2)^2/f/c₀\), where D is the transducer diameter and \(c₀\) the sound velocity in water] in order to be probed with plane waves; see Table III. The receiver was facing the emitter and located close to the sample (i.e., 20 mm away) in order to avoid possible diffraction effects by the sample-screen junction.

The emitting transducer was fed with a standard pulser-receiver (reference 5072 from Olympus). The transmitted signal detected by the receiving transducer was acquired at a 5 × 10⁹ samples per second sampling rate and averaged over 1024 acquisitions using a digital oscilloscope (this resulted in a frequency resolution of 5 kHz of the Fourier transform of the signals). Figure 2 displays two sets of 1 MHz and 2.25 MHz central frequency (\(f_i\)) signals transmitted through water and a 8.82 mm thick slab of a dispersion of 397 μm beads with volume fraction \(φ = 5\%\). Comparing the signals received after propagation through the beads dispersion (signal S in Fig. 2) to the signals received after propagation through water (signal S₀) and anticipating the spectral signal analysis, one can state that: (i) the earlier arrival of the signal is due to the larger velocity of longitudinal waves, (ii) the signal lengthening is due to the strong dispersion, (iii) the apparent change in central frequency of the 1 MHz signal results from the large attenuation for frequencies larger than 0.5 MHz.

As the acoustic pulse propagates through the suspension, it is scattered by the beads. The resulting wave propagating through a sample can be described as the superposition of a coherent wave independent of the beads distribution and an incoherent wave which depends on the beads distribution. The contribution of the incoherent wave to the received signal
depends on the pulse central frequency, which determines the efficiency of the scattering, on the sample thickness and acoustic beam diameter, which both determine the number of irradiated scatterers, but also on the receiver diameter since the spatial fluctuations of the incoherent wave in the receiver plane are averaged over the receiver surface. In order to reduce the contribution of the incoherent wave to the received signal, the latter is also averaged over 16 acquisitions performed at 16 different sample positions corresponding to the nodes of a $4 \times 4$, 2 mm step squared grid lying in the sample plane. By doing this, an averaging over the scatterers distribution is performed. The contribution of the incoherent wave to the signals can be evaluated by comparing a signal acquired along a given path through the sample; dashed curve, signal labeled $S_0$ and through a 8.82 mm thick sample filled with volume fraction $\phi = 5\%$ of 397 $\mu$m beads (solid curve, signal labeled $S$) acquired along a given path through the sample; dashed curve, signal labeled $S_r$ averaged over 16 different paths through the sample). The signals transmitted through the sample have been multiplied by a factor of 10 and 35 in (a) and (b), respectively.

signal $S_0(t)$ transmitted through water, i.e., in absence of sample, according to the following procedure.

In order to isolate the signal having propagated through the sample only once, only the part of $S_0$ beginning just before the main peak and lasting $2c/c_{\text{max}}$ was retained, the other part of $S_0$ begin padded with zeroes. $c_{\text{max}} = 2600 \text{ m s}^{-1}$ is the maximum velocity measured in the samples (see below). More precisely, the end of the rectangular window applied to $S_0$ was slightly tuned to coincide with a zero of $S_0$ in order to avoid any signal discontinuity. The same filter was applied to $S_r$. Then, the Fourier transforms of $S_0$, $S_0(f)$ and $S_0$, $S_0(f)$, were computed using Python language. With the aim of accurately unwrapping the phase of $S_0$, $S_0$ later, $S_0$ (respectively, $S_0$) was multiplied by $\exp[-\imath \omega t_0]$ (respectively, $\exp[-\imath \omega t_0]$), where $\omega = 2\pi f$ is the angular frequency and $t_0$ (respectively, $t_0$) is the instant of detection of the main peak of $S_0$ (respectively, $S_0$). Finally, the real $k'$ and imaginary $k''$ parts of the complex acoustic wavenumber $k_{\text{eff}}$ in the sample were determined independently as functions of the frequency by fitting separately the modulus and the phase of the complex transmission factor $T_1 T_2 \exp[i(k_{\text{eff}} - k_0) \ell] \exp[i\omega(t_0 - t_0')]$ to the ratio

