# Ultrasonic Properties of a Suspension of Microspheres Supporting Negative Group Velocities

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The ultrasonic attenuation coefficient, phase velocity, and group velocity spectra are reported for a suspension that supports negative group velocities. The suspension consists of plastic microspheres with an average radius of 80 $\mu$m in an aqueous medium at a volume fraction of 3%. The spectra are measured using a broadband method covering a range from 2 to 20 MHz. The suspension exhibits negative group delays over a band near 4.5 MHz, with the group velocity magnitude exceeding $4.3 \times 10^8$ m/s at one point. The causal consistency of these results is confirmed using Kramers-Kronig relations.

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In highly dispersive media, the propagation of broadband ultrasonic pulses is marked by the severe reshaping of the pulse envelope. In describing waves undergoing dispersive propagation, up to five different velocities (i.e., phase, group, signal, front, and energy) have been defined, with each characterizing a specific aspect of the evolving pulse [1]. In any given situation, one or more of these velocities may not be clearly defined or physically meaningful. The most fundamental of these five is the phase velocity $c_p$ which, along with the attenuation coefficient, is a basic constituent of the acoustic complex wave number. When known for all Fourier components of a signal, the complex wave number can be used to predict the evolution of a pulse in a linear medium. The group velocity $c_g$ in a medium can almost always be defined in the mathematical sense, although its direct physical significance or utility is not always apparent. The group velocity does have some intriguing properties, not least of which is the fact that it can take on both arbitrarily large and negative values. [Unusually large (e.g., $c_g \gg c_p$) or negative group velocities are collectively labeled ''abnormal''.] In this Letter, we report broadband measurements of the attenuation coefficient, phase velocity, and group velocity spectra for an aqueous suspension of narrowly sized (80-$\mu$m nominal radius) polymer microspheres. The group velocity spectrum includes a band of negative velocities near 4.5 MHz, and exceeds $4.3 \times 10^8$ m/s in magnitude at one point. This represents an increase of more than 5 orders of magnitude relative to ordinary ultrasonic velocities in water or the bulk plastic.

Propagation of acoustic waves in biphasic media composed of random or periodic arrangements of inclusions in fluids (e.g., suspensions or phononic crystals, respectively) has been of interest in recent years due to the wide array of physical phenomena that such systems make accessible [2–7] (e.g., diffuse and coherent multiple scattering, acoustic band gaps, phonon tunneling). In suspensions, the ballistic/coherent component of the transmitted field carries the imprint of the elastic properties of both the scatterers and the host medium while maintaining phase coherence across various random arrangements of inclusions. With a fluid host, the coherent component (which includes the ballistic contribution [3]) can be efficiently extracted by averaging signals over a number of spatial distributions via agitation [8,9]. It is this coherent component that is the basis of the measurements reported here. The suspension used in this work has a solids volume fraction of 3%, which was sufficient to generate the strength of dispersion necessary to yield significant negative group delays. Calculations based on a theoretical model indicate that this system could reach the negative group velocity threshold at volume fractions as low as 2%. This work confirms earlier predictions that microsphere suspensions of this type would exhibit superluminal velocities at moderate volume fractions [10].

The existence of spectral bands exhibiting abnormal group velocities is a general feature of wave propagation in dispersive media. With electromagnetic waves, superluminal (i.e., $>c$, the speed of light) and negative group velocities are well known and of recent interest in the study of negative-index metamaterials [11,12]. The idea that group velocities of light pulses could exceed $c$ dates back at least to the work of Sommerfeld in the early 20th century [1], although it was not clear at the time that such velocities would be physically meaningful. Eventually, it was shown that superluminal group velocities can have a physical manifestation as a pulse velocity [13] while remaining consistent with relativistic causality. The term superluminal implies that the time interval marking the appearance of a point-of-reference at two distinct locations is less than the time it would take light to go the same distance in vacuum, and thus negative delays (and negative speeds) are naturally included in this interpretation. Since 1982 superluminal group velocities have been observed in a variety of systems [11,12,14–18]. In acoustics, the observation of superluminal group velocities for acoustic waves in the sonic region of the spectrum in several isolated bands between 300–3000 Hz has recently been reported [19].

