

Determination of acoustic nonlinearity parameters using thermal modulation of ultrasonic waves

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ABSTRACT

This study presents a test method and its theoretical framework to determine the acoustic nonlinearity parameters (α , β , δ) of material using thermal modulation of ultrasonic waves. Temperature change-induced thermal strain excites the nonlinear response of the material and modulates the ultrasonic wave propagating in it. Experimental results showed a strong correlation between the relative wave velocity change and the temperature change. With a quadratic polynomial model, the acoustic nonlinearity parameters were obtained from the polynomial coefficients by curve fitting the experimental data. Their effects on thermal-induced velocity change were discussed. The parameters α , β , and δ govern the hysteretic gap, average slope, and curvature of the correlation curve, respectively. The proposed theory was validated on aluminum, steel, intact and damaged concrete samples. The obtained nonlinear parameters show reasonable agreement with values reported in the literature. Compared to other nonlinear acoustic methods using vibration or acoustic excitation, the thermal modulation method generates more uniform, slow changing, and larger strain field in the test sample. Employing the thermal effect as the driving force for nonlinearity instead of an undesired influencing factor, this method can measure the absolute values of α , β , and δ with good accuracy using a simple ultrasonic test setup.

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Nonlinear acoustic/ultrasonic techniques show high sensitivity to microcracking damage, especially in complex materials. Following the phenomenological description of stress-strain hysteresis of rock by McCall and Guyer,^{1,2} the elastic modulus of a complex material can be modeled as a strain and strain rate-dependent variable and described with three nonlinear parameters β , δ , and α . The first two parameters represent the classical nonlinear perturbation parameters, and α is the nonclassical nonlinear parameter, which is introduced as a measure of material hysteresis. Meurer *et al.*³ proposed a general form of a nonlinear constitutive model, and they showed that the McCall and Guyer model is a special case of this general model when only the leading order effect of α is considered.

Acoustic nonlinearity of material may manifest as softening of the modulus with increasing strain or higher harmonic generation. Modulus softening can be measured as the change in resonance frequency or ultrasonic wave velocity at different strain levels. The nonlinear resonance acoustic spectroscopy (NRAS) method^{4,5} measures the resonance frequency shift Δf when the resonance mode in a sample is excited at different strain levels. The linear slope between Δf and the strain change ϵ is defined as the relative nonlinear parameter

α . The dynamic acousto-elastic testing (DAET) method is a recently developed nonlinear ultrasonic method that measures the relative ultrasonic wave velocity change due to a low-frequency strain modulation.^{6,7} Nonlinear parameters β , δ , and α_{DAET} were derived from the correlation curves between the relative velocity change and strain. Higher harmonic generation is a phenomenon wherein the ultrasonic waveform is distorted by the nonlinear response of the material, and then higher harmonic waves are generated. Second harmonic generation (SHG)^{8,9} measures the amplitudes of the fundamental and the second harmonics to calculate the nonlinear parameter β . Because the absolute displacement amplitudes are challenging to measure, a relative value of β is typically used for characterizing fatigue of metal materials¹⁰ and concrete damage.¹¹

Although these nonlinear ultrasonic testing methods show different degrees of success in extracting nonlinear parameters and characterizing material damage, some limitations hinder the application of these methods in practice. First, the absolute nonlinear parameters are not easy to measure. The NRAS method uses the acceleration instead of the strain to calculate an alternative nonlinear parameter α_f . In addition, NRAS is only applicable to small samples since it

needs to excite the global vibration of the test sample. Second, many nonlinear ultrasonic methods use an impact or an actuator to excite nonlinear responses of the test sample. The excitation applied at a single point or a small area creates a nonuniform strain field in a local area of the sample. The measurement results are affected by the strain distribution in the sample.

