Intrinsic and coupling-induced elastic nonlinearity of lanthanum-doped lead magnesium niobate–lead titanate electrostrictive ceramic

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The elastic nonlinearity of lanthanum-doped lead magnesium niobate–lead titanate electrostrictive ceramic has been investigated by using ultrasonic second-harmonic generation technique and field-induced strain measurement. A large ultrasonic nonlinearity parameter, $\beta=30$, was observed at room temperature, which is more than twice of the values for lead zirconate–lead titanate piezoelectric ceramics. The third-order elastic constant c_{111} has been calculated from the β value. By introducing an effective nonlinearity parameter β_e that includes both the intrinsic and the electromechanical coupling generated elastic nonlinearities, the nonlinearity parameter can be accurately characterized. The strong field dependence of the nonlinearity parameter shows excellent potential for designing field tunable nonlinear devices using this material. © *2000 American Institute of Physics.* [S0003-6951(00)04335-7]

The solid solution $Pb_{0.985}La_{0.01}(Mg_{0.3}Nb_{0.6}Ti_{0.1})O_3$ [La-doped PMN-PT] is a relaxor material, which has a giant dielectric constant at room temperature and gives rise to large field-induced strain through the electrostriction effect. It was reported that a strain of 10^{-4} could be obtained under an electric field of less than 1 kV/cm.[1] The strong electrostrictive properties of this material make it a good material for transducers, sensors, and actuators.[2] This intrigued interest in both the materials and physical communities to study its elastic, dielectric, electrostrictive properties[3,4] and electromechanical coupling coefficients.[5] In those studies, however, linear elasticity of the material was assumed, although very strong nonlinear behavior was observed under electric field.[4] The assumption of linear elasticity is only conditionally valid since solids are inherently nonlinear and the PMN-PT system is inherently a strong nonlinear material.[6] The knowledge of elastic nonlinearity is important in fundamental studies as well as device designs. In the present letter we report the measurement results on the elastic nonlinearity of PMN-PT and their field dependence.

The lanthanum-doped PMN-PT ceramic was manufactured by TRS Ceramics (State College, PA 16801) and the measured sample is a cylinder with a diameter of 2.54 cm and 2.16 cm long. The ultrasonic second-harmonic generation setup used in the experiment is shown in Fig. 1. A 5 MHz toneburst signal from a function generator is amplified by a power amplifier, then, applied to the transmitting transducer, which sends a finite amplitude acoustic wave into the sample. When the ultrasonic wave arrives at the other end of the sample, its wave form is distorted due to the material nonlinearity and the second-harmonic wave is generated. The nonlinear ultrasonic phenomenon is originated from several sources: nonlinearity of the interatomic forces, geometrical nonlinearity, and nonlinear electromechanical coupling effects. For a general dielectric, the dynamical process is described by the nonlinear electroacoustic coupling equations in material reference frame.

$$\rho_0 \ddot{u}_i = T_{ij,j}, \tag{1a}$$

$$D_{k,k}=0, \quad (i,j,k=1,2,3), \tag{1b}$$

and the nonlinear constitutive relations are given by

$$\begin{aligned} T_{ij} = {} & c^E_{ijkl}u_{k,l} - e_{kij}E_k + \tfrac{1}{2}(c^E_{ijklmn} + \delta_{ik}c^E_{ljmn} + \delta_{im}c^E_{njkl} \\ & + \delta_{km}c^E_{ijln})u_{k,l}u_{m,n} - (e_{kijmn} + \delta_{im}e_{knj})u_{m,n}E_k \\ & - \tfrac{1}{2}m_{klij}E_kE_l, \end{aligned} \tag{2a}$$

$$\begin{aligned} D_k = {} & \varepsilon_{kl}E_l + e_{klm}u_{l,m} + \tfrac{1}{2}(e_{klmij} + \delta_{li}e_{kmj})u_{l,m}u_{i,j} \\ & + m_{klmn}E_l u_{m,n} + \varepsilon_{klm}E_lE_m, \end{aligned} \tag{2b}$$

where ρ_0 is mass density in unstrained state, **u** is particle displacement, **D** is material electric displacement, **T** is the stress tensor, c_{ijkl}, e_{ijk}, and ε_{ij} are the second-order elastic constants, piezoelectric coefficients, and dielectric constants, respectively. The coefficients c_{ijklmn}, e_{ijklm}, and ε_{ijk} are tensor components of the third-order elastic constant, nonlinear piezoelectric coefficient, and nonlinear dielectric constant, respectively, and m_{ijkl} are the electrostriction coefficients. The second harmonic generation is a special case of