*Theory.*—The propagation of an acoustic wave packet across a passive linear medium can be described by a transfer function of the form $H(\omega, \Delta z) = \exp[iK(\omega)\Delta z]$,

 

where $K(\omega) = i\alpha(\omega) + \omega/c_p(\omega)$ is the complex wave number, $\omega$ $(= 2\pi f)$ is the frequency, and $\Delta z$ is the thickness of the medium [20]. The complex wave number describes the dispersive properties of the system and is defined in terms of the phase velocity $c_p(\omega)$ and attenuation coefficient $\alpha(\omega)$ [21]. (An ergodic suspension of spheres can be considered statistically homogeneous and isotropic; $K(\omega)$ then represents the suspension's properties in the ensemble-averaged sense [22].) If $\mathrm{Re}K(\omega)$ is expanded in a Taylor series, the first order expansion coefficient defines the group velocity

$$\frac{1}{c_g(\omega)} = \frac{1}{c_p(\omega)}\left(1 - \frac{\omega}{c_p(\omega)}\frac{dc_p(\omega)}{d\omega}\right). \quad (1)$$

For propagation in dispersive materials, the physical significance of the group velocity is not always clear. With some restrictions, it has been shown that $c_g(\omega)$ is the envelope velocity of a Gaussian-gated continuous wave [13]. Calculations suggest that this physical manifestation of $c_g(\omega)$ as the speed of a peak holds for many singly peaked, smoothly enveloped (e.g., Hann windowed) narrowband signals. The key condition is that the bandwidth of the signal must be sufficiently narrow that the wave number can be accurately represented by its 2nd-order Taylor expansion over that frequency range. Under these conditions the temporal peak of the Gaussian-like modulation envelope of the signal will effectively propagate at the group velocity, although it may only be observable over a limited depth.

Derived from Eq. (1), the condition for the existence of superluminal group velocities is $dc_p/d\omega > (c_p/\omega) \times (1 - c_p/c)$. Since $c \gg c_p$, this is functionally equivalent to the negative velocity condition $dc_p/d\omega > c_p/\omega$. Note that in (1) the relation specifically defines the inverse group velocity, also known as the slowness $s_g(\omega) \equiv 1/c_g(\omega)$. This quantity is equivalent to the group delay time per unit depth of propagation and is referred to in this work simply as the group delay. As discussed later, the delay is a more natural parameter for describing the negative velocity regime.

*Materials and methods.*—The microspheres (Duke Scientific Corporation, Palo Alto, CA) studied in this work are made of polystyrene divinylbenzene and packaged suspended in an aqueous solution. The nominal radius of the microspheres is 80 $\mu$m and the volume fraction of microspheres was 3%. The suspensions were contained in a custom designed sample chamber and were agitated during acquisition via pipette mixing, a method shown to be effective in obtaining reliable data from suspensions [8,9].

The broadband ultrasonic spectroscopy technique uses the Fourier transforms of time-localized signals to derive material parameters over a broad range of frequencies simultaneously [20]. In this work, the various spectra were determined from 2 to 20 MHz ($ka = 0.67$ to 6.7) using the Fourier spectra of the through-sample and reference signals $[F_s(\omega) = |F_s|\exp(i\phi_s)$ and $F_{\mathrm{ref}}(\omega) = |F_{\mathrm{ref}}|\exp(i\phi_{\mathrm{ref}})$, respectively]. The attenuation and phase velocity determinations are discussed in Ref. [9]. The group velocity relation is

$$c_g(\omega) = \left(\frac{1}{c_{g,\mathrm{ref}}} - \frac{1}{h}\frac{d\Delta\phi}{d\omega}\right)^{-1} \quad (2)$$