In a recent work by the authors,¹² we investigated ambient temperature-induced ultrasonic wave velocity changes in concrete samples with microcracking damage and defined a thermal modulation coefficient to represent the sensitivity of wave velocity to the temperature effect. It is found that the thermal modulation coefficient increases with the damage level in concrete samples, which shows its potential for nondestructive evaluation of material. In this Letter, we propose using the thermal modulation of nonlinear ultrasonic waves to determine the nonlinear parameters α , β , and δ . The slow temperature change creates a relative uniform thermal strain field in the test sample. The thermal strain can be used as the driving force to excite nonlinear behaviors of a material and modulate ultrasonic waves propagating in the material. Based on the experimental results from the thermal modulation test, we built a theoretical model to determine the absolute values of nonlinear parameters. The proposed theory was validated on classical nonlinear materials (metals) and nonclassical nonlinear materials with hysteresis (concrete).

Temperature effects are often regarded as undesired noise in most experimental studies. The dependence of elastic wave velocity on temperature has been studied by many researchers.^{12–15} Previous researchers^{13,14} have noticed a linear relationship between the relative velocity change dv/v and the temperature change ΔT on metals. For materials with hysteresis, Fig. 1 shows a diagram of $dv/v \sim T$ correlation, where a sample experienced a heating/cooling thermal cycle ($T_0 \rightarrow T_1 \rightarrow T_0$). The relative velocity change dv/v decreases from points A to B during the heating and increases from B to C while cooling. In this Letter, through analytical derivations and experimental data analyses, we prove that the nonlinear acoustic parameters can be derived from the $dv/v \sim T$ correlations on metal materials and concrete.

A quadratic polynomial is used to model the dv/v vs temperature relationship as

$$dv/v = k_0 + k_1 \Delta T + k_2 \Delta T^2, \quad (1)$$

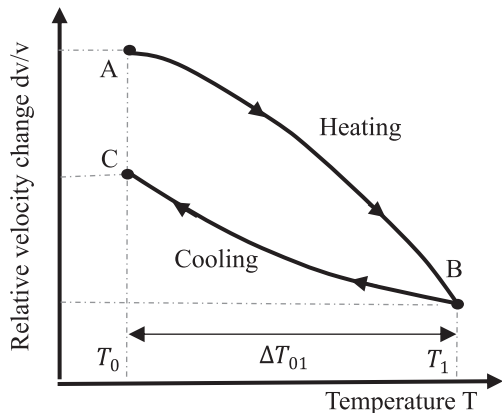


FIG. 1. Diagram for correlation between the relative velocity change and the temperature change.

where k_0 , k_1 , and k_2 are the coefficients of the polynomial and ΔT is the temperature change relative to a reference temperature. These coefficients are obtained from curve fitting of the correlation curves in Fig. 1 and are denoted as the thermal modulation coefficients.

For nonlinear materials with hysteresis, the modulus can be described with a strain and strain rate-related model as⁴

$$E(\varepsilon, \dot{\varepsilon}) = E_0 \{1 - \beta \varepsilon - \delta \varepsilon^2 - \alpha [\Delta \varepsilon + \varepsilon \text{sign}(\dot{\varepsilon})]\}, \quad (2)$$

where E_0 is the linear modulus and β and δ are the quadratic and cubic nonlinear parameters (classical nonlinear parameters). α describes the hysteretic behavior of nonclassical nonlinear materials. ε and $\dot{\varepsilon}$ are the strain and strain rate; $\text{sign}(\dot{\varepsilon}) = 1$ if $\dot{\varepsilon} > 0$, and $\text{sign}(\dot{\varepsilon}) = -1$ if $\dot{\varepsilon} < 0$. $\Delta \varepsilon$ is the maximum strain experienced in the previous loading cycles.

Since the velocity is related to \sqrt{E} , the relative velocity change is expressed as

$$dv/v = \frac{1}{2} \frac{\Delta E}{E} = -\frac{1}{2} \{\beta \varepsilon + \delta \varepsilon^2 + \alpha [\Delta \varepsilon + \varepsilon \text{sign}(\dot{\varepsilon})]\}. \quad (3)$$

Temperature changes will generate thermal strain ε in a material, which can be expressed as $\varepsilon = \alpha_T \Delta T$, where α_T is the thermal expansion coefficient. If the sample experiences the temperature cycle as shown in Fig. 1, then $\Delta \varepsilon = \alpha_T \Delta T_{01}$ represents the maximum strain that the sample experienced for both the heating and cooling processes.

