Test of Hyperelasticity in Highly Nonlinear Solids: Sedimentary Rocks

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We report measurements of three-wave mixing amplitudes on systems whose third order elastic constants have also been measured by means of the elastoacoustic effect. Because attenuation and diffraction are an important aspect of our measurement technique we analyze our results using a modified Khoklhov-Zabolotskaya-Kuznetsov equation in the frequency domain. We find that the value of β so deduced for polymethyl methacrylate agrees quite well with that predicted from the stress dependent sound speed measurements, establishing that polymethyl methacrylate may be considered as a hyperelastic solid. The β values of sedimentary rocks, though they are typically 2 orders of magnitude larger than, e.g., polymethyl methacrylates, are still a factor 3–10 less than those predicted from the elastoacoustic effect.

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The most commonly investigated nonlinear acoustic properties of a liquid or a solid are: (a) the extent to which the speed(s) of sound change upon the application of a known stress [elastoacoustic (EA) effect] and (b) the extent to which two different initial frequencies, f_1 and f_2, mix to generate frequency components $f_1 - f_2$, $f_1 + f_2$, $2f_1$, and $2f_2$ [three-wave mixing (3WM)]. Within the context of hyperelasticity, both these processes are governed by the same nonlinear material parameters called third order elastic (TOE) constants if one is dealing with a solid. An isotropic solid has three such coefficients, A, B, and C. These effects are described in Ref. [1]. A straightforward discussion of the underlying theory is given in the chapter by Norris (p. 263) who makes connection with other systems of notation.

Apparently a test of the prediction of three-wave mixing efficiencies based on values of A, B, and C deduced from the elastoacoustic effects has only recently been done on any solid [2] and this is the focus of the present Letter. As a practical matter we are particularly interested in these effects in sedimentary rocks, which are known to have values of the TOE constants that are orders of magnitude larger than those of their constituent minerals [3]. In the search for hydrocarbon reservoirs it is often most useful to have some knowledge of the local distribution of stress around a borehole. Local stress distribution affects most aspects of hydrocarbon production including drilling, perforating, casing, and sanding during production. Because rocks are so sensitive to applied stress, one can often deduce, e.g., the direction of maximal horizontal stress by means of the effect on borehole acoustic modes of propagation; see Refs. [4]. If one had an independent means to measure the TOE constants, such as by three-wave mixing, one could deduce the actual stress magnitudes in addition to their directions. Unfortunately sedimentary rocks, while being extremely nonlinear, are also extremely attenuative; this makes it difficult to quantify the amplitudes of nonlinearly generated signals.

We have done the following: on a sample of polymethyl methacrylate (PMMA), a commercially available nonporous plastic, and of three different sedimentary rocks we have overdetermined the three parameters A, B, and C by measuring the rate of change of five sound speeds with applied uniaxial and hydrostatic stress. We chose PMMA partly because of convenience and partly because we can make the effects of attenuation in the 3WM experiments as large as those in the rocks (see below).

Using a water immersion technique with a slab of each material we have independently measured the (linear) attenuation in each sample as a function of frequency from 100 kHz to 2 MHz. We have measured the efficiency of sum and difference frequencies, etc. generated by the insonification of the sample with a collimated beam having very narrow band pulses at $f_1 = 0.95$ MHz and $f_2 = 1.05$ MHz. Because the attenuation at frequencies in the vicinity of 2 MHz is so large for the rocks, we have focussed on the difference frequencies for them, but we have quantified all four nonlinear signals for the PMMA sample. In order to handle the simultaneous effects of nonlinear acoustics, attenuation, and dispersion we have analyzed our nonlinear results using a modified Khoklhov-Zabolotskaya-Kuznetsov (KZK) equation. The nonlinearity of this KZK equation enters via a dimensionless parameter, β, which is related to A, B, C in hyperelastic materials by [1]

$$\beta = -\left[\frac{3}{2} + \frac{A + 3B + C}{K + (4/3)\mu}\right]. \quad (1)$$

We find that all four nonlinearly generated signals in PMMA are accurately described by the same value of β as that deduced from the elastoacoustic measurements, thus indicating that the deformation energy of PMMA is hyperelastic to third order in the strain. In all three

samples of the sedimentary rocks we find values of β that are typically a factor 3–10 smaller than those predicted from the elastoacoustic measurements, but still 2 orders of magnitude larger than those encountered for homogeneous solids.