where $\Delta\phi = \phi_s - \phi_{\mathrm{ref}}$, $f = \omega/2\pi$ is the frequency, $h$ is the sample chamber width (measured ultrasonically), and $c_{g,\mathrm{ref}}$ is the velocity in the chamber fluid with all the spheres settled out. The phases were unwrapped using a time shifting method that minimizes the linear term in the phase. Once the phase curves are unwrapped they are compensated for the introduced time offsets. The data were acquired using a through-transmission immersion system with two coaxially aligned poly(vinylidene fluoride) transducers in a water bath. The transmitter was excited by a broadband pulse and the received signals were amplified and digitized (Gage Applied Technologies CompuScope 12400, run in 12-bit mode at $200 \times 10^6$ samples/s). During each acquisition, 4096 time traces of through-transmitted ultrasound were captured. The signal acquisition steps used in the substitution method are illustrated in Figs. 1(a) and 1(b), and a diagram of the setup is shown in Fig. 1(c). Through-sample and reference waveforms are shown in Figs. 1(d) and 1(e). The 25 175 point signals were Hann windowed before FFT analysis $(\Delta\omega/2\pi = 0.0079$ MHz). The derivative in the group velocity relation was approximated by a finite-differencing method that is accurate to order $(\Delta\omega)^2 d^3\Delta\phi/d\omega^3$.

*Results.*—The attenuation coefficient and phase velocity spectra are shown in Fig. 2. In the region near 4.5 MHz, the respective spectra exhibit a narrow, sharply peaked attenuation band and a rapid dispersive sweep. The attenuation coefficient has nine peaks and varies by a factor of 40 from just below 0.03 $\mathrm{mm}^{-1}$ to 1.07 $\mathrm{mm}^{-1}$. The phase velocity

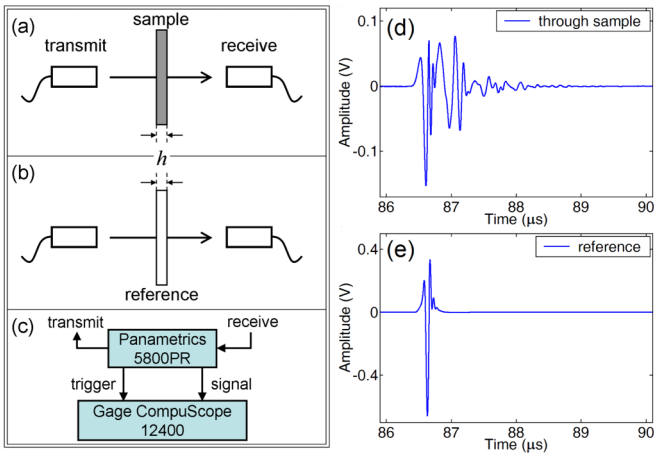


FIG. 1 (color online). (a) The through-sample and (b) reference signal acquisition steps of the substitution method. (c) Diagram of the measurement system. (d) The through-sample and (e) reference waveforms (4096 averages).

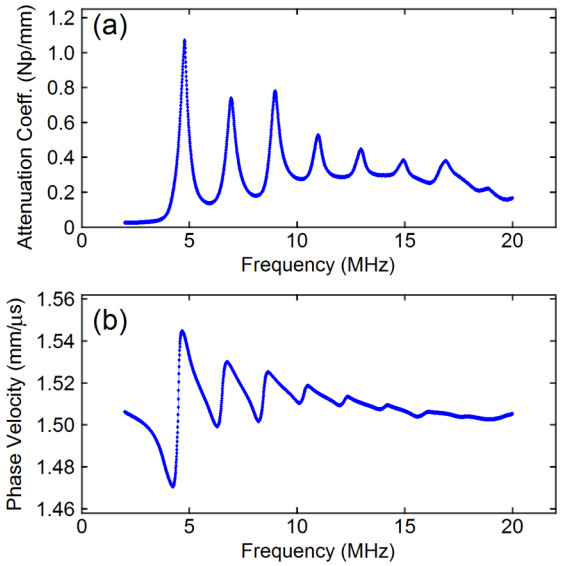


FIG. 2 (color online). (a) The attenuation coefficient and (b) the phase velocity spectra for the microsphere suspension.

