Short-Period Nonlinear Viscoelastic Memory of Rocks Revealed by Copropagating Longitudinal Acoustic Waves

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Abstract We describe a new sensitive acoustic method to measure nonlinear viscoelastic properties of rocks directly in the time domain. Unlike static stress-strain measurements, or dynamic acousto-elastic methods that use steady state resonance to load a strain field into a rock, our method uses two copropagating longitudinal acoustic waves, one to perturb the strain in the sample and the other to probe the effect of that strain. Experiments on Crab Orchard Sandstone and Lucite samples are presented. We evaluate the time-dependent nonlinear effects and find strong evidence that rocks have a short-period nonlinear viscoelastic memory that is a function of the time history of the loaded strain. We develop a phenomenological model to describe both this nonlinear viscoelastic memory and the nonlinear elastic behavior of rock. Our model shows that the viscoelastic memory is controlled by both the traditional nonlinear elastic coefficients and a memory strength parameter. These new observations and methods have significance for quantifying changes in microstructure. This short-term memory effect, on the order of a small fraction of a cycle, is shown to be related through viscoelasticity to absorption in the sample.

Plain Language Summary By using propagating ultrasound waves to probe rocks, we have developed measurement methods to characterize very small changes in the stiffness of rocks in response applied dynamic strains. When strained, rocks become softer and have a short-term memory. Our sensitive methods reveal that rock microstructure is responsive to environmental conditions such as overload and fluid content. These new approaches may allow earth scientists to better understand earthquake fault zones or sense environmental problems such as fluid leaks in near-surface reservoirs.

1. Introduction

It is well known that rocks are highly nonlinear (Johnson et al., 1996). Since the nonlinear elastic response of rocks has been shown to be caused by the rock's microstructure (Darling et al., 2004), measurements of the nonlinear response of a rock may help us to better characterize its microstructure and to identify differences among rocks.

Unlike previous work using the dynamic acousto-elastic method that uses acoustic resonance to impart strain (e.g., Renaud et al., 2012) and our earlier work with shear waves (Gallot et al., 2015; TenCate et al., 2016), we use copropagating longitudinal acoustic waves to load a strain field and investigate the nonlinear characteristics of rock. The method involves sending two copropagating longitudinal waves into the rock and receiving them as illustrated by Figure 1. One of the waves is a high-energy low-frequency (LF) longitudinal wave (the pump) that loads a temporally varying strain field in the rock, and the other is the low-energy high-frequency (HF) longitudinal wave (the probe) that is used to detect the elastic modulus variation induced by the loaded strain. Because the pump wave frequency is much lower than the probe frequency, we refer to the imposed strain field as slowly time varying for purposes of this paper. This dynamic strain field offers a way to study the evolution of the elastic modulus variation between different strain cycles of the pump waveform directly in the time domain.

A nonlinear version of Hooke's law can be expressed using a power series expansion (TenCate et al., 1996; VandenAbeele, 1996),

\[ \sigma = E\varepsilon + \beta E\varepsilon^2 + \delta E\varepsilon^3 + O(\varepsilon^4), \]  

(1)
where $E$ is elastic modulus and terms $\beta$ and $\delta$ are the quadratic and cubic nonlinear elastic coefficients, respectively. Without considering the fourth-order error, the generic nonlinear elastic modulus $M$ can be written as follows:

$$M = E + \beta E \varepsilon + \delta E \varepsilon^2.$$  \hfill (2)

Therefore, the elastic modulus variation with strain is

$$\Delta M/E = (M - E)/E = \beta \varepsilon + \delta \varepsilon^2.$$  \hfill (3)

However, dynamic acousto-elastic experiments conducted on rocks have shown that the elastic modulus variation is at least a second-order polynomial function of strain (Renaud et al., 2011, 2012; Renaud, Riviere, Haupert, et al., 2013; Renaud, Riviere, Le Bas, et al., 2013). Therefore, in order to fit the data, an additional term $C$ must be included:

$$\Delta M/E = C + \beta \varepsilon + \delta \varepsilon^2.$$  \hfill (4)

Unlike previous work on resonant steady state configurations, we have extended this model to freely propagating, general time-varying strain waveforms by making the $C$ term dependent on the loading waveform time history as well as the maximum pump strain level. In section 3, we will show that this new time-dependent term describes observed short-term memory effects. In addition, our experiments, which load a slowly time-varying strain field into rocks, show that $C$ also empirically describes an observed offset of the modulus variation that is due to nonlinear material conditioning (Guyer & Johnson, 2009; Renaud et al., 2011; Renaud, Riviere, Le Bas, et al., 2013; Riviere et al., 2015; TenCate, 2011).