### Table II

<table>
<thead>
<tr>
<th>Beads volume fraction $\phi$ (%)</th>
<th>Beads diameter $2R$ ($\mu$m)</th>
<th>Sample thickness $e \pm$ standard deviation (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>397</td>
<td>9.43 ± 0.01</td>
</tr>
<tr>
<td>2</td>
<td>397</td>
<td>5.51 ± 0.04</td>
</tr>
<tr>
<td></td>
<td>397</td>
<td>8.48 ± 0.01</td>
</tr>
<tr>
<td></td>
<td>397</td>
<td>19.17 ± 0.04</td>
</tr>
<tr>
<td>5</td>
<td>397</td>
<td>5.10 ± 0.09</td>
</tr>
<tr>
<td></td>
<td>397</td>
<td>8.82 ± 0.02</td>
</tr>
<tr>
<td></td>
<td>397</td>
<td>19.16 ± 0.04</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>5.15 ± 0.09</td>
</tr>
<tr>
<td>7.5</td>
<td>500</td>
<td>3.23 ± 0.12</td>
</tr>
<tr>
<td>10</td>
<td>397</td>
<td>3.36 ± 0.06</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>5.68 ± 0.03</td>
</tr>
</tbody>
</table>

### Table III

<table>
<thead>
<tr>
<th>Olympus reference</th>
<th>V301</th>
<th>V302</th>
<th>V323</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter (inch)</td>
<td>1</td>
<td>1</td>
<td>0.25</td>
</tr>
<tr>
<td>Central frequency $f_i$ (MHz)</td>
<td>0.5</td>
<td>1</td>
<td>2.25</td>
</tr>
<tr>
<td>Exploitble frequency range (MHz)</td>
<td>0.1 0.6</td>
<td>0.1 1.2</td>
<td>0.3 2.5</td>
</tr>
<tr>
<td>Emitter sample distance (mm)</td>
<td>70</td>
<td>120</td>
<td>40</td>
</tr>
</tbody>
</table>
the case of strong $T$ dissipation in the matrix, i.e., when shear waves are strongly attenuated along their path from a scatterer to another.

For the case of randomly distributed elastic spheres in elastic media, explicit models, in which effective wavenumber is given by an explicit formula, and self-consistent models, in which it is the solution of a characteristic equation, have been proposed. In the following, the explicit model derived in Ref. 42 is preferred because it allows to clearly identify the contributions of wave conversions ("coupling terms"). More precisely, the effective wavenumbers of coherent $L$ and $T$ waves are derived in the spirit of Fikioris and Waterman’s approach, which is based on the quasi-crystalline assumption and takes account of the non-overlap between the scatterers (“hole correction”). In this model, the parameter $b$ (hole radius) introduced in the course of the field averaging process satisfies $b = 2R$. In the following, the effective wavenumber of the coherent compression wave predicted using this model is labeled LCN model. Interestingly, direct numerical simulations of multiple scattering in concrete have confirmed the validity of LCN model in the case of weak scattering up to densities of 25%.

Note that, in the long wavelength limit, LCN ($b \rightarrow 0$) model identifies with Linton and Martin’s model (hereafter, labeled LM model) with an additional contribution due to wave conversion. This remark will later lead us to conclude on the influence of wave conversion on coherent propagation from the benchmarking of LCN and LM models against experiments.

B. Experimental results

We now confront the above presented theoretical predictions to the measurements. Figures 3, 4, and 5 display the experimental variations of the velocity and attenuation of longitudinal coherent waves together with their predictions using LM and LCN models as functions of $f$ and $k_L^0 R$ ($k_L^0 = c_L/\bar{c}_L$, $c_L^0$ being the zero-frequency value of the longitudinal velocity in epoxy $c_L = 2276.4$ m s$^{-1}$) in dispersions of 397 $\mu$m diameter beads in epoxy with volume fractions $\phi = 2\%$, 5\%, and 10\%, respectively. The discrepancy between the superposed experimental curves obtained using different transducers (see above) can be regarded as an evaluation of the overall uncertainty associated to the measurements.