The relevant material parameters of our four samples are listed in Table I. We chose one limestone and two sandstones and the porosities range from 0.18 to 0.23. The sound speeds were measured in a time-of-flight technique through a rectangular parallelepiped under ambient, hydrostatic, and uniaxial stress, as described previously [3]. A simple curve fitting algorithm allows us to extract the slopes $\frac{dV}{d\sigma}$ where σ is either the hydrostatic or uniaxial stress, as the case may be. We have so-measured five such slopes that overdetermine the three TOE constants, as described previously [3]. Our best estimates of β(stress) are given in Table I.

We performed our three-wave mixing measurements in water using a high-power source transducer and calibrated receivers in a tank approximately 2 m $\times$ 1 m $\times$ 1 m deep. The source signal comprised the beating of two 50 cycle tone bursts, one at $f_1 = 0.95$ MHz and the other at $f_2 = 1.05$ MHz. The drive signal (f_1 and f_2) voltage was varied in precisely known steps, such that the transducer face pressure ranged from approximately 1 to 1000 kPa. The source transducer was an air-backed, 1.0 MHz, 1.0 in diameter, planar-piston transducer.

We positioned receivers at a distance of 203 mm from the source face, on axis with the transmitter beam. One receiver was a B&K 8103, used for measuring the difference frequency (100 kHz). The other was a 0.4 mm Sonic Technologies membrane-hydrophone probe, used for all of the measurements around and above 1 MHz. Both receivers were calibrated by the manufacturers in Pa over wide frequency ranges.

With our calibrated receivers in a water-only setup ($\beta = 3.5$) we used the KZK calculations (below) to calibrate the transmitter pressures at each drive voltage. We did this by quantifying the axial and radial variations of all received signals.

We used standard immersion techniques to derive the linear velocity-dispersion and attenuation-dispersion curves for each sample, which we measured over as wide a frequency range as possible, depending on the sample. We measured the sample densities. The samples were then located in the beam of the high-power transmitter, typically at a standoff of 50.8 mm from the transmitter face. We varied the drive signal (f_1 and f_2) voltage in the same steps as for the water-only test, recording the received tone bursts for both high and low frequency receivers. The received signals were then processed into calibrated pressure values at each pertinent frequency, as above.

In order to interpret our nonlinear measurements, we need a theoretical tool that incorporates nonlinearity, diffraction, attenuation, and reflection losses at the various interfaces. We used the modified KZK equation for this purpose [1]:

$$\frac{\partial^2 p}{\partial z \partial \tau} + \frac{\partial F}{\partial \tau} - \frac{V}{2}\nabla_\perp^2 p = \frac{\beta}{2\rho V^3}\frac{\partial^2 p^2}{\partial \tau^2}. \tag{2}$$

Here, $p(r, z, \tau)$ is the acoustic pressure, V is the linear speed of sound, ρ is the density, and β is the nonlinear parameter given by Eq. (1). The linear frequency dependent attenuation coefficient, $\gamma(\omega)$, is incorporated in the quantity F, which is conveniently written in the frequency domain as

$$\tilde{F} = \gamma(\omega)\tilde{p}. \tag{3}$$

If $\gamma \propto \omega^2 \leftrightarrow F \propto \frac{\partial^2 p}{\partial \tau^2}$, one recovers the usual KZK equation appropriate to a viscous fluid. Equation (2) is solved in the frequency domain by expanding the solution as a Fourier series, as described in Refs. [5]. The code was modified by us to include discontinuities of material properties and to allow for an arbitrary frequency dependence for the attenuation, $\gamma(\omega)$. The code accounts for reflection losses at boundaries but does not account for multiple reflections, which, in fact, are negligibly small for our attenuative samples.