In the last decade, models have been developed to capture the interplay between conditioning and relaxation processes in the field of nonlinear elasticity (Guyer & Johnson, 2009; Vakhnenko et al., 2005). To include both nonlinearity and viscoelasticity, Favrie et al. (2015) developed a phenomenological model in which nonlinearity was included through a "soft ratchet" mechanism and viscoelasticity through a generalized Zener model with multiple relaxation rates. Another way of combining nonlinear and viscoelastic methods is an empirical approach involving complex nonlinear coefficients at each frequency (Trarieux et al., 2014). Our methodology is also empirical; however, it differs from the previous works that focus on the use of continuous waves because our data are collected in the time domain and our model is therefore time domain based and includes viscoelasticity and causality (Szabo, 1995, 2014 Chapter 4).

We show results for two samples. One is a Crab Orchard Sandstone (Benson et al., 2005) sample from the Cumberland Plateau, Tennessee, USA. The other is a Lucite sample. In each case, measurements are made using several input voltages supplied to the pump, which result in different strain amplitudes in the samples that are measured using a laser vibrometer. We find that Lucite is much less nonlinear than the Crab Orchard Sandstone. We propose a phenomenological model that includes the classical nonlinear elastic effects in addition to a memory effect to explain our measurements on the sample of Crab Orchard Sandstone.

2. Experiment

2.1. Experiment Setup

Figure 1 shows the setup of the copropagating longitudinal acoustic wave experiment. A Crab Orchard Sandstone sample $15 \text{ cm} \times 15 \text{ cm} \times 5 \text{ cm}$ is used for the experiment. A pair of 1 MHz HF (T1 and R1) 12.7 mm diameter compressional ultrasound transducers centered at a distance of 8.5 cm from the top edge are mounted on opposite sides of the rock sample to generate and receive probe signals, respectively. The pump transducer, centered at a distance of 4 cm from the top edge of the sample, is a high-amplitude
compressional source that loads a relatively high strain into the sample through a 39 mm diameter 0.1 MHz LF (T2) transducer. The pump and probe source transducers are mounted side by side on the sample as shown in Figure 1. Ideally, we would prefer to have these two sources mounted coaxially on the sample; however, our set up provides an adequate first-order approximation to that configuration since the major component of the pump strain is in the x direction along the probe source-receiver direct path. The probe is a low-amplitude signal that is used to measure small differences in arrival time that are induced by the large strains imposed by the pump transducer. The pump beam overlaps the probe beam as shown in Figure 1 by the shaded areas representing the −20 dB amplitude contours of the pump and probe transducer radiation patterns, which show the amplitude as a function of angle. Although the pump and probe transducers were placed so as to minimize boundary effects, boundary reflections will arrive late and inverted in the time window of interest but with low amplitude relative to the direct arrival due to the radiation pattern of the P wave transducers (shaded areas in Figure 1) and the longer travel paths through an attenuating medium (see Appendix A). Waves generated by the pump and probe propagate at the same velocity. A waveform generator excites both a 74 kHz signal, which is amplified by a radio frequency power amplifier and sent to T2, and a 620 kHz signal, which is sent to T1. The choice of frequencies was made to maximize the amplitudes of the signals that were coupled into the sample while maintaining a large difference in frequencies between the pump and probe to facilitate measurements. We made measurements using five input voltage levels to the pump. Each voltage level gave differing maximum strain amplitudes in the sample. In subsequent discussions, we refer to measurements by the maximum strain measured in the sample. A three-component laser vibrometer (R2) measures the particle vibration velocity, \( v \), of the pump signal in the overlap region, which is used to estimate the strain, \( \varepsilon(t) \), induced into the sample (see Appendix A). Probe signals received by R1 are filtered using a 600 kHz high-pass filter to remove contamination by the pump signals. Then the probe signals are amplified. We measure probe time of flight (TOF) signals under two conditions at R1. One is the probe signal recorded when the pump signal is present (TOF). The other one is the probe signal in the absence of the pump signal (TOF\(_0\)). Comparing TOF and TOF\(_0\), we obtain a measure of the TOF delay, \( \Delta \text{TOF} \), which is used to calculate the elastic modulus variation induced by the pump.

### 2.2. Estimation of Elastic Modulus Variation

In our experimental setup, the triggers of the probe signal were sequentially delayed over a range of 0–43 \( \mu s \) with 1 \( \mu s \) increments relative to the pump trigger to allow the probe signal to interact with various segments of the positive and negative strains induced by the pump signal (see Appendix A for more details). For each probe trigger time delay, we measured probe signals with and without the presence of the pump signal. Figure 2 shows the two types of probe signals recorded by transducer R1 on the sample of Crab Orchard Sandstone. In the figure, the x axis shows the trigger delay time. Figure 2b, which zooms in the selected area of Figure 2a, clearly shows a TOF delay, \( \Delta \text{TOF} \), between the two types of probe signals that depends on the trigger delay time. Therefore, \( \Delta \text{TOF} \) can be easily measured by a cross-correlation algorithm. The elastic modulus variation, \( \Delta M/E \), is estimated as being proportional to the \( \Delta \text{TOF} \) induced by the application of the pump divided by the original non pump TOF\(_0\) (Renaud et al., 2012):

\[
\frac{\Delta M}{E} = -2 \frac{\Delta \text{TOF}}{\text{TOF}_0}.
\]  

where TOF\(_0\) (about 52.3 \( \mu s \)) is the TOF of the probe in the absence of the pump signal.