exhibits a variation of 74 m/s through the low frequency attenuation band. The group delay spectrum is shown in Fig. 3, and the negative delay band is shown in detail in Fig. 4. In Fig. 4(b), the magnitude of the group velocity is plotted logarithmically. Note that the most negative delays in Fig. 4(a) correspond to velocities of smaller magnitude in Fig. 4(b), illustrating how velocity varies inversely to the intensity of the dispersive effect in the negative band. The largest velocity in terms of magnitude is $4.34 \times 10^5$ mm/$\mu$s which is about 1.4 times the speed of light in vacuum. Note that the delay curve crosses zero, indicating that with sufficient frequency resolution this system could exhibit any arbitrarily large value as it diverges at these crossover points. This is an indication that the group delay, not the group velocity, is the appropriate parameter to use for describing the abnormal regime. First of all, the divergence of the velocity at the zero-crossing points in the delay gives the impression that there is some abrupt physical discontinuity at these points and this is not the case. With the delay, the zero-crossing points are merely part of a continuum which is consistent with the physical manifestation of the dispersive effects. Secondly, where the dispersive effects are strongest, the delay exhibits its greatest deviations from the baseline while the velocity actually gets smaller in magnitude.

*Discussion.*—The highly dispersive behavior observed in this suspension results from resonances in the scattering amplitudes of the individual microspheres in tandem with the relative uniformity of the radii across the microsphere population. The band exhibiting negative delay around 4.5 MHz corresponds to a mode with a moderate $Q$ factor of 10. An experimentally validated theoretical model of these suspensions [8,9] indicates that the ratio of the scattering-to-geometric cross sections for the individual spheres varies from a low of 0.1 at 2 MHz to a maximum of 3.8 at 4.5 MHz. In an earlier report, this model predicted that negative group delays would occur in these suspensions at moderate volume fractions [10], a result confirmed by these measurements. Model-based calculations indicate that the suspension should reach the negative delay threshold at a volume fraction of about 2%. It is not obvious that the suspension would exhibit such dispersive strength, especially considering the moderate level of mechanical contrast between the host and the spheres and the modest volume fraction of solids (3%). The attenuation bands are wide enough to be straightforward to locate experimentally, yet still display an order of magnitude in variation over a moderate spectral region. This system may therefore have utility for studies of phenomena associated with propagation in random media, especially when signals with moderate-to-large fractional bandwidths are of interest. Its strongly dispersive effects are of potential interest for studies of broadband pulse propagation with possible

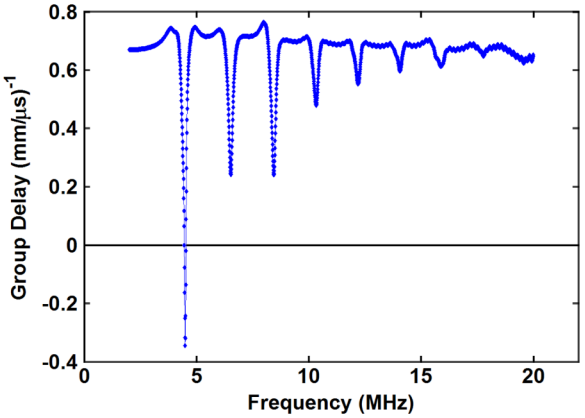


FIG. 3 (color online). The group delay spectrum for the microsphere suspension. The negative delay band runs from 4.45 to 4.53 MHz.