Although we list attenuations at three frequencies in Table I, we have, in fact, used the actual attenuation values for each frequency component that we measured. The attenuation of water is quite negligible at all relevant frequencies, for our purposes. The attenuation values at 100 kHz for all the solid samples, though not negligible, are small enough that accuracy of those values is not an issue (see below).

First, we consider the sample of PMMA. We have found that the most sensitive determination of β is obtained by using a rather thick sample (6 in) immediately facing the

TABLE I. Material properties. $\gamma_{1,\Delta,2}$ refer to the measured attenuation at 1 MHz, 100 kHz, and 2 MHz, respectively. β for water is taken from Ref. [1]. A, B, C are determined from stress dependence of the acoustic speeds.

	ρ(gm/cc)	V(km/s)	γ_1(1/m)	γ_Δ(1/m)	γ_2(1/m)	$-A$(GPa)	$-B$(GPa)	$-C$(GPa)	β(stress)	β(3WM)
Water	1.0	1.48	2.5×10^{-2}	2.5×10^{-4}	1.0×10^{-1}	...	...	...	...	3.5
PMMA	1.19	2.72	12.4	1.55	23.0	22 ± 6	18 ± 3	19 ± 3	9 ± 2	7.1 ± 0.5
Portland	2.33	3.47	64.0	6.25	129	2362 ± 147	1310 ± 106	1846 ± 210	332 ± 28	60–150
Indiana	2.40	3.89	77.0	0.89	295	6559 ± 313	$11\,715 \pm 286$	9377 ± 644	1201 ± 43	400–525
Berea	2.26	2.45	288	14.3	711	-2234 ± 395	4939 ± 269	8787 ± 517	1155 ± 93	170–670

transmitter. This is because the β value is not particularly large compared to that of water and we therefore wish to maximize the path length through PMMA. In Fig. 1 we plot the measured amplitudes of the received signals comprising the original components f_1 and f_2 as well as the nonlinearly generated ones at $f_2 - f_1 = 100$ kHz, $2f_1 = 1.9$ MHz, $f_1 + f_2 = 2.0$ MHz, and $2f_2 = 2.1$ MHz. In this geometry we do not know the extent to which the sample loads the transmitter. Accordingly, we plot the measured signals vs the received f_1 signal amplitude, and not vs the initial amplitude on the transducer face.

We also plot the results of our solution to Eq. (1) using a value of $\beta = 7.1$ for PMMA. We see that it does an excellent job of describing all four nonlinear signals over a wide range of amplitudes. Over the range of amplitudes considered, the behavior of the two fundamentals is governed primarily by linear acoustics; the nonlinear signals are proportional to the square of the initial amplitude, as can be seen from the plot. We have repeated the calculation for a narrow range of β values in order to find what value best fits each nonlinear signal. This range is reflected in the error bars quoted in Table I. Our value of $\beta = 7.1$ is to be compared to the value $\beta = 10$ deduced by Landsberger and Hamilton [6] from the measured amplitude of second harmonic generation in a similar geometry and the value $\beta = 6$ deduced from earlier elastoacoustic measurements (Winkler and Liu [3]).

In order to emphasize the role of attenuation and diffraction, we plot the computed amplitudes of all relevant waves as a function of distance along the z axis, in Fig. 2. We see destructive interference due to diffraction effects in the fundamental (f_1 and f_2) components as well as in the higher harmonics within the first 5 cm of the sample. We see a large decrease in the amplitudes due to attenuation; we have plotted the plane wave attenuation coefficients $\exp[-\gamma z]$ for $f = 1$ MHz, $f = 2$ MHz, and $f = 100$ kHz as guides to the eye. We see the discontinuity in the transmitted amplitudes at the PMMA-water interface.