### 2.3. Observed Elastic Modulus Variation With Time-Varying Strain

Since the amplitude of a pump signal is time dependent, delaying the probe onset relative to the pump onset allows us to observe nonlinear behaviors for different strain amplitudes and pump-induced strain histories. The pump induces propagating packets of strain, and the probe wave rides within a particular pump-induced strain state along its path. Therefore, the relative trigger delay time of the probe to pump allows us to vary the strain and strain history that the probe experiences.

Figure 3a shows the average strain induced by the pump signal as a function of trigger delay time. In our experiments, the probe transducer (T1) trigger is delayed relative to the pump transducer (T2) trigger from 0 to 43 \( \mu s \) in increments of 1 \( \mu s \). We conducted the experiments using five different maximum input voltages (high voltage induces high strain) to the pump and obtained five strain curves that are displayed by solid color lines in the figure. Figure 3b shows the elastic modulus variation, \( \Delta M/E \), measured by the probe.
signals as a function of relative trigger delay time. Because five different pump input voltages were used, we obtained five curves of the elastic modulus variation displayed by solid color lines in the figure.

3. Short-Period Nonlinear Viscoelastic Memory

3.1. Elastic Modulus Variation as a Function of Dynamic Strain

Figure 4 shows the strain and modulus change as functions of trigger delay time for five of the pump input voltages, and Figure 5 shows the relation between modulus change and instantaneous strain. We display TOF delays using trigger delays corresponding to three cycles of pump waves where the pump amplitude increases with each cycle. These three cycles are identified by solid, dashed, and dotted curves in Figures 4 and 5. Demarcating these three cycles will be useful later in showing differences among models and in elastic constant transitions. In the figures, the black and red lines indicate results for the samples of Lucite and Crab Orchard Sandstone, respectively.

Our convention is that compressive axial strain is negative, while a positive strain represents tension. Figures 4 and 5 show that each cycle of the pump, having higher amplitude than the previous one, brings the rock to a new state. The relative compression and tension cause the modulus to oscillate about this state to form the hysteretic nonlinear behavior that is observed both in compression and in tension. We also observe that the relative maxima/minima of the elastic modulus variation are consistently delayed relative to those of the strain (see, for example, the maximum strain at a trigger value of 32 μs results in the

![Figure 2](image-url)  
**Figure 2.** (a) Probe signals measured by R1 when the pump input results in a maximum strain of $9.7 \times 10^{-7}$. The solid red lines and blue dashed lines indicate the probe signals in the absence and presence of pump signals, respectively. The trigger delay time is the time that the probe trigger is delayed relative to the pump onset. (b) Zoom of the selected area marked by green dotted lines in (a).

![Figure 3](image-url)  
**Figure 3.** (a) Average strain induced by the pump signals and (b) the elastic modulus variation measured by the probe signals as a function of the trigger delay time. The trigger delay time is the time that the probe trigger is delayed relative to the pump onset. Different solid color lines indicate different strain maxima that vary with the voltages applied to the pump transducer. The circle markers indicate the measurement points.
Figure 4. The strain and the elastic modulus variation, $\Delta M/E$ as a function of the trigger delay time. (a–f) Results for maximum strain of $4.7 \times 10^{-7}$, $6.0 \times 10^{-7}$, $7.7 \times 10^{-7}$, $8.8 \times 10^{-7}$, $9.7 \times 10^{-7}$, and $9.7 \times 10^{-7}$, respectively. In each subfigure, the top figure shows strain as a function of trigger delay time, and the bottom figure indicates the elastic modulus variation, $\Delta M/E$, as a function of the trigger delay time. Positive strain is defined as extensional and negative strain as compressional. The solid, dashed, and dotted lines indicate the first, second, and third cycles of the pump, respectively. The circles indicate the measurement points for the Crab Orchard Sandstone sample. The black and red lines indicate measurement results for the Lucite and Crab Orchard Sandstone samples, respectively. The blue lines indicate the fit of our model to the Crab Orchard Sandstone data using only the first two cycles of the data in (a–f), the fit of the model to the data using a constant (with respect to time) $C$ for each cycle in equation (4).
maximum modulus variation at 34 μs in Figure 4). Figure 5 depicts hysteretic nonlinear elasticity loops. Note that our hysteresis loops are much more asymmetric than the loops previously reported (Renaud et al., 2011, 2012; Renaud, Riviere, Haupert, et al., 2013; Renaud, Riviere, Le Bas, et al., 2013; Riviere et al., 2015) for sinusoidally driven resonant configurations, suggesting that the use of propagating waves provides additional hints into how the rock pore structure behaves differently under tension and compression. Trarieux et al. (2014) also found highly asymmetric viscoelastic hysteresis loops for air-filled glass beads whose behavior they believed was similar to highly nonlinear rocks. In Figure 5, the three hysteresis loops for each maximum strain level (i.e., for each pump voltage level) correspond to the first, second, and third cycles, respectively. The circles indicate the measurement points for the Crab Orchard Sandstone. The black and red lines indicate measurement results for the Lucite and Crab Orchard Sandstone samples, respectively. The blue lines indicate the fit of our model to the data using only the first two cycles of the data in (a–e); (f) shows the fit of the model to the data using a constant (with respect to time) C for each cycle in equation (4).
3.2. Key Indicators of the Short-Period Nonlinear Viscoelastic Memory