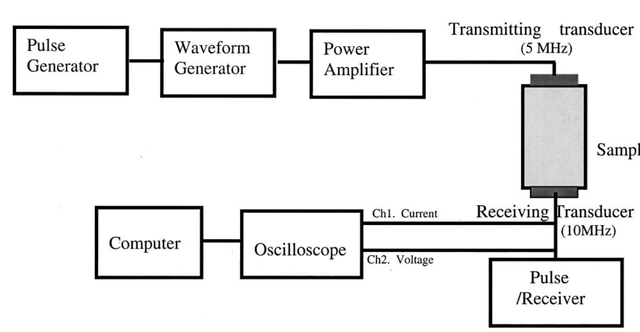

FIG. 1. Experimental setup for measure the absolute amplitudes of the first and second harmonic ultrasonic waves.

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TABLE I. The ultrasonic nonlinearity parameter β and the third-order elastic constant c_{111} of $Pb_{0.985}La_{0.01}(Mg_{0.3}Nb_{0.6}Ti_{0.1})O_3$ ceramic.

β	$c_{11}(10^{10}\ N/m^2)$	$c_{111}(10^{10}\ N/m^2)$
30	14.1	−465.3

three–phonon interactions in solids, i.e., the self-interaction of one fundamental phonon that generates another phonon with twice of the frequency.[7] Because the amplitude of the second harmonic is much smaller compared to the fundamental wave, only quadratic nonlinearity is considered in Eqs. (2a) and (2b).

Our second-harmonic generation measurements were conducted at room temperature without bias field. In this case, there is no electromechanical coupling to the fundamental acoustic wave but only electrostrictive coupling. The ceramic sample can be treated as isotropic. For a longitudinal wave propagating in an isotropic medium, the Eqs. (1a)–(2b) can be simplified as

$$\rho_0 \ddot{u}_1 = T_{11,1}, \tag{3}$$

$$T_{11} = c_{11} u_{1,1} + \tfrac{1}{2}(3c_{11} + c_{111}) u_{1,1}^2. \tag{4}$$

This is a pure elastic nonlinear equation. The solution of Eq. (3) is

$$u_1 = A_1 \sin(\omega t - kx) - \tfrac{1}{8}\beta k^2 x_x A_1^2 \cos 2(\omega t - kx), \tag{5}$$

where A_1 and A_2 are the amplitudes of fundamental and second-harmonic waves, respectively, k is the wave number, ω is the angular frequency, x is the distance in the wave propagation direction, and β is the ultrasonic nonlinearity parameter of the material defined as

$$\beta = -\frac{3c_{11} + c_{111}}{c_{11}}. \tag{6}$$

From Eq. (5), it is seen that β can be experimentally determined if the absolute amplitudes of A_1 and A_2 are known

$$\beta = \frac{8}{k^2 L}\frac{A_2}{A_1^2}, \tag{7}$$

where L is the sample length. In order to measure A_1 and A_2, the receiving transducer is calibrated by following the calibration procedure given in Ref. 8. Once β is determined, the third-order elastic constant c_{111} can be calculated by

$$c_{111} = -(3 + \beta) c_{11}, \tag{8}$$

where the second-order elastic constant $c_{11} = \rho_0 v^2$ can be determined by the material density ρ_0 and the longitudinal wave velocity v.

The measured results are listed in Table I. It is seen that the nonlinearity parameter β of La-doped PMN-PT is twice as large as that of common single crystals ($\beta = 2-14$)[9] and normal ferroelectric lead zirconate–lead titanete (PZT) ceramics ($\beta = 3-18$).[10,11]

Usually, the electromechanical devices using electrostrictive material as the active part are biased by an external electric field. Under this situation there is an electromechanical coupling nonlinearity in addition to the pure elastic nonlinearity mentioned above. Macroscopically, relaxor systems, such as the PZN-PT system under study, have a center of symmetry, therefore, the third order tensors coefficients are all vanished and the constitutive equations for a biased electrostrictive material can be expressed as

