Second Harmonic Generation of Shear Waves in Crystals

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***Abstract*—Nonlinear self-interaction of shear waves in electro-elastic crystals is investigated based on the rotationally invariant state function. Theoretical analyses are conducted for cubic, hexagonal, and trigonal crystals. The calculations show that nonlinear self-interaction of shear waves has some characteristics distinctly different from that of longitudinal waves. First, the process of self-interaction to generate its own second harmonic wave is permitted only in some special wave propagation directions for a shear wave. Second, the geometrical nonlinearity originated from finite strain does not contribute to the second harmonic generation (SHG) of shear waves. Therefore, unlike the case of longitudinal wave, the second-order elastic constants do not involve in the nonlinear parameter of the second harmonic generation of shear waves. Third, unlike the nonlinearity parameter of the longitudinal waves, the nonlinear parameter of the shear wave exhibits strong anisotropy, which is directly related to the symmetry of the crystal. In the calculations, the electromechanical coupling nonlinearity is considered for the 6mm and 3m symmetry crystals. Complement to the SHG of longitudinal waves already in use, the SHG of shear waves provides more measurements for the determination of third-order elastic constants of solids. The method is applied to a Z-cut lithium niobate ($LiNbO_3$) crystal, and its third-order elastic constant c_{444} is determined.**

I. Introduction

The three-phonon interaction in solids has been extensively investigated [1]–[11], and the second harmonic generation (SHG) is a special case of this three-phonon interactions, i.e., a phonon with twice of the frequency of the input phonon is generated through a process of the input fundamental phonon interacts with itself (self-interaction) due to the nonlinearity of solids [7]. This SHG can be used to determine third-order elastic constants (TOECs) of single- and poly-crystals [2]. In the SHG measurements, amplitudes of the fundamental and the second harmonic longitudinal wave are determined. Then, a so-called ultrasonic nonlinearity parameter β is obtained [2]. The ultrasonic nonlinearity parameter, which characterizes the SHG of longitudinal waves, involves combinations of the second- and the third-order elastic (TOECs) constants. With the known second-order elastic constants, a certain TOEC or a combination of TOECs can be determined.

However, in previous studies the SHG of the shear waves always has been ignored due to the following two reasons. First, the process of self-interaction of a shear wave to generate its own second harmonic wave is prohibited for isotropic solid [3] and in many purely polarized wave directions for other symmetries (details will be given in this paper). Second, there is no reported method to measure the absolute amplitude of a shear wave.

However, from the viewpoint of nonlinear property characterization of materials, the SHG of shear wave can be a powerful tool. First, the propagation and polarization directions are different in a shear wave, which can help to probe varieties of processes that are polarization sensitive. As will be presented in this paper, the SHG of shear waves is strongly dependent upon polarization direction. Second, the quadratic nonlinearity of a longitudinal wave comes from both material (nonlinear Hooke's law) and geometrical (finite strain) nonlinearities, and includes electromechanical nonlinearity if piezoelectric coupling exists. But the quadratic nonlinearity of a purely polarized shear wave only reflects the material nonlinearity. Therefore, it is a more direct manifestation of the material nonlinear properties. Third, unlike the longitudinal nonlinearity, we found that the quadratic nonlinearity of shear waves often exhibits particular symmetry, which is associated with the symmetry of the crystal itself.

If one only uses the SHG of longitudinal wave to determine the TOECs, there is not enough equations along high symmetric purely polarized wave directions to isolate all of the independent TOECs. For example, for a cubic crystal there are six independent TOECs, but one can only measure the second harmonic longitudinal wave in [100], [010], and [111] directions. Thus, at least three more equations are needed to uniquely determine the six independent TOECs [4]. If SHG of the shear waves can be used, it will add more combinations of the TOECs to help resolve this problem.