The hysteresis loops in Figure 5 show clearly that there is a consistent pattern of modulus change with strain that occurs over several pump cycles for a given pump input voltage and over all input voltages used in our experiments. We summarize the key indicators of short-period nonlinear viscoelastic memory of rocks that are illuminated by our experiments as follows:

- Rocks become softer: Figure 4 shows that the elastic modulus change has a general trend toward negative values; for example, the rock becomes softer independent of the sign of the applied strain. The trend becomes clearer with increasing pump input amplitude. When the pump input is sufficiently large the elastic modulus variation is always negative whether the strain is positive or negative. Earlier work (Johnson et al., 1996) with numerous different rock types shows a similar trend.

- Rocks have viscoelastic memory: There is a lack of instantaneous reaction of the modulus to changing strain indicating a memory effect that we attribute to viscoelasticity. The delay times where the peaks and troughs of $\Delta M/E$ occur are shifted slightly ($\Delta t$) toward later times compared to the peaks and troughs of the applied strain. This is consistent in all of our measurements. At the time of the maximum compression, the value of $\Delta M/E$ is near a local maximum; for example, the rock is locally stiffer even though it is softer than it was prior to the experiment. There is also consistent evidence of double peaks at trigger delay times of 14, 27, and 39 μs shown in Figure 4.

- Transitions between strain cycles suggest rock memory: We find three hysteresis loops for each input pump amplitude (strain level) shown in Figure 5 because we used propagating waves to load three cycles of strain into the rock, with each cycle having different amplitude and shape. We consider that each hysteresis loop begins and ends when the strain is zero. There are transitional regions between the hysteresis loops related to each pump input. For example, Figure 5d shows a transitional region most clearly between the first and the second hysteresis loops where the curve type changes from solid to dashed. When strain amplitude increases, the rock moves from one state, through a transitional region and into the next state in the subsequent cycle of strain. In the transitional region, the modulus change is affected by the previous strain state, suggesting that the rocks need a short time to “forget” the previous state, which indicates a memory effect. The memory effect is described by the parameter $\gamma$ that is introduced in the modeling section and is on the order of tenths of a microsecond for the Crab Orchard sample and frequencies used (i.e., a fraction of a cycle).

- Lucite has negligible nonlinearity: In both Figures 4 and 5 we compare the Crab Orchard data to measurements made on Lucite. Lucite has been used in many previous studies as a reference (linear) material, compared to the high nonlinearity of rocks (Guyer & Johnson, 1999; Winkler & McGowan, 2004). The nonlinearity of the Lucite is not apparent in our data and is assumed to be smaller than can be measured by our experimental procedure.

- A time-dependent $C$ term allows our model to fit complex observed data: Using the viscoelastic nonlinear model described in more detail in section 4, we compare our model with the time domain data in Figures 4 and 5. A key difference in our model from previous models is our inclusion of a time-dependent $C$ term, whereas in previous work (Riviere et al., 2013), $C$ is only a function of maximum strain. We show in Figure 4e that our model fits the complicated waveform data very well to first-order because we used the complete prior history of the waveform. In Figure 4f, we attempted to use equation (4) to fit the data while using a constant (with respect to time) $C$ term to fit each of the three cycles independently. The fits using such a value of $C$ are poor by comparison (Figure 4e). Similarly in Figure 5, we compare our model to data plotted as hysteresis loops for the three cycles for each maximum strain. The agreement is quite good for the first two cycles at each maximum strain level, with the second cycle having the best agreement. Although the third cycle fit is poor it should be noted that we only used the first two cycles of data in the inversion because the third cycle could be more contaminated by boundary reflections. Even so the model predicts the overall shape of the third cycle data. We compare fits obtained using our model with fits obtained using a constant (with respect to time) $C$ term model for all hysteresis loops in Figures 5e and 5f, respectively. As is evident from Figure 5f, such a $C$ term is unable to describe the observed effects that we capture with our model using a time-dependent $C$ term (Figure 5e).