As a preamble, we note that strong dispersion and attenuation, which are the signature of a resonance, occur around $k^0 R \approx 0.3$. This demonstrates that this resonance is actually subwavelength. First, LCN model is observed to fit to the experimental data globally much better than LM model for all volume fractions. In particular, the velocity peak predicted by LM model (and not by LCN model) is not reproduced by the experimental data. This definitely demonstrates that wave conversions play a key role in the propagation of coherent waves in elastic media and that LCN model fairly describes the influence of wave conversion. Second, for $\phi = 5\%$ and 10\%, LCN model is observed to fit to the experimental attenuation better than LCN ($b \rightarrow 0$) model. This confirms the relevance of the hole correction in LCN model.
The agreement between LCN model and experimental data is found to be excellent for \( \phi = 2\% \) and for 5\% for frequencies larger than 0.5 MHz. This demonstrates the accuracy of LCN model. Conversely, the discrepancy between LCN model and the experimental data for \( \phi = 10\% \) is quite large. This can be attributed to the loss of accuracy of analytical models at high volume fractions in the case of strong scattering due to the sphere resonance. The discrepancy between theory and experiment observed in this frequency range could also be due to the anisotropy of the samples, which could blur this fine behavior. Noticeably, LCN model displays negative attenuation and simultaneously positive velocity around 0.4 MHz for \( \phi = 5\% \) and 10\%, whereas the experimental velocity and attenuation are both positive in this frequency range. This unphysical attenuation has already been observed by different authors\(^{38,44,45} \) without explanation.

**IV. FURTHER THEORETICAL DEVELOPMENTS**

As pointed out by Moon and Mow,\(^{36} \) the physical phenomenon responsible for the strong acoustic dispersion and attenuation observed around 0.5 MHz in elastic dispersions is the resonance corresponding to the particle deformation eigenmode labeled \( n = 1 \), also called the dipolar resonance of the scatterers. This allows us to call this resonance a local resonance, in contrast to spatial interferences involving inter-particle distance, like in phononic crystals.

The physical ground of dipolar scattering is the following. When the density (inertia) of the particles differs from that of the surrounding elastic matrix, the cyclic variations of the mechanical stress associated with the wave lead to oscillating motions of the matrix and of the particles having different amplitudes,\(^{46} \) thus, on an oscillating relative motion of the particle with respect to the matrix. It follows that the particle scatters acoustic energy according to a dipolar pattern (associated to the dipolar resonance \( n = 1 \)) if the particle is small with respect to the incident wavelength. The resonant behavior of the scatterer motion and the associated acoustic scattering relies on the combination of the particle...
The scatter excited by an incident longitudinal wave have a minor influence on the amplitudes of the modes $n = 1$ of the longitudinal and transverse scattered waves in the matrix contrary to its mass. Given the large contrast in elastic properties between TC and epoxy resin ($\tilde{\mu} \approx \mu_p/\mu_0 = 253$), it is thus reasonable to consider the scatterer as a moving rigid particle. In this limit, its dipolar vibration mode can be seen as a simple oscillatory translation, which is exactly the motion exploited in core-shell particles based LRM.

LCN model involves the scattering coefficients $T_{LL}^n$ that describe the scattering of a longitudinal plane wave by a unique particle in the form of a longitudinal wave, as well as $T_{LT}^n$ (respectively, $T_{TT}^n$) that describes the scattering of a longitudinal (respectively, transverse) plane wave in the form of a transverse (respectively, longitudinal) wave, i.e., the scattering coefficient associated to mode conversion $L \rightarrow T$ (respectively, $T \rightarrow L$). The effective wavenumber $k_{LL}^{\text{eff}}$ depends on these scattering coefficients as follows:

$$
\left(\frac{k_{LL}^{\text{eff}}}{k_{LL}}\right)^2 = 1 - i \frac{3\phi}{(kLR)^2} \sum_{n=0}^{\infty} (2n + 1)T_{LL}^n + \phi^2 F(T_{LL}^n, T_{LT}^n, T_{TT}^n),
$$

where the function $F(T_{LL}^n, T_{LT}^n, T_{TT}^n)$ is given in Ref. 42. Accordingly, $L \rightarrow T$ and $T \rightarrow L$ mode conversions do not come up at first order in $\phi$. This result is predicted by all the existing multiple scattering models.