For the heating process ($A \rightarrow B$), $\text{sign}(\dot{\varepsilon}) = 1$ and T_0 is used as the reference temperature. For the cooling process ($B \rightarrow C$), $\text{sign}(\dot{\varepsilon}) = -1$ and T_1 is the reference temperature. Then, Eq. (3) can be written as below for the heating (+) and the cooling (−) processes:

$$dv/v^{(\pm)} = -\frac{1}{2} \{\beta(\alpha_T \Delta T) + \delta(\alpha_T \Delta T)^2 + \alpha(\alpha_T \Delta T_{01} \pm \alpha_T \Delta T)\}. \quad (4)$$

Equation (1) for heating and cooling cycles can be rewritten as

$$dv/v^{(\pm)} = k_0^{\pm} + k_1^{\pm} \Delta T + k_2^{\pm} \Delta T^2, \quad (5)$$

where k^{\pm} represents the coefficients for heating and cooling processes.

By comparing the first degree term ΔT in Eqs. (4) and (5), we can get the following solution for k_1^{\pm} :

$$\begin{cases} k_1^+ = -\alpha_T(\beta + \alpha)/2, \\ k_1^- = -\alpha_T(\beta - \alpha)/2. \end{cases} \quad (6)$$

The nonlinear parameters β and α are solved as

$$\begin{cases} \beta = -(k_1^- + k_1^+)/\alpha_T, \\ \alpha = -(k_1^+ - k_1^-)/\alpha_T. \end{cases} \quad (7)$$

Equation (7) shows that for classical nonlinear materials without hysteresis ($\alpha = 0$), the coefficients k_1^+ and k_1^- should be equal. This statement is validated in the experiments presented in this Letter.

By comparing the second degree term ΔT^2 in Eqs. (4) and (5), we obtain two solutions to the cubic nonlinear parameter δ from the heating and cooling processes, which is related to the curvatures of the correlation curves,

$$\delta^{\pm} = -2k_2^{\pm}/\alpha_T^2. \quad (8)$$

To perform the thermal modulation test, the samples were placed in an environmental chamber with a controllable temperature changing rate. The sample experienced a slow temperature changing rate (1°C/h) to minimize the temperature gradient. Two 2.25 MHz ultrasonic transducers (Olympus A106S) were installed on the two opposite surfaces of the metal samples as the transmitter and receiver. For concrete samples, two 15 mm diameter piezoelectric disks (STEMINC SMD15T21R111WL) were used to excite and receive ultrasonic waves around the frequency of 150 kHz. Ultrasonic signals were acquired using a digital oscilloscope (PICO 4224), and the temperature was monitored using a type T thermocouple and a data logger.

For common structural materials (aluminum, steel, and concrete), the relative velocity change with temperature is on the order of $10^{-4}/^\circ\text{C}$. In order to accurately measure such small velocity changes, we used the coda wave interferometry (CWI) method.¹⁶ If two signals differ only by a dilation, which is typically true for temperature-induced signal distortion, we can determine the dilation by stretching the disturbed signal and obtaining the stretching factor when the cross correlation between the signals reaches maximal. In the experiments, we were able to measure the relative velocity change with a precision of 10^{-6} .

The dilation $\delta_t = dt/t$ represents the relative time delay dt at the time window t of a signal. In the thermal modulation test, two effects contribute to the relative time delay δ_t : thermal expansion of the sample and temperature dependence of the wave velocity.^{14,16} The relative velocity change will be calculated as $dv/v = -(\delta_t - \alpha_T \Delta T)$. If the thermal strain is negligible ($\varepsilon = \alpha_T \Delta T \ll dv/v$), the relative velocity change can be calculated as $dv/v = -dt/t = \delta_t$.