“Slow dynamics” effects were described by Johnson and Sutin (2005) and TenCate (2011). While slow dynamics occurs for longer periods of time (seconds and longer), what we call short-period memory is an effect we attribute to combined viscoelastic and nonlinear effects occurring at small fractions of an
acoustic period (for our experiments on the submicrosecond scale). In a previous study utilizing a similar setup of propagating waves, we did not observe a slow dynamics effect (Gallot et al., 2015). This could be due to the fact that the pump excitation is of short duration (a few cycles) and relatively small amplitude ($<10^{-9}$) compared to other studies (Shokouhi et al., 2017).

4. Modeling Short-Period Nonlinear Viscoelastic Memory and Nonlinear Elasticity

In materials obeying a simple Hooke’s law, stress is directly proportional to strain, but as explained above, we need a better model to track the changes we observe. Instead of an instantaneous correspondence between stress (or elastic modulus) and changes in strain, we see both a nonlinear relationship and a memory effect.

Since instantaneous correspondence is inconsistent with our observations, we include a time-dependent parameter $C$ in equation (4). Our experimental results show that this parameter $C$ scales with pump input voltage (maximum pump strain). Because the strain is a function of the pump input, the absolute value of $C$ is some function of strain and its time of application. Therefore, we postulate that

$$C = C(\bar{t}, \varepsilon),$$

where $\bar{t}$ is the trigger delay time.

In order to describe the memory effect, $C$ is expressed as a convolution of a material impulse response function (explained in Appendix B in more detail), which describes the viscoelasticity of the medium, and the classical nonlinear response to the steady state nonlinear modulus variation (equation (3)) represented as $s(\varepsilon(t))$.

To include the effect of viscoelasticity, we approximate the material impulse response function as a simple decaying exponential function with decay constant $\gamma$ as outlined in Appendix B. Our phenomenological model to describe the nonlinear viscoelastic parameter, $C$, at trigger delay time, $\bar{t}$, becomes

$$C(\bar{t}, \varepsilon) = \int_0^{\bar{t}} e^{-\gamma(t-\bar{t})} s(\varepsilon(t)) dt$$

or

$$C(\bar{t}, \varepsilon) = \int_0^{\bar{t}} e^{-\gamma(t-\bar{t})} \left( \beta \varepsilon(t) + \delta \varepsilon(t)^2 \right) dt,$$

where $\gamma$ is a viscoelastic parameter. Therefore,

$$\Delta M(\bar{t}) / E = C(\bar{t} - \Delta t) + \beta \varepsilon(\bar{t} - \Delta t) + \delta \varepsilon(\bar{t} - \Delta t)^2$$

$$= \int_0^{\bar{t} - \Delta t} e^{\gamma(t-\bar{t})} \left( \beta \varepsilon(t) + \delta \varepsilon(t)^2 \right) dt + \beta \varepsilon(\bar{t} - \Delta t) + \delta \varepsilon(\bar{t} - \Delta t)^2,$$

where $\Delta t$ is the delay time between when the modulus variation occurs relative to the strain change, which is a phenomenon of viscoelasticity. There are several forms that equation (9) might take. The one we have chosen is straightforward to interpret (see Appendix B) and numerically implement (Appendix C). It fits our data quite well as we have shown. In the equation, $\beta$ and $\delta$ are the coefficients that describe the nonlinear elasticity, $\gamma$ is the coefficient that describes viscoelasticity, and $\Delta t$ is the time shift between the peak strain and the peak nonlinear modulus change. The value of $\gamma$ is obtained empirically (Appendix C) to give the best fit of our phenomenological model to the data and also corresponds well with independent measurements of sample absorption that are described in Appendix B.

5. Nonlinear Parameter Estimation

We use a least squares minimization to estimate the nonlinear coefficients $\beta, \delta,$ and $\gamma$ that provide the best fit of our model (equation (9)) to our measured data $\Delta M(\bar{t}) / E$.

Appendix C provides the details of the inversion process. The parameters $\beta$ and $\delta$ differ by 5 orders of magnitude, so one of the important considerations in the inversion is to include scaling constants to insure convergence to a stable solution.
Because the values of $\Delta M/E$ found during our experiments on Lucite are almost 2 orders of magnitude smaller than those for Crab Orchard Sandstone, the nonlinear coefficients, $\beta$ and $\delta$, for Lucite are too small to be accurately determined for our model described by equation (9). This result agrees with the observation that Lucite is much less nonlinear than the rock.

Using the $\Delta M/E$ and the strain data for the first two cycles of each experiment on the Crab Orchard Sandstone, we determined the four coefficients, $\gamma$, $\Delta t$, $\beta$, and $\delta$, which are listed in Table 1. Note that all the parameters are all relatively independent of pump input amplitude. Using the four coefficients found for each experiment and equation (9), we modeled the $\Delta M/E$ data using the strain measured on the Crab Orchard Sandstone. The modeling results fit the experimental data well for the second cycle (Figure 6). This agreement would not be possible with a non-time-varying $C$ model as previously discussed in section 3.2. Our model using these four parameters is able to capture the qualitative complexity of the data including the asymmetric strain loops in Figures 5 and 6 and the variability seen in the modulus changes versus delay time in Figure 4.