We now aim to determine the eigenfrequency associated to the vibration eigenmode $n = 1$. Since at the dipolar resonance $|kLR| < 1$, we consider for simplicity purpose the approximate expression of the dipolar scattering coefficient $T_{LL}^1$ in the long wavelength regime $|kLR| \ll 1$ (Ref. 49),

$$
T_{LL}^1 = \frac{i(kLR)^3}{9} (\tilde{\rho} - 1) \frac{3(kLR)^2 + 9i \kappa R - 9}{(2\tilde{\rho} + 1)(kLR)^2 + 9i \kappa R - 9},
$$

where the mass density ratio appears as

$$
\tilde{\rho} = \frac{\rho_p}{\rho}.
$$

Generally speaking, resonances correspond to the poles of the scattering coefficients. If the matrix is assumed to be purely elastic, i.e., using for $\mu$ its zero-frequency, real value $\mu_0 = \rho(\zeta_T^0)^2$, with $\zeta_T^0 = 977$ m s$^{-1}$ the zero-frequency value of the transverse velocity, one of the poles of the denominator of Eq. (3) gives the following simple expression for the resonance frequency $f_{\text{res}}$:

$$
f_{\text{res}} = \frac{3\zeta_T\sqrt{8\tilde{\rho} - 5}}{4\pi R(2\tilde{\rho} + 1)}
$$

and the quality factor $Q$ of this resonance is given by

$$
Q = \frac{1}{3} \sqrt{8\tilde{\rho} - 5}.
$$

Using these approximate expressions, one finds $f_{\text{res}} = 425$ kHz and $Q = 3.4$ (respectively, $f_{\text{res}} = 337$ kHz and $Q = 3.4$)
for 397 μm (respectively, 500 μm) diameter beads in epoxy matrix. These approximate values of the resonance frequency differ from their exact values ($f_{\text{res}} = 465$ kHz for 397 μm diameter beads and $f_{\text{res}} = 366$ kHz for 500 μm diameter beads) by $<10\%$, which confirms the relevance of approximation (5). The approximate values of the quality factor are even closer to their exact values ($Q = 3.2$ for 397 μm diameter beads and $Q = 3.1$ for 500 μm diameter beads). It is also worth noting that, in this limit, $Q$ only depends on the density contrast and is a monotonously increasing function of $\tilde{\rho}$ from $\tilde{\rho} = 5/8$. The same conclusion has already been drawn concerning the resonance of core-shell particles through numerical investigations.19,50

Finally, we address the influence of the matrix viscoelasticity on the dipolar resonance frequency. Viscoelasticity is indeed expected to play a noticeable role in the present case since the resonance frequency is close to the inverse of the first relaxation time $\tau_1$. The actual resonance frequency can be numerically evaluated by solving Eq. (5) recursively: at the first iteration, the low frequency value of the matrix shear modulus is used for determining the resonance frequency $f_{\text{res,1}}$; at the second iteration, the value of the matrix shear modulus at frequency $f_{\text{res,1}}$ is used for determining the resonance frequency $f_{\text{res,2}}$, etc. One ultimately finds $f_{\text{res}} = 483$ kHz (respectively, $f_{\text{res}} = 381$ kHz) for 397 μm (respectively, 500 μm) diameter particles in epoxy. Viscoelasticity induces a relative frequency shift on the order of 4%.