We first tested metal materials (aluminum and steel) using the thermal modulation method. Figure 2 shows the thermal modulation results of an aluminum 6061 block ($5\text{ cm} \times 5\text{ cm} \times 18\text{ cm}$) and a stainless steel 304 block ($3\text{ cm} \times 6\text{ cm} \times 30\text{ cm}$) with the temperature histories and relative velocity change histories. The two samples were first heated to a specific temperature and then cooled down to room temperature. The temperature changing rate was about 0.96°C/h , which was close to the designed temperature rate. The relative velocity change in Fig. 2(b) shows an opposite trend to the temperature change history.

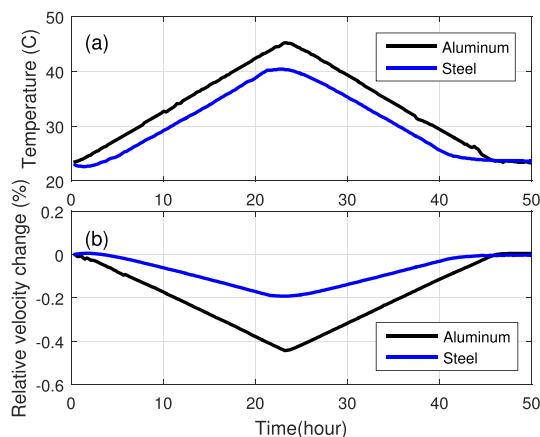


FIG. 2. Thermal modulation test results of aluminum and steel: (a) temperature history and (b) relative velocity change history.

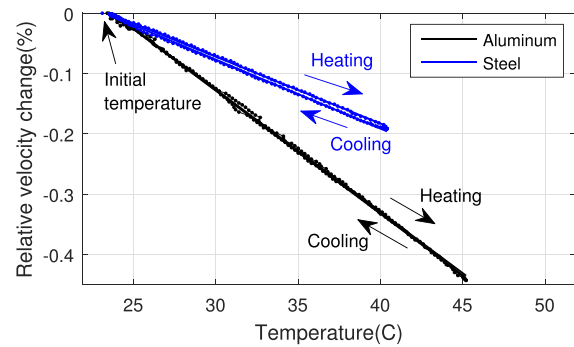


FIG. 3. Thermal modulation test results on aluminum 6061 and stainless steel samples.

Figure 3 presents the $dv/v \sim T$ curves for the aluminum and steel samples. The extracted thermal modulation coefficients are summarized in Table I. The steel sample has a smaller absolute value of k_1^\pm (slope) than the aluminum sample. Both correlation curves show very good linearity, which indicates small values of $|k_2^\pm|$ for aluminum and steel.

The nonlinear parameters α , β , and δ can be calculated from Eqs. (7) and (8) by using the thermal expansion coefficients $\alpha_T = 23 \mu\text{e}/^\circ\text{C}$ for aluminum and $\alpha_T = 17.3 \mu\text{e}/^\circ\text{C}$ for steel. The results are also given in Table I. From the thermal modulation tests, we obtained $\beta = 15.4$ for aluminum and $\beta = 10.7$ for steel. The absolute value of β reported in the literature varies from 4 to 12 for aluminum alloys and 2–4.5 for stainless steel.^{17–20} The β values obtained from the thermal modulation test are slightly larger than the reported values in the literature for both aluminum and steel. A possible explanation is the differences in the strain measurement and strain range. In the thermal modulation test, the samples had a very slow strain changing rate, relatively large strain level, and uniform strain distribution, while most other nonlinear acoustic methods (NRAS, DAET, and SHG) generate dynamic strain at very low strain levels.

For both the aluminum and steel curves, the heating and cooling slopes are equal $k_1^+ = k_1^-$, which gives the nonclassical parameters $\alpha = 0$. Although both curves show good linearity, the δ^\pm values for the steel are almost 10 times higher than those for the aluminum sample. The steel sample also shows a narrow hysteresis area. Just like the difference between k_1^+ and k_1^- gives the hysteresis parameter α , the authors believe that the difference between δ^+ and δ^- may represent a higher order hysteresis response ($\sim \varepsilon^2$). This higher order effect of hysteresis has been predicted by Meurer *et al.*³ in their general nonlinear

TABLE I. Nonlinear parameters for aluminum and steel.