### 6. Discussion and Conclusions

The time domain method of using copropagating waves that we present provides a robust means of measuring the nonlinear parameters of rocks that is consistent with other measurement techniques (e.g., Renaud et al., 2012). Our values for $\beta$ and $\delta$ are also quite consistent for a large variation in maximum strain level (Table 1). In addition, this method yields values for a viscoelastic parameter, $\gamma$, and a time- and strain-dependent value of $C$. We compare the fit to our data using our time-dependent $C$ with a best fit using a constant value (with respect to time) for $C$ for the three cycles in our waveforms in graphs e and f of Figures 4 and 5. Comparing panels (e) and (f) in these two figures, we find that our model provides a better fit to the data.

In our model, the term $e^{-\gamma(\tau-t)}$ describes a viscoelastic effect acting on the signal waveforms through a convolution operation. This means that at each time $\tau$, the modulus is dependent on the shape and previous values of the strain waveform, indicating a short-term memory effect. In addition, $C$ is found to be dependent on the input level of strain, as well as being a function of $\beta$, $\delta$, and $\gamma$, which allows us to model more general

### Table 1

| Maximum strain ($\times 10^{-7}$) | $\gamma$ ($\mu s^{-1}$) | $\Delta t$ ($\mu s$) | $\beta$ ($\times 10^2$) | $\delta$ ($\times 10^9$) |
|-------------------------------|-------------------|-----------------|-----------------|-----------------|
| 4.7                           | 0.21              | 0.7             | -1.4            | -0.4            |
| 6.0                           | 0.24              | 0.8             | -1.6            | -0.5            |
| 7.7                           | 0.24              | 0.9             | -1.6            | -0.4            |
| 8.8                           | 0.22              | 0.8             | -1.7            | -0.4            |
| 9.7                           | 0.20              | 0.8             | -1.8            | -0.4            |

**Figure 6.** Second cycle of the hysteresis curves for different maximum pump strain amplitudes. The elastic modulus variation, $\Delta M/E$ as a function of the strain for Crab Orchard Sandstone samples in the second cycle of the strain. Different colors indicate different maximum strain. The solid and dash-dotted lines indicate the experimental and modeling data, respectively. The circle markers indicate the data points taken at 1 $\mu s$ intervals in delay times.
experimental geometries. The values of $\beta$, $\delta$, and $\gamma$ are relatively independent of the level of the pump strain amplitude (Table 1) and appear to be an intrinsic mechanical property of the rock.

Darling et al. (2004) showed that the individual minerals in a rock behave linearly, while the composite rock is nonlinear, indicating that nonlinear elasticity is controlled by grain contacts and the pore structure of the rock. In our results, the nonlinear behavior of the Crab Orchard material indicates considerable softening under applied tensile strain yet little recovery during compression. In Figures 4a–4e, the modeling clearly displays the continuous trend of the elastic modulus variation toward more negative values. The modeling results fit the observation that the elastic modulus variation is always negative whether the strain is compressive or dilatational when the pump input is large. That means that when pump input is large enough, the rock always softens. This could be indicative of a microstructure containing small closed cracks or, alternatively, cracks containing fluid in which the cracks open more readily than they close. Our results also show significantly greater asymmetry in the hysteresis loops (Figures 5 and 6) than previous studies using resonance methods (e.g., Renaud et al., 2012; Riviere et al., 2015). This asymmetry is a result of the time-dependent pump waveform changes and the strong rock softening effects. These results point to the possibility that the use of propagating waves to estimate nonlinear parameters may provide additional information about the crack and pore structure behavior differences in tension and compression.

Our phenomenological model shows a good match to the peaks and troughs of the modulus variation as a function of the measured strain. The nonlinear parameters were derived using the data from the first two cycles of the pump input waveform. We did not use the data from the third cycle in our inversion because of possible contamination from edge effects from our finite sample. Even so, the model prediction for the third cycle captures the major trends in the data. In addition to possible boundary effects, our phenomenological model uses only the $x$ component of the velocity measured by the laser vibrometer to approximate the strain. Even with these limitations, however, the model provides a good first-order approximation for most of the complicated waveform structures we observed.

In this paper we discuss only the application of our methods to the study of the nonlinearity of Crab Orchard Sandstone and how it can be parameterized using the three parameters $\gamma$, $\beta$, and $\delta$. It is important to ask how general a description this is for other rocks and what physical mechanisms these model parameters depend upon. Previous studies using other experimental methods (Johnson et al., 1996; Renaud et al., 2012; Riviere et al., 2015) have shown that the nonlinear parameters vary with rock type. We have made initial measurements on several other sedimentary rocks under dry conditions that support this observation. These results lead us to speculate that rocks can be classified using measurements such as those we have made and that nonlinear parameters are sensitive to differences in the pore space microstructure in rocks such as populations of microcracks (Renaud et al., 2012) that have been long known to impart anelasticity. Our present hypothesis, which needs to be investigated by more detailed experiments, is that the asymmetric nonlinear response to loading conditions leads to different amplitudes during the compressional and tension half cycles in the hysteresis curves (see Figure 6).