B. Analogy with Minnaert resonance in bubbly liquids

In the $\tilde{\rho} \gg 1$ limit, $f_{\text{res}}$ has the following approximate expression:

$$f_{\text{res}} \sim \frac{3\sqrt{2}}{4\pi} \frac{1}{R} \sqrt{\frac{\mu}{\rho_p}}.$$  (7)

This resonance frequency $f_{\text{res}}$ depends on the shear modulus of the matrix, i.e., of the most deformable under shear material, and on the inertia of the particle, i.e., of the densest material. This hybrid combination of mechanical properties in the factors determining this dipolar resonance is very similar to the Minnaert resonance of the volume oscillations (monopolar resonance) of a gas bubble in a liquid, for which the relevant stiffness is the one of the most compressible material (the gas) and the relevant inertia is the one of the densest material (the liquid). This is precisely the kind of hybridization of mechanical properties of the two-components material which makes both the Minnaert resonance of bubbly liquids and the dipolar resonance of dense particles dispersed in an elastic matrix subwavelength.

C. Influence of the density contrast on resonance

Finally, we aim to numerically determine the influence of particle density and shear modulus on the propagation of the coherent wave. For simplicity purposes, the matrix is assumed to be purely elastic and has the epoxy density $\rho = 1100$ kg m$^{-3}$ and the Lamé constants $\lambda = \lambda_0 = 3.6 \times 10^9$ Pa and $\mu = \mu_0 = 1.05 \times 10^9$ Pa. The beads diameter and volume fraction are fixed equal to 500 μm and $\phi = 2\%$. First, a numerical study is performed by varying the bead density $\rho_p$ between 5500 kg m$^{-3}$ and 22000 kg m$^{-3}$ ($\rho$ ranging from 5 to 20) for fixed values of the bead Lamé constants $\lambda_p = 2.09 \times 10^{11}$ Pa and $\mu_p = 2.66 \times 10^{11}$ Pa, so that $\tilde{\rho} = 253$. The frequency dependences of the dimensionless velocity $c_L^{\text{eff}}/c_L$ and dimensionless attenuation $\xi_L^{\text{eff}}R$ of the longitudinal coherent wave are plotted in Figs. 6(a) and 6(b), respectively, for four different values of $\tilde{\rho}$, together with the corresponding resonance frequencies $f_r$, $i=1 \ 4$, computed using Eq. (5). First, we note that, as in the case of Minnaert resonance of bubbly liquids, the resonance frequency coincides with the inflection of attenuation and with the minimum of velocity. Moreover, the larger $\tilde{\rho}$ is, the larger are the variations of velocity and the attenuation at resonance, as expected from the predicted increase of the quality factor of the resonance with $\tilde{\rho}$. These observations complete the analytical study presented hereinabove.

D. Link between particle dynamics and effective acoustic properties

As analyzed in Refs. 36, 51, and 52, under the hypotheses of linear, Hookean elasticity Newton’s principle applied to a single rigid particle moving in a viscoelastic matrix leads to the following differential equation that couples the particle displacement $u_p$ and the matrix displacement $u$ associated to the coherent wave at the particle location:

$$\frac{4}{3} \pi R^3 \rho_p \ddot{u}_p = 6 \pi R \mu (u - u_p) + 6 \pi R \mu \frac{R}{c_T} (\dot{u} - \dot{u}_p)$$

$$+ \frac{2}{3} \pi R^3 \rho (\ddot{u} - \ddot{u}_p) + \frac{4 \pi R^3 \rho \dddot{u}}{3}.$$  (8)