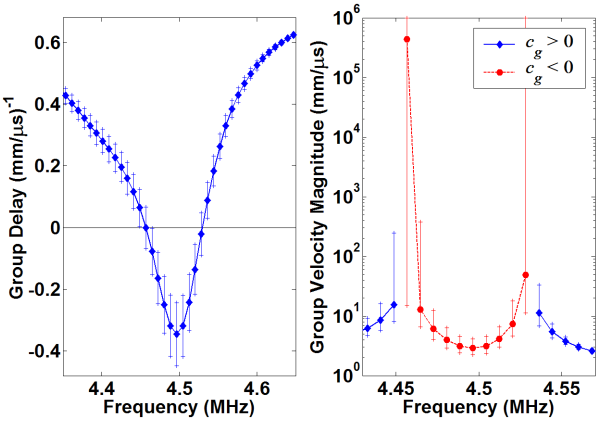


FIG. 4 (color online). (a) The group delay spectrum, zoomed in to resolve the negative delay band. (b) The group velocity magnitude in the abnormal region plotted logarithmically. This is the absolute value of the inverse of the delay curve. The maximum velocity magnitude is $4.34 \times 10^5$ mm/$\mu$s at 4.46 MHz.

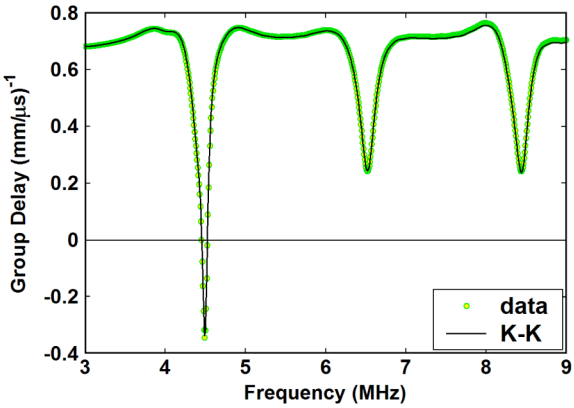


FIG. 5 (color online). A comparison of the measured group delay with the predicted spectrum from Kramers-Kronig relations. The plot is zoomed to resolve the negative delay band.

applications to biomedical imaging with contrast agents and wideband microwave systems.

*Causal Consistency of Results.*—The measurements in this Letter were performed using broadband pulses (i.e., bandwidths large in comparison with the attenuation bands in the data). Consequently, there are no directly observable time-domain features in these signals that indicate that the group delay curve has gone negative. The implication of a negative delay is that some reference point in a waveform reaches a more distant observer before an observer closer to the source. Such delays can be directly observed in experiments using the properly constructed waveforms with sufficiently narrow bandwidths as detailed in earlier work [10,19]. The compatibility between negative group delays and causality has been widely noted, and is commonly attributed to the ''pulse reshaping'' effect. (For a description of the time-domain manifestation of this effect see Ref. [10]).

Kramers-Kronig (KK) relations provide a quantitative means of verifying the mutual consistency of these attenuation and dispersion data. KK relations are the direct consequences of the simple causality, that a system cannot respond to a stimulus before the stimulus has occurred. In earlier work, a finite-bandwidth approximation of the KK relations for the direct determination of the group velocity in terms of the attenuation coefficient has been shown to provide accurate predictions for data from suspensions [21,23]. The comparison of the KK prediction and the measured group velocity spectrum is shown in Fig. 5. The strong agreement is readily apparent, verifying the consistency of these results as required by KK in accordance with causality.

In summary, the linear transmission properties of a 3% suspension of polymer microspheres supporting abnormal group velocities have been reported. Using a broadband technique, the transmission spectra were measured over a bandwidth from 2 to 20 MHz, with the group velocity entering the negative delay regime over an 80 kHz band near 4.5 MHz. Extremely large group velocities were found at the edges of the negative band, although the group delay $(1/c_g)$ is the more appropriate parameter to use in the abnormal regime. The causal consistency is quantitatively verified using Kramers-Kronig relations. This system may have utility for studies of phenomena associated with coherent and diffuse propagation in random media, with potential relevance for medical imaging and wideband microwave applications.

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