The value determined from these three-wave mixing measurements, $\beta(\mathrm{3WM}) = 7.1$, is in substantial agreement with that determined from the stress dependence of the sound speeds, $\beta(\mathrm{stress}) = 9$. We draw the following conclusions: (1) through third order in the strain the deformation energy in PMMA is well described by standard hyperelastic theory. (2) Our experimental techniques for determining β both from the stress dependence of the sound speeds and from the three-wave mixing measurements are accurate. (3) The modified KZK equation, Eq. (1), accurately describes the combined effects of nonlinearity, diffraction, attenuation, and the amplitude loss at the PMMA-water interface. It would be unlikely if the agreement between theory and experiment would hold for all four nonlinear signals if these three conclusions were not true. As far as we are aware, this represents the first such confirmation of this aspect of the hyperelastic hypothesis in a solid.

Armed with this confidence in our technique, we turn to the investigation of the sedimentary rock samples. Here, the samples are much more nonlinear, having β values 1–2 orders of magnitude larger than that of PMMA, and they are much more attenuative. Thus, the nonlinearly generated signals are created, not throughout the sample thickness, but primarily within the first decay length or so of the fundamentals; this means the attenuation coefficient, γ, of the fundamentals must be known

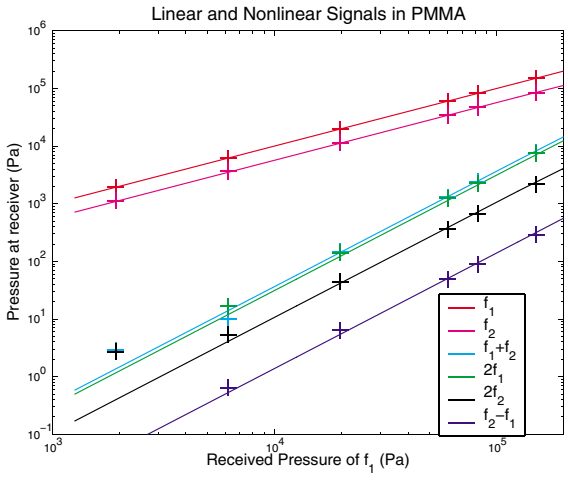

FIG. 1 (color). Amplitudes of the two fundamental frequencies, f_1 and f_2, as well as those of the nonlinearly generated signals plotted against the received amplitude of f_1. The acoustic path is 146 mm of PMMA and less than 1 mm water. The symbols represent the measured data and the solid curves are the solutions to the KZK equation with $\beta = 7.1$ for PMMA.

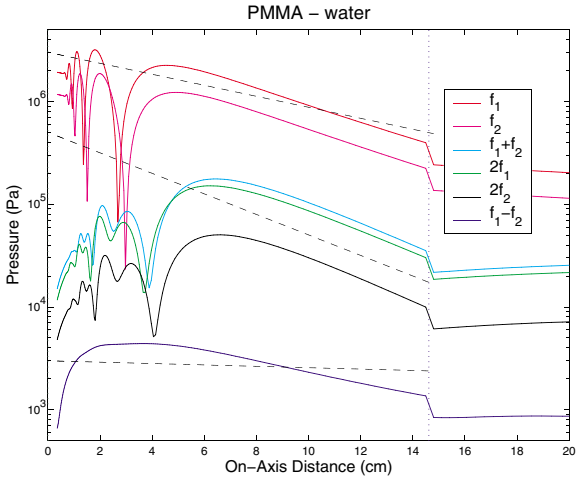

FIG. 2 (color). Computed amplitudes of the relevant frequency components of Fig. 1 on the z axis from the transmitter to the receiver. The dashed lines represent linear decay at 1 MHz, 2 MHz, and 100 kHz, from top to bottom. The vertical dotted line is the PMMA-water boundary