Physical interpretation of the terms appearing in this differential equation is not present in the literature in the case of a solid matrix, contrary to the case of a fluid matrix. In this context, we compare the terms of the right-hand side (RHS) of Eq. (8) to those for the case of a viscous fluid matrix, i.e., Stokes viscous drag, Basset-Boussinesq history force, and Archimedes force.49 The first term of the RHS of Eq. (8) is an elastic spring force exerted by the matrix on the moving particle.53,54 This spring force is associated to the matrix deformation which extends over a distance comparable to $R$ and adapts to the particle displacement through the propagation of shear waves. The fourth term of the RHS can be interpreted as a time-dependent buoyancy (Archimedes-like force) associated to the matrix acceleration. In the low frequency $k_T R \ll 1$ regime considered here, adaptation of the matrix acceleration to the incident stress wave is instantaneous so that this time-dependent buoyancy is in phase with the matrix acceleration. On the contrary, since $c_T < c_L$, the spring force exerted by the matrix on the moving particle is expected to adapt to the particle displacement with a time delay on the order of $R/c_T$, highlighting a relaxation process of the spring force over the characteristic timescale $R/c_T$. The third term of the RHS is the so-called added mass effect which has a noticeable influence on the resonance frequency although it has been neglected in Ref. 36. We note
and the viscoelastic nature of the matrix shear modulus elastic relaxation, which occurs without energy dissipation, shear stresses associated to its displacement, called hereafter of propagation through the matrix close to the particle of the damping of the particle motion has two origins: the delay originates from the viscoelastic properties of the matrix. Thus, that an additional source of time delay of the spring force associated acoustic scattering.

This expression evidences the link between the translational motion of the particle, the particle vibration mode \( n = 1 \), and associated acoustic scattering.

\[
\begin{align*}
\text{FIG. 6.} & \text{ (Color online) Variations of (a) dimensionless effective longitudinal velocity and (b) dimensionless effective longitudinal attenuation as function of frequency for various values of the mass density contrast } \rho \text{ for a fixed value of the elasticity ratio } \mu = 253. \text{ The resonance frequencies corresponding to each value of } \rho \text{ are also indicated.}
\end{align*}
\]

that an additional source of time delay of the spring force originates from the viscoelastic properties of the matrix. Thus, the damping of the particle motion has two origins: the delay of propagation through the matrix close to the particle of shear stresses associated to its displacement, called hereafter elastic relaxation, which occurs without energy dissipation, and the viscoelastic nature of the matrix shear modulus \( \mu \), which involves energy dissipation, called hereafter viscoelastic relaxation. As shown in Ref. 55, damping of the particle motion due to elastic relaxation occurs through acoustic radiation.

The solution of Eq. (8) in the harmonic regime can be expressed as \( u_p = t_p u \) where

\[
\begin{align*}
\text{E. Influence of relaxation on effective acoustic properties}
\end{align*}
\]

We now aim to analyze the impact of both elastic and viscoelastic relaxations on the dispersion equation and on effective acoustic properties of the dispersion. For simplicity purpose, we adopt Foldy’s approximation for the dispersion equation, i.e., we consider Eq. (2) without the second-order term in concentration. In the low frequency regime \( |k_L R| \ll 1 \) considered here, the multipolar expansion in Eq. (2) can be safely restricted to the two first vibration eigenmodes \( (n = 0 \text{ and } n = 1) \). The simplified dispersion equation we now consider is thus

\[
\left( \frac{k_{LL}^0 R}{k_L} \right)^2 = 1 - i \frac{3\phi}{(k_L)^2} \left( T_{LL}^{01} + 3T_{LL}^{11} \right),
\]

where

\[
\begin{align*}
T_{LL}^{11} & = -i(k_L R)^2/3 \text{ for a rigid particle.} \quad 49
\end{align*}
\]

In both the numerator and denominator of Eq. (3), the imaginary number \( 9k_L R \) represents the contribution of elastic relaxation on the phase shift of \( u_p \) with respect to \( u \). Neglecting both elastic and viscoelastic relaxations, i.e., dropping \( 9ik_L R \) in Eq. (3) and using for \( \mu \) its zero-frequency, real value \( \mu_0 \), leads to the following expression for the effective wavenumber

\[
\left( \frac{k_{LL}^0 R}{k_L} \right)^2 = 1 - \phi + \frac{\phi(\hat{\rho} - 1)(k_L)^2}{(2\hat{\rho} + 1)(k_L)^2 - 9}.
\]