In the present paper, SHG of shear waves in crystals is investigated based on the rotationally invariant state function. In the first part of this paper, the state function and basic equations for the nonlinear process are given. Based on this, purely polarized wave directions for shear waves in nonlinear regime are discussed. The quadratic nonlinearity of shear waves is calculated for cubic, hexagonal, and trigonal symmetry crystals in those directions in which the nonlinearity exists. The corresponding nonlinearity parameters are provided. Also, the contribution of electromechanical coupling nonlinearity to SHG is considered for 6mm and 3m symmetry crystals. The experimental results for a Z-cut $LiNbO_3$ crystal are reported.

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II. The State Function and Nonlinear Electro-Acoustic Equations for Anisotropic Crystals

The equation of motion describes a thermodynamically reversible process for an electro-elastic crystal. Therefore, its state can be described by a potential or scalar state function $\psi(\eta, \mathbf{W})$, which is rotationally invariant if η and $\mathbf{W}$ defined below are taken as independent variables [5]. Here η is the Green's finite strain tensor:

$$\eta_{IJ} = \frac{1}{2}\left(\frac{\partial u_I}{\partial X_J} + \frac{\partial u_J}{\partial X_I} + \frac{\partial u_K}{\partial X_I}\frac{\partial u_K}{\partial X_J}\right), \tag{1}$$

and W is related to the ordinary spatial electric field $\mathbf{E}^*$ by:

$$W_L = E_i^* x_{i,L}, \tag{2}$$

where X in (1) is the position vector of a matter point at the reference state, i.e., at time t_0, and x in (2) is the position vector of the same matter point at the final state, i.e., at time t, and u is the particle displacement. When X and t are taken as the independent variables, one uses the material description, and one uses spatial description when x and t are taken as independent variables. The lower and upper case letters represent quantities in spatial coordinate and material coordinate, respectively. Also, the comma in subscript stands for differentiation with respect to corresponding spatial or material coordinate, e.g., $x_{i,L} = \partial x_i/\partial X_L$.

For an electro-elastic crystal, the state function can be expanded into Taylor's series with respect to its reference state for which the strain and electric field are both zero [9]:

$$\begin{aligned}\psi = {} & \frac{1}{2}c_{IJKL}\eta_{IJ}\eta_{KL} - e_{IJK}W_I\eta_{JK} - \frac{1}{2}\chi_{IJ}W_IW_J \\ & + \frac{1}{6}c_{IJKLMN}\eta_{IJ}\eta_{KL}\eta_{MN} - \frac{1}{2}e_{MIJKL}W_M\eta_{IJ}\eta_{KL} \\ & - \frac{1}{2}b^S_{IJKL}W_IW_J\eta_{KL} - \frac{1}{6}\chi_{IJK}W_IW_JW_K.\end{aligned} \tag{3}$$

Up to cubic terms are preserved in (3). The material constants c_{IJKL} and c_{IJKLMN} are the second- and third-order elastic constants, e_{IJK} are linear piezoelectric constants, χ_{IJ} and χ_{IJK} are the second- and third-order dielectric susceptibility, b^S_{IJKL} are the electrostrictive constants, and e_{MIJKL} are the third-order piezoelectric coefficients.

The equation of motion and quasistatic electric equation can be written as:

$$\rho_0\ddot{u}_I = \delta_{iI}T_{iJ,J}, \qquad (I, J, K = 1, 2, 3), \tag{4a}$$

$$D_{L,L} = 0, \qquad (L = 1, 2, 3), \tag{4b}$$

where T_{iJ} is the total Piola-Kirchhoff stress tensor (material plus Maxwell electrostatic stress):

$$T_{iJ} = P_{iJ} + M_{iJ}, \tag{5a}$$

$$P_{iJ} = x_{i,K}\frac{\partial\psi}{\partial\eta_{KJ}}, \tag{5b}$$

$$M_{iJ} = JX_{J,k}m_{ki}, \tag{5c}$$

$$m_{ki} = \varepsilon_0\left(E_i^*E_k^* - \frac{1}{2}E_j^*E_j^*\delta_{ik}\right), \tag{5d}$$

where $J = \det\left(\frac{\partial x_i}{\partial X_J}\right)$ is the Jacobian, δ_{ik} is the Kronecker delta, and ε_0 is the dielectric permittivity of free space.