In conclusion, we developed a sensitive method that uses propagating waves to quantitatively measure the nonlinearity of samples. We propose a phenomenological model that includes the classical nonlinear parameters in addition to viscoelasticity to explain our measurements on a sample of Crab Orchard Sandstone. We have shown that these factors as well as waveform history produce a measurable short-term memory effect, shorter in length than a probe waveform cycle, which is predicted by our model. Our new observations and methods have significance for quantifying changes in microstructure of rocks. These developments may allow earth scientists to take advantage of the advances in other fields, such as medical imaging, material science, and nondestructive testing, to characterize the Earth with applications to earthquake physics, energy, and environmental problems including the production or injection of fluids in near surface reservoirs.

**Appendix A: Estimation of Pump-Induced Strain**

Figure A1 shows the $x$ axis component of the pump particle vibration velocity as a function of position and time measured on the surface of the sample along a line connecting the probe source position to the probe receiver position. We take this as the approximate strain in the sample. In the figure, the $x$ axis shows the offset of the measurement point from the transducer T1. The cyan dashed lines indicate the position of the
probe signals as a function of time when the probe transducer (T1) trigger is delayed 0, 10, 20, 30, and 40 μs, relative to the pump transducer (T2) trigger, respectively. We averaged the pump signals along the survey line for each probe delay and calibrated the elapsed time between the pump signals at the surface and the pump signals in the interaction region. Accordingly, we obtain the average $x$ axis particle vibration velocity as a function of trigger delay time. Based on the average $x$ axis particle vibration velocity $v(t)$, the average $x$ axis strain induced by the pump signals can be calculated:

$$\varepsilon(t) = \frac{-v(t)\,v_p}{C_0}.$$  

(A1)

where $v_p$ is the longitudinal wave velocity (about 2,870 m/s).

Boundary reflections from the edges of our physical model (Figure 1) will begin to arrive at a delay of about 40 μs compared to the direct pump arrival shown in Figure A1. Such reflections are assumed to have small impact on the pump strain values due to the transducer radiation pattern, which excites little energy at high angles from the transducer center line as shown in Figure 1 (Tang et al., 1994; Szabo, 2014, Chapter 6). In addition, amplitudes are further reduced by geometrical spreading and the greater absorption on the longer reflected paths. The relative simplicity of the waveforms in Figure A1 for traveltimes of 0–40 μs supports this assumption. These reflections will, however, have the most impact at late times and larger measurement distances. For this reason we use only the first two cycles of strain data in our inversion for nonlinear parameters.

**Appendix B: Relation Between Memory Effect and Attenuation**

We can interpret $\gamma$ in terms of well-known quantities used to describe seismic wave attenuation. For frequency-dependent quality factor $Q$ (Sato et al., 2012), we can define the absorption by a frequency power law (Szabo, 1995, 2014 Chapter 4),

$$\alpha(f) = \alpha_0|f|^y,$$  

(B1)

where $\alpha_0$ is the absorption coefficient and $y$ represents the frequency dependence of absorption. Using the spectral ratio method to estimate $\alpha_0$ and $y$ from pulse transmission data collected on samples of Crab Orchard Sandstone and Lucite (He, 1999), we find that $\alpha_0 = 0.371$ neper/(cm-MHz$^y$) or 3.222 dB/(cm-MHz$^y$) and $y = 1.17$. This corresponds to a $Q$ of 29.5 at 1 MHz, which is similar to the value found in the laboratory for dry Berea Sandstone by Toksöz et al. (1979). The $P$ wave velocity ($V$) is found to be 2,870 m/s. We used Lucite as a reference for the spectral method and used the substitution method (He, 1999) to correct spectral ratios for the attenuation in the Lucite. Absorption is accompanied by velocity dispersion as required by causality and expressed by a phase term $\varphi(f)$ (Aki & Richards, 2002; Szabo, 1995). We thus describe the wave propagation through a viscoelastic medium by the material transfer function.

$$R(z, f) = \exp(-\alpha_0|f|^y z - i(2\pi f z / c + \varphi(f) z)).$$  

(B2)
The real and imaginary components of $R$ at $z = 150$ mm based on power law fits to absorption of our Crab Orchard sample and the causal model are shown in Figure B1.