Under these assumptions, \( (k_{LL}^0)^2 \) is real. Thus, depending on its sign, \( k_{LL}^0 \) is either real or pure imaginary, i.e., the coherent wave is either progressive or evanescent. Analysis of Eq. (11) leads to identify a stop band, i.e., a frequency range for which evanescence occurs (Re[\( k_{LL}^0 \) ] = 0, Im[\( k_{LL}^0 \) ] \neq 0). The lower boundary of the stop band is the resonance frequency \( f_{\text{res}} = 3\tau R/2\pi R \sqrt{2\hat{\rho} + 1} \), which differs from \( f_{\text{res}} \) as given by Eq. (5) as a consequence of the further simplifications applied to the propagation model. Its upper frequency is a cutoff frequency which has the following expression:

\[
f_c = f_{\text{res}}^0 \sqrt{1 + \frac{2\phi(\hat{\rho} - 1)^2}{\phi(\hat{\rho} - 4) + 2\hat{\rho} + 1}}.
\]

Note that such an expression for the upper boundary of stop band has been previously derived by Moon and Mow \(^{36}\) and has been found to fit experimental measurements on dispersions of lead spheres in epoxy resin. \(^{35}\)

This stop band is visible in Figs. 7(a) and 7(b), which display, respectively, the variations of the real and imaginary parts of \( k_{LL}^0 \) as defined by Eq. (11) for the TC-epoxy dispersion as function of frequency for several values of \( \phi \). We now compare in Figs. 7(a) and 7(b) the frequency variations of \( k_{LL}^0 \) without relaxation, i.e., as given by Eq. (11), to the ones of \( k_{LL}^0 \) with elastic relaxation and with or without viscoelastic relaxation, i.e., as given by Eq. (10). The values chosen for the mechanical properties of the materials are those of TC-epoxy dispersion. Strikingly, elastic relaxation results in a considerable smoothing of the variations of Re[\( k_{LL}^0 \) ] and
Im[\(k_L^{\text{eff}}\)]. In particular, the stop band disappears, i.e., Re[\(k_L^{\text{eff}}\)] is positive for all frequencies. Adding viscoelastic relaxation only slightly increases the frequency at which Re[\(k_L^{\text{eff}}\)] is maximum and slightly decreases the maximum value of Im[\(k_L^{\text{eff}}\)] compared to the case with elastic relaxation only. We conclude that the main cause of smoothing of the dipolar resonance of the TC-epoxy dispersions studied here is elastic relaxation and not viscoelastic dissipation.

Finally, we address the question of the impact of relaxation on effective mass density, which is expected to be affected by the dipolar resonance of the scatterers and is of particular interest in the context of acoustic metamaterials. In the frame of Foldy’s model, the effective mass density of the dispersion \(\rho_{\text{eff}}\) is defined as\(^{56}\)

\[
\frac{\rho_{\text{eff}}}{\rho} = 1 - \frac{3i\phi}{2(k_L R)^2} \sum_{n=0}^{\infty} (2n + 1)[1 - (-1)^n] T_n^{LL}.
\]

(13)

In the low frequency regime \(|k_L R| \ll 1\) considered here, the multipolar expansion in Eq. (13) can, here, also be safely restricted to the two first vibration eigenmodes, which results in the following approximate expression for \(\rho_{\text{eff}}\):

\[
\frac{\rho_{\text{eff}}}{\rho} \approx 1 - \frac{9i\phi}{(k_L R)^3} T_1^{LL},
\]

and using Eq. (10), it yields

\[
\frac{\rho_{\text{eff}}}{\rho} \approx \phi + \left(\frac{k_L^{\text{eff}}}{k_L}\right)^2.
\]

(14)