The material electric displacement D is related to ordinary spatial electric displacement D^* by,

$$D_L = JX_{L,i}D_i^*, \tag{6a}$$

and

$$D_L = \varepsilon_0E_L + P_L, \tag{6b}$$

where E is related to ordinary spatial electric field E^* by:

$$E_L = JX_{L,i}E_i^*. \tag{6c}$$

Let

$$E_1^* = -\phi_{,1}, \tag{6d}$$

from (2), one can obtain:

$$W_L = -\phi_{,L}, \tag{6e}$$

where ϕ is the electric potential. In (6b) P_L is the material electric polarization,

$$P_L = -\frac{\partial\psi}{\partial W_L}. \tag{6f}$$

Based on the above expressions, the constitutive equations up to square terms can be expressed as:

$$\begin{aligned}T_{iJ} = \delta_{iI}\Big[& c_{IJKL}u_{K,L} + e_{MIJ}\phi_{,M} + \frac{1}{2}c_{IJKL}u_{S,K}u_{S,L} \\ & + c_{MJKL}u_{I,M}u_{K,L} + \frac{1}{2}c_{IJKLMN}u_{K,L}u_{M,N} \\ & + e_{MKJ}u_{I,K}\phi_{,M} + e_{MIJKL}u_{K,L}\phi_{,M} \\ & - \frac{1}{2}b_{MNIJ}\phi_{,M}\phi_{,N}\Big],\end{aligned} \tag{7a}$$

$$\begin{aligned}D_L = {} & -\varepsilon_{LM}\phi_{,M} + e_{LIJ}u_{I,J} + \frac{1}{2}e_{LIJ}u_{S,I}u_{S,J} \\ & + \frac{1}{2}e_{LIJKL}u_{I,J}u_{K,L} - b_{LMIJ}u_{I,J}\phi_{,M} \\ & + \frac{1}{2}\chi_{LMN}\phi_{,M}\phi_{,N},\end{aligned} \tag{7b}$$

where ε_{LM} are the components of the linear dielectric permittivity tensor, b_{LMIJ} are components of the electrostrictive tensor.

III. SHG of Shear Waves in Nonpiezoelectric Crystal

For simplicity, the piezoelectric effect is not considered in the first step. Although 20 of the 32 crystal groups are piezoelectric, it will be shown that there exist some directions in a piezoelectric crystal along which the propagating shear wave does not couple to piezoelectricity.

A. Purely Polarized Shear Acoustic Waves

Most of SHG experiments are conducted using purely polarized acoustic waves for simplicity and easy realization. Purely polarized waves and purely polarized wave directions have been defined by Brugger [6]. (Brugger used the term pure mode. Here we use purely polarized waves to avoid the confusion with the modes for guided waves.) Brugger's calculations were for infinitesimal amplitude acoustic waves, i.e., in the linear regime. For large but finite amplitude acoustic waves, the nonlinear effects become significant. In the nonlinear regime, the purely polarized wave concept for the shear waves is not exact. It is known that a finite amplitude acoustic wave will encounter waveform distortion when it propagates in a nonlinear medium, i.e., noticeable second harmonic will be generated. When a longitudinal wave propagates in a crystal along a purely polarized wave direction defined for infinitesimal amplitude acoustic wave, the self-interaction of the fundamental longitudinal wave only generates its own second harmonic wave. Neither fundamental nor second harmonic shear waves are produced. Thus, purely polarized wave is meaningful for a longitudinal wave, even for the case of a finite amplitude acoustic wave. But the situation is different for shear waves. It has been noticed that the self-interaction of a shear wave does not generate its own second harmonic wave but the second harmonic wave of the longitudinal wave instead, when it propagates in an isotropic solid [3]. In anisotropic solids, it also is possible that the self-interaction of one kind of shear wave generates the second harmonic of another kind of shear wave [7]. Therefore, there always exist the second harmonic waves of either quasilongitudinal wave or quasishear wave accompanying the fundamental shear wave. Based on this fact, it is argued that the purely polarized wave does not exist for a shear wave when nonlinearity, which is inherent for all solids, is taken into account.