The time domain transfer function, $r(t-z/V)$, for the sample is the inverse Fourier transform of (B2) and is plotted in Figure B1 for a propagation distance of 150 mm. For a signal $s(t)$ with a spectrum, $S(f)$, injected into a material the output spectrum at a distance $z$ is

$$V(z, f) = R(z, f)S(f)$$  \hspace{1cm} (B3)

and the waveform at distance $z$ is

$$v(z, t) = r(z, t)s(t),$$  \hspace{1cm} (B4)

where $r$ is the inverse Fourier transform of $R$. Note that in Figure B2, this $r$ is an asymmetric real function of time and the tail can be viewed as a type of memory effect such that the output signal $v(z, t)$ is influenced by past values of the input $s(t)$. For the purely elastic case, this function would revert to an impulse function centered on the propagation delay time. If a simple linear symmetric approximation of the form $\exp(-\gamma|t|)$ is made to match the points on $r(t, z = 150$ mm) that have amplitude of $e^{-1}$ of the maximum value, we find that $\gamma$ is 0.13 $\mu$s$^{-1}$. This is close to the value obtained for our sample of Crab Orchard Sandstone by fitting $\Delta M/E$ in section 5. This exponential approximation of $v(z, t)$ is

$$v(z, t) \approx e^{-\gamma t} s(t),$$  \hspace{1cm} (B5)

and can be recognized as the same convolutional form in equation (7).

In this approximate model we capture the dominant effect of material absorption. In their comprehensive study of the nonlinear properties of rocks, Renaud et al. (2012) measured a variation of absorption with strain, $\alpha_0 - \alpha_0$, from the nonstrained sample value, $\alpha_0$. We have not included this small variation in absorption, calculated by Renaud et al. (2010) as 1.2% of the total measured absorption in our approximation, which captures the dominant broadband absorption effect to first order.

### Appendix C: Inversion for Model Coefficients

We use a least squares minimization procedure to find the model coefficients. To estimate the nonlinear coefficients $\beta$, $\delta$, and $\gamma$, we start from equation (9), to obtain

$$\Delta M(\bar{t})/E = \left(\int_0^{\bar{t} - \Delta t} e^{\bar{t} - t} \varepsilon(t) dt + \varepsilon(\bar{t} - \Delta t)\right) \beta + \left(\int_0^{\bar{t} - \Delta t} e^{\bar{t} - t} \varepsilon(t)^2 dt + \varepsilon(\bar{t} - \Delta t)^2\right) \delta,$$  \hspace{1cm} (C-1)

We define

$$B = \Delta M(\bar{t})/E A_1 = 10^2 \times \left(\int_0^{\bar{t} - \Delta t} e^{\bar{t} - t} \varepsilon(t) dt + \varepsilon(\bar{t} - \Delta t)\right) A_2$$

$$= 10^8 \times \left(\int_0^{\bar{t} - \Delta t} e^{\bar{t} - t} \varepsilon(t)^2 dt + \varepsilon(\bar{t} - \Delta t)^2\right) x_1 = 10^{-2} \times \beta x_2$$  \hspace{1cm} (C-2)

The conversion to $x_1$ and $x_2$ is done to scale the inversion for convergence. Then, these can be expressed as

$$x_1 = \frac{10^8 \times \beta}{10^{-2} \times \beta}$$

$$x_2 = \frac{10^8}{10^{-2}}$$
\[ A \times X = B [A_1, A_2] \times X = B \begin{bmatrix} a_{11} & a_{21} \\ \vdots & \vdots \\ a_{1N} & a_{2N} \end{bmatrix} \begin{bmatrix} x_1 \\ x_2 \end{bmatrix} = B \begin{bmatrix} b_1 \\ \vdots \\ b_N \end{bmatrix}, \tag{C-3} \]

where \( N \) is the number of trigger delay time times. We solve the equation for trial values of \( \gamma \) and \( \Delta t \), and get

\[ X = B / A. \tag{C-4} \]

To find \( x_1 \) and \( x_2 \) for a range of values of \( \gamma \) and \( \Delta t \). Finally, we obtain

\[ \tilde{\beta} = 10^3 \times x_1 \quad \delta = 10^6 \times x_2. \tag{C-5} \]

To complete the solution, we find

\[ O(\gamma, \Delta t) = \int_0^{\Delta t} e^{(\gamma - \Delta t)^2} \left[ \beta \delta (t) + \tilde{\beta} \delta (t - \Delta t) + \tilde{\beta} \delta (t - \Delta t)^2 \right] dt. \tag{C-6} \]

We can define an error function,

\[ e(\gamma, \Delta t) = \left\| O(\gamma, \Delta t) - \Delta M(\Delta t) / E \right\|^2 \tag{C-7} \]

which we minimize using a grid search to find the optimal \( \gamma \) and \( \Delta t \). The grid search procedure is to choose \( \gamma \) and \( \Delta t \), solve equations (C-4) and (C-5) to find \( \beta \) and \( \delta \), and then calculate the error using equation (9). We evaluate a range of values for \( \gamma \) and \( \Delta t \) to find the best fit of the model to the experimental data.

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