Sample	Aluminum 6061	Stainless steel 304
α_T	23.0×10^{-6}	17.3×10^{-6}
k_1^+, k_2^+	$-1.77 \times 10^{-4}, -9.3 \times 10^{-8}$	$-0.93 \times 10^{-4}, -5.8 \times 10^{-7}$
k_1^-, k_2^-	$-1.77 \times 10^{-4}, -1.1 \times 10^{-7}$	$-0.93 \times 10^{-4}, +6.0 \times 10^{-7}$
β	15.4	10.7
δ^+, δ^-	351, 415	3875, -4009
α	0	0

model, while Eq. (3) only includes the lower order term ($\sim \varepsilon$). Since samples will experience large thermal strains ($10^{-4} \sim 10^{-3}$) in the thermal modulation test, the higher order effects of α may need to be considered in future studies.

Concrete is a typical nonclassical nonlinear material. We tested two concrete cylinders (10 cm diameter \times 20 cm height), with one control sample cast with normal concrete mix and the other sample with microcracking damage induced by alkali-silica reaction (ASR). ASR is a chemical reaction in concrete, which occurs between alkali hydroxides in hydrated cement and reactive silica in certain types of aggregates. ASR produces an expansive gel, which will eventually cause microcracks and degradation of concrete. The mix design and curing condition of the ASR sample were described in a previous work by the authors.²¹ At the test time, the ASR sample had moderate damage (0.1% expansion) with minor surface cracks, while the control sample was in an intact condition. For the thermal modulation test, the two concrete samples were first heated from 23.2 °C to 44.1 °C and then cooled down to 23.2 °C. The correlation curves between the relative velocity change and temperature are shown in Fig. 4 for the two concrete samples.

The thermal modulation coefficients k_1^+ , k_2^+ and k_1^- , k_2^- were obtained from curve fitting and are summarized in Table II. All fittings have the goodness-of-fit R^2 larger than 0.99. For both the control and ASR samples, there are $|k_1^+| > |k_1^-|$, which means that the relative velocity change is more sensitive to the temperature change in the heating process than in the cooling process. The ASR sample has larger coefficients $|k_1^+|$ and $|k_2^+|$ than the control sample, which indicates that microcracking damage would increase the sensitivity of velocity changes to temperature. In the previous work by the authors,¹² we also found that the $|k_1^-|$ value increased with the damage level in concrete samples.

The thermal expansion coefficients were measured for both the control and ASR samples. The results were very close, so that we used $\alpha_T = 10 \mu\epsilon/^\circ\text{C}$ for both samples. The nonlinear parameters α , β , and δ were calculated using Eqs. (7) and (8). The absolute values of β are 100 for the control sample and 221 for the ASR sample. For the control sample, the result agrees with the literature reported values in the range of 40–157 for concrete.^{15,22} The ASR sample shows a higher β value than the control, which indicates that microcracking damage increases the quadratic nonlinearity.

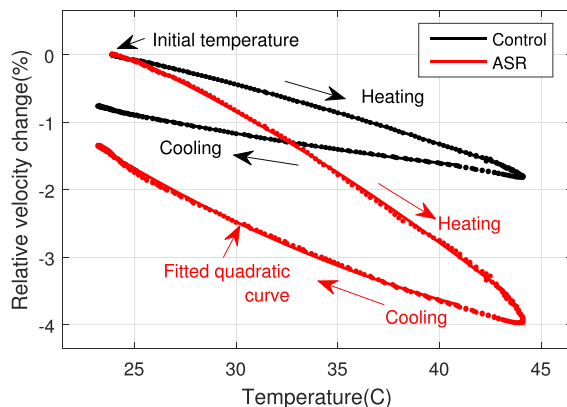


FIG. 4. Thermal modulation test results of concrete.

TABLE II. Nonlinear parameters for concrete samples.