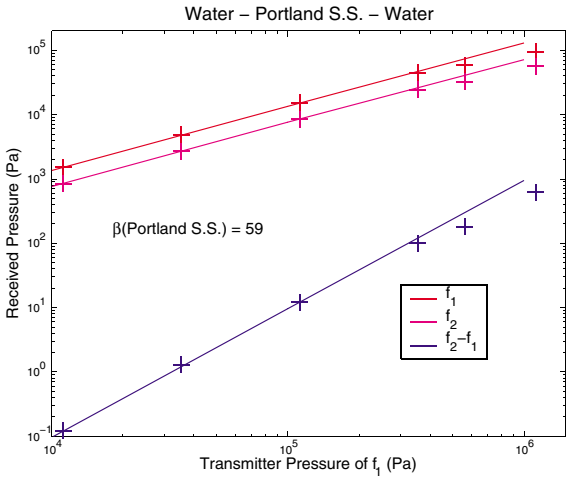

FIG. 3 (color). Amplitudes of fundamentals and of difference frequency in Portland S.S. with an acoustic path as indicated.

quite accurately. Because γ_1 (Portland, Indiana) $\approx 6\gamma_1$(PMMA), the total attenuation in the 6 in of PMMA is comparable to the total attenuation in 1 in of both the Portland and Indiana samples, $\exp(-\gamma_1 L) \approx 0.15$. This fact makes PMMA a good nonrock with which to test our system and compare the results against those of the rocks.

Even though we use rock samples only ≈ 1 in thick, we are unable to quantify the nonlinear signals near 2 MHz due to high attenuation in the 1–2 MHz region. Accordingly, we focus our attention exclusively on the difference frequency generation, $f_2 - f_1 = 100$ kHz. Here, the decay rate, $\gamma(100\ \text{kHz})$, is small enough that the KZK calculation is relatively insensitive to its actual numerical value. This is the motivation as to why we have looked at the difference frequency generation for these attenuative samples.

Our resultant three-wave mixing values are listed in Table I as β(3WM). We note that all three rock samples have values that are large compared to that in PMMA, but still a factor 3–10 less than those implied from Eq. (1), β(stress). The range of values listed represents values obtained from different sample thicknesses, different standoffs from the transmitter, as well as fits to data at different transmitter face pressures. For example, in Fig. 3 we show results for the Portland Sandstone. Although the difference signal initially grows quadratically, while the fundamentals are still approximately linear, we see that there is significant deviation between theory and experiment above 3×10^5 Pa. The amplitudes of the fundamentals are no longer linearly related to the initial amplitudes; the amplitude of the difference signal is no longer quadratic in the initial amplitude. Unlike the case of the PMMA data in Fig. 1 there is no single value of β that will fit all the Portland data, and similar behavior holds for the other rock samples. We consider it to be a manifestation of the breakdown of the validity of the KZK equation for these samples, and possibly the breakdown of the hyperelastic assumption, even for these small strains. It is quite possible that the deformation energy as a function of strain does not have the simple power-law behavior implicit in the conventional hyperelastic expansion. A specific example of this behavior does occur in the theoretical case where there are so-called Hertz-Mindlin contacts, for which $U \propto \epsilon^{5/2}$ [7]. It may well be the case that a new paradigm is needed to describe the nonlinear acoustics of sedimentary rocks [8], as is clearly the case for large, hysteretic strains.

In conclusion, we have demonstrated the validity of the KZK equation to understand three-wave mixing experiments in PMMA. The numerical value of β so determined agrees with that determined independently by means of the stress dependence of the sound speeds, establishing that PMMA may be considered hyperelastic in this context. Measurements on sedimentary rocks indicate a very different situation. The numerical values of β determined from the amplitude of difference frequency generation are approximately 2 orders of magnitude larger than in PMMA but still a factor 3–10 smaller than those implied by the stress dependence of the sound speeds.

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