Consequently, in the case of a steep resonance, we expect the stop band to approximately coincide with a frequency band of negative effective mass density. The real and imaginary parts of \(\rho_{\text{eff}}\) are plotted as functions of frequency, respectively, in Figs. 8(a) and 8(b) for several values of \(\phi\) and for three cases: (i) absence of relaxation, (ii) elastic relaxation only, (iii) elastic and viscoelastic relaxation. The values chosen for the mechanical properties of the materials are, here, also those of TC-epoxy dispersion. As expected, in
absence of relaxation, \( \rho_{\text{eff}} \) is purely real and actually displays a frequency band of negative effective mass density. Taking into account elastic relaxation considerably smooths the variations of \( \rho_{\text{eff}} \). In particular, the frequency band of negative mass density disappears, i.e., \( \text{Re}[\rho_{\text{eff}}] \) is positive for all frequencies. Adding viscoelastic relaxation only slightly increases the frequency at which \( \text{Re}[\rho_{\text{eff}}] \) and \( \text{Im}[\rho_{\text{eff}}] \) are maximum and slightly decreases the maximum values of \( \text{Re}[\rho_{\text{eff}}] \) and \( \text{Im}[\rho_{\text{eff}}] \) compared to the case with elastic relaxation only. We conclude that the main cause of smoothing of the resonant behavior of the effective mass density of the TC-epoxy dispersion studied here is elastic relaxation.

Given these conclusions, the key phenomenon to be minimized for emphasizing the dipolar resonance is thus elastic relaxation around the resonance frequency, i.e., \( k_T R \) has to be minimized at \( f \sim f_{\text{res}} \). Given Eq. (5), \( k_T R = [3\sqrt{8\hat{\rho}} - 5]/[2(2\hat{\rho} + 1)] \) at \( f = f_{\text{res}} \), i.e., at resonance, \( k_T R \) depends only on \( \hat{\rho} \) and not on \( \hat{\mu} \). This is in agreement with the independence versus \( \hat{\mu} \) of the quality factor of the resonance as given by Eq. (6). Thus, the larger the mass density contrast between the particle and the matrix, the steeper is the dipolar resonance.

We nevertheless recall that these conclusions have been drawn using Foldy’s model. An open question is whether these conclusions hold in the frame of LCN model, which has been shown to precisely fit experimental data because it takes wave conversions into account. We note that there is presently no realistic model expressing the effective mass density in the case of an elastic matrix. Thus, investigation of the behavior of the effective mass density of such dispersions in which wave conversion noticeably affects acoustic propagation requires further theoretical developments.

V. CONCLUSION

Our experimental study of the acoustic transmission through random dispersions of rigid spheres in an elastic matrix in the vicinity of their subwavelength dipolar resonance, and its comparison with a recent model of effective propagation that fully accounts for the elastic nature of the matrix, have allowed us to unambiguously demonstrate that wave conversion has a noticeable influence on the propagation of coherent longitudinal waves through such composite materials. Analyzing the dynamics of the scatterers has allowed us to evidence that the main cause of smoothing of the dipolar resonance is elastic relaxation, i.e., the finite time required for the shear stresses associated to the translational motion of the scatterers to propagate through the matrix. According to Foldy’s multiple scattering model, we have shown that the only parameter that determines the dipolar resonance steepness is the particle to matrix mass density ratio. We have also shown that the effective mass density is affected by elastic relaxation in the same way as the dipolar resonance. This leads us to refine the benchmarking of rigid particles in an elastic matrix against dense core-soft shell particles regarding their ability to display negative effective mass density. We note that a possible asset of core-shell particles is a lesser impact of elastic relaxation on their dipolar resonance as the consequence of the confinement of elastic deformations in the thin shell. The phase delay associated to the propagation of shear stresses over the shell thickness \( \epsilon \), \( k_T R \) is indeed expected to be less than \( k_T R \) provided that \( \epsilon < R \). But at the same time, the confinement of elastic deformations in the thin shell is expected to lead to larger viscoelastic dissipation as compared to rigid particles in an elastic matrix, where deformations extend over a distance on the order of \( R \). Thus, relaxation has an equivocal influence on the performances of both composite materials. This causes rather easy-to-make dispersions of rigid spheres in an elastic matrix appealing candidates as metamaterials.

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