Sample	Control	ASR sample
α_T ($^\circ\text{C}$)	10.0×10^{-6}	10.0×10^{-6}
k_1^+, k_2^+	$-6.8 \times 10^{-4}, -9.5 \times 10^{-6}$	$-13.9 \times 10^{-4}, -2.8 \times 10^{-5}$
k_1^-, k_2^-	$-3.2 \times 10^{-4}, +8.9 \times 10^{-6}$	$-8.2 \times 10^{-4}, +2.9 \times 10^{-5}$
β	100	221
δ^+, δ^-	$1.9 \times 10^5, -1.8 \times 10^5$	$5.6 \times 10^5, -5.8 \times 10^5$
α	36	57

For both samples, $|\delta^+|$ and $|\delta^-|$ in the heating and cooling processes are very close, and they are much larger than $|\delta^\pm|$ of the aluminum and the steel samples. The ASR sample has larger $|\delta^\pm|$ than the control sample, but the values are still on the same order of magnitude 10^5 and are close to the reported value of 10^6 for rock.²³

The ASR sample also shows a larger value of $\alpha = 57$ than the control sample $\alpha = 36$, which suggests that the damage in concrete increases the hysteresis. In Eqs. (4) and (5), α can also be solved from the constant term $|k_0^\pm|$. In a closed thermal cycle with the same starting and ending temperatures, α is related to the gap in the dv/v curves at the starting/ending points, i.e., $\alpha = \Delta(dv/v)/(\alpha_T \Delta T_{01})$. The α values calculated from the gap are 40 for the control sample and 73 for the ASR sample.

The effects of nonlinear parameters on the $dv/v \sim T$ curve are summarized. In Eq. (7), β and α were derived from the first degree coefficients k_1^+ and k_1^- , where β is related to the average slope and α is associated with the difference between the heating/cooling slopes. Therefore, β gives the sensitivity of the relative velocity change to the temperature change, and α describes the material hysteresis. In addition, α contributes to the gap in dv/v between points A and C in Fig. 1. For materials with very small hysteresis, the velocity will return to the original value after a closed thermal cycle. For mesoscopic materials (concrete, rock), the velocity may not go back to the original value, and this phenomenon has been reported by several researchers.^{24,25} The parameter δ is linked to the curvature of the correlation curves. The aluminum and steel samples have very small δ values, and their $dv/v \sim T$ curves show high linearity. For concrete samples, the correlation curves have similar curvatures during the heating and cooling processes, but with opposite signs. The ASR sample has a larger curvature than the control sample, which indicates a larger δ value. The difference between δ^+ and δ^- may be related to a higher order hysteresis behavior predicted by Meurer *et al.*³ In summary, for $dv/v \sim T$ curves from a thermal modulation test, hysteresis (α) determines the gap, the difference between the heating/cooling slopes and curvatures; β affects the average slope; δ controls the curvature of curves. Similar analyses were also given by Shokouhi *et al.*⁷ using the DAET method. Comparing the three parameters for the control and ASR concrete samples, we also conclude that microcracks and cracks increase both classical nonlinearity (β and δ) and nonclassical nonlinearity hysteresis (α).

In this Letter, we present a nonlinear ultrasonic test method based on thermal modulation to determine the absolute values of acoustic nonlinearity parameters of materials. Using a quadratic model, we derived the nonlinear parameters α , β , and δ from the $dv/v \sim T$ curve and determined their values by curve fitting of experimental data.

Compared to other nonlinear acoustic methods based on vibration or acoustic excitation, the thermal modulation method generates nonlinear behaviors of materials with a larger, relatively uniform, and slowly changing strain field. Therefore, the relative velocity change and the thermal strain can be readily measured with reasonable accuracy without using complicated test systems and calibration procedures. The experimental results showed reasonable agreement with literature-reported values, which further validates that the nonlinear response is induced by material strain, either mechanical strain or thermal strain. Ongoing studies are focused on the quantitative correlation between the nonlinear parameters and ASR-induced microcracking damage in concrete, which could provide a new nondestructive approach to evaluate material damage in the laboratory and *in situ* tests.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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