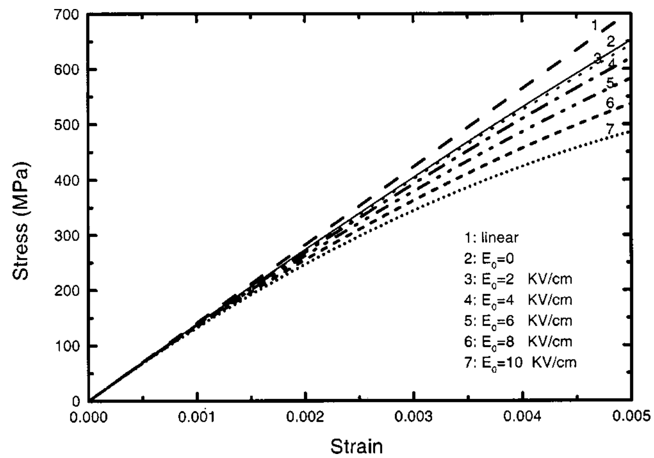

FIG. 2. Strain–stress relation for lanthanum doped PMN-PT under different bias field. The solid line is for the case without coupling nonlinearity.

$$T_{11} = c_{11}^E u_{1,1} + \tfrac{1}{2}(3c_{111}^E + c_{111}) u_{1,1}^2 - \tfrac{1}{2} m_{11} E_1^2, \tag{9}$$

$$D_1 = \varepsilon_{11} E_1 + m_{11} E_1 u_{1,1}, \tag{10}$$

and

$$E_1 = E_0 + E(x,t), \tag{11}$$

where E_0 and $E(x,t)$ are applied bias and electrostrictively generated internal electric fields, respectively. From equation (1b), i.e., $D_{1,1} = 0$, the electrostrictively generated fundamental electric field $E(x,t)[\ll E_0]$ can be calculated as

$$E(x,t) \approx -\frac{m_{11} E_0}{\varepsilon_{11}} u_{1,1}. \tag{12}$$

Substituting the result into Eq. (9), the nonlinear strain–stress relation can be obtained as

$$T_{11} = \left(\overline{c_{11}} + \frac{m_{11}^2 E_0^2}{\varepsilon_{11}}\right) u_{1,1} + \tfrac{1}{2}\left(3c_{11} + c_{111} - \frac{m_{11}^3 E_0^2}{\varepsilon_{11}^2}\right) u_{1,1}^2. \tag{13}$$

Thus, one can define an effective nonlinearity parameter

$$\beta_e = -\frac{3c_{11} + \overline{c_{111}}}{\overline{c_{11}}}, \tag{14}$$

where

$$\overline{c_{11}} = c_{11} + \frac{m_{11}^2 E_0^2}{\varepsilon_{11}},$$

$$\overline{c_{111}} = c_{111} - \frac{m_{11}^3 E_0^2}{\varepsilon_{11}^2}.$$

Using the measured values of $m_{11} = 8.5 \times 10^{-5}$ F/m, $\varepsilon_{11} = 2 \times 10^4 \varepsilon_0$ ($\varepsilon_0 = 8.86 \times 10^{-12}$ F/m), and $c_{11} = 14.1\ N/m^2$,[12] we can calculate the effective nonlinearity parameter β_e and the effective third order elastic constant $\overline{c_{111}}$. Then, the nonlinear strain–stress relation for the PMNPT sample can be calculated. Figure 2 shows a few curves for several values of the bias field. The solid line is for the case of zero external

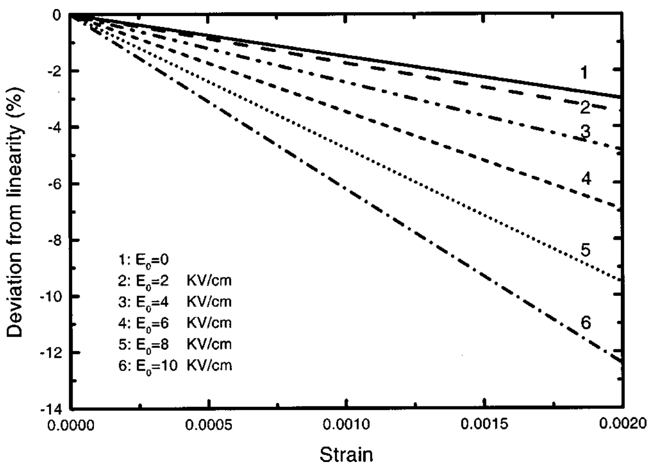

FIG. 3. Relative deviation form linear strain–stress relation for lanthanum doped PMN-PT under different bias field.

fields. We can see that the system is quite nonlinear. This nonlinearity increases with bias field level and the adjustability is fairly large. We show the relative change of the nonlinearity caused by the applied electric field in Fig. 3.

By defining an effective nonlinearity parameter, we have also successfully quantified the relative change of the nonlinearity induced by the application of electric field. The strong dependence of β_e on the bias-electric field suggests a way to the control nonlinearity of PMN-PT, which may be used to design field tunable nonlinear electroacoustic devices.

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