However, it is known that the second harmonic longitudinal wave generated by the self-interaction of a fundamental shear wave is not phase-matched [8] because velocities of the two waves are much different. Usually, the two shear waves in anisotropic solids will have different velocities; therefore, the second harmonic of one kind of shear wave generated by the self-interaction of another kind of fundamental shear wave also is not phase matched. Lacking of phase matching makes it impossible to accumulate the energy of the second harmonic waves with the propagation of the fundamental wave. Therefore, such nonphase matched second harmonics exist but are always very weak.

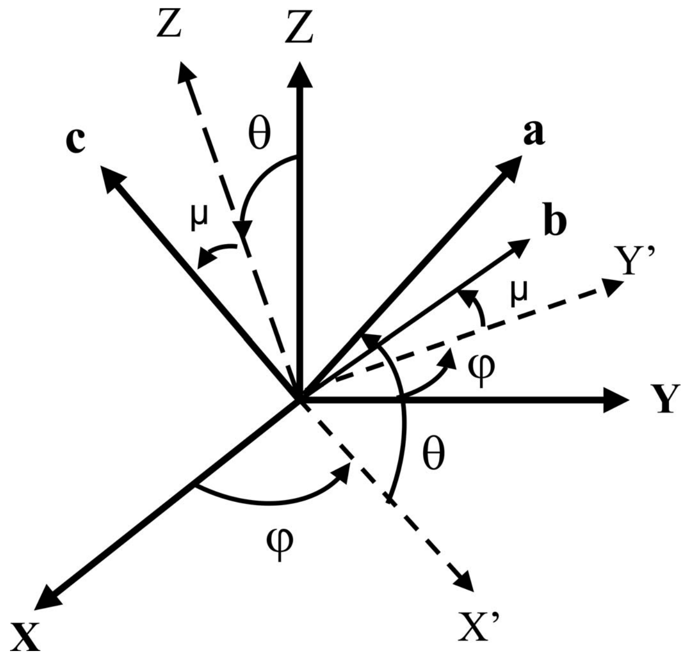

Fig. 1. Transformation from the constitutive coordinate system to the working coordinate system.

However, if a shear wave can generate its own second harmonic wave for a given wave propagation direction, the process is phase matched, and the energy of the generated second harmonic shear wave will accumulate with propagation distance. Thus, when the wave propagates for a finite distance, the nonphase matched second harmonic waves can be ignored compared to the phase matched second harmonic shear wave. Under this circumstance, one can assume the shear wave to be a purely polarized wave and the given wave propagation direction a purely polarized wave direction. Using this definition, the concept of purely polarized wave in linear regime still can be used in the nonlinear regime. This means that one can use the way similar to that for SHG of longitudinal waves to investigate the SHG of shear waves.

The state function (3) is given in a coordinate system coincident with constitutive coordinate system (XYZ) of a crystal. For a plane wave propagating along an arbitrary direction, a new coordinate system (abc) can be constructed through a rotational transformation. Hereafter, the discussion is performed always in material coordinates; hence, the distinction between the lower and upper case will be ignored. In the rotated coordinate system, the a-axis is chosen to be coincident with the wave propagation direction, and the b- and c-axes are chosen to be orthogonal to a and make (abc) a right-hand coordinate system. The particle displacement $\mathbf{u}(u_1, u_2, u_3)$ is a function of a only for a plane wave, which renders the problem one-dimensional. We call the coordinates (abc) the working coordinate system.

As shown in Fig. 1, the working coordinate system can be obtained by rotating the constitutive coordinate (XYZ) three times. First the coordinate is rotated about the Z-axis by φ to form a coordinate $(X'Y'Z)$. Then, the $(X'Y'Z)$ coordinate is rotated about Y' by θ to form coordinate $(aY'Z')$. The $(aY'Z')$ coordinate then is rotated about a by μ to form the working coordinate system (abc). The former two rotations decide the wave propagation direction; the third rotation gives the particle displacement direction of the shear waves. Obviously, the vector W (W_X, W_Y, W_Z) in the old system (XYZ) can be expressed as a vector $W' = (W'_a, W'_b, W'_c)$ in the new system (abc) by a simple transformation:

$$[W] = [\alpha]^T \cdot [W']. \tag{8}$$

Also the strain tensor η in the (XYZ) system can be represented by the strain tensor η' in (abc) through:

$$[\eta] = [\alpha]^T \cdot [\eta'] \cdot [\alpha], \tag{9}$$

where $[\alpha]$ is the coordinate transformation matrix, $[\alpha]^T$ is its transpose (10) (see next page). Thus, the state function, which is rotationally invariant, can be expressed as the function of W' and η' by substituting (8) and (9) into (3), i.e., $\psi = \psi(\eta', W')$.

For a plane wave in the working system (abc) the equation of motion without considering piezoelectric effect can be written as:

$$\rho_0 \ddot{u}_i = \frac{\partial P_{i1}}{\partial a}, \tag{11a}$$

where

$$P_{i1} = \left[(u_{i,k} + \delta_{ik}) \frac{\partial \psi}{\partial \eta_{k1}} \right], \tag{11b}$$

$(i, j, k = a, b, c \text{ or } 1, 2, 3)$. Here the primes, upper and lower cases are ignored, but u_i and η_{ij} should be considered as particle displacement and strain component in the working coordinate system. To find the purely polarized wave directions under the assumption given above, only the linear part of (11) needs to be analyzed. If one can choose the angle φ, θ, and μ to make the coefficients before the product terms $\eta_{11}(\eta_{21}+\eta_{12})$ and $\eta_{11}(\eta_{31}+\eta_{13})$ in the state function to become zero simultaneously, one has found the first kind of purely polarized wave direction [6]. In this direction the longitudinal and two shear waves are all purely polarized waves. If the product terms $\eta_{11}(\eta_{21}+\eta_{12})$ or $\eta_{11}(\eta_{31}+\eta_{13})$ do not appear in the expression of state function at all for certain symmetry crystals, one has the second kind of purely polarized wave direction. Along such a direction, one of the shear waves is purely polarized and another shear wave couples to the longitudinal wave; hence, they are quasilongitudinal and quasishear waves, respectively. By using this method we have searched for purely polarized wave directions of shear waves for cubic, hexagonal, and trigonal crystals. The results are in agreement with that calculated by Brugger in the linear regime [6]. After angles φ, θ, and μ have been found, one can investigate the SHG in those purely polarized wave directions.

B. Second Harmonic Generation of Shear Waves in Purely Polarized Wave Directions

To investigate the process of self-interaction of a shear wave that generates its own second harmonics, the partial differentiations of the state function with respect to strain components in (11) are calculated in the working coordinate system. For a purely polarized shear wave, the bilinear equation of motion finally can be expressed as:

$$\rho_0 \ddot{u}_i - K_2 u_{i,11} = K_3 u_{i,1} u_{i,11} \qquad (i = 2 \text{ or } 3). \tag{12}$$

Here u is the particle displacement in a purely polarized wave. The phase-matched second harmonic generation is permitted for shear waves if K_3 in (12) is nonzero; (12) has the same form as that for a purely polarized longitudinal wave. But K_3, the coefficient before the nonlinear term $u_{i,1}u_{i,11}$ in (12) involves only the third-order elastic constants or their combination for shear waves, whereas it involves both second-order and third-order elastic constants for a longitudinal wave. This means that the quadratic nonlinearity could come from both material and geometry nonlinearity for a purely polarized longitudinal wave, but only from the material nonlinearity for a shear wave as shown below.

Assuming that there is a purely polarized shear wave, without losing generality, we may set $u_1 = u_3 = 0$ and $u_2 = u_2(a, t)$. From (1) we have:

$$[\eta] = \begin{bmatrix} \frac{1}{2}u_{2,1}^2 & \frac{1}{2}u_{2,1} & 0 \\ \frac{1}{2}u_{2,1} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}. \tag{13}$$

It is seen that η_{12} (or η_{21}) is always a linear strain for a purely polarized shear wave. In the working coordinate system, the bilinear equation for u_2 can be rewritten as:

$$\rho_0 \ddot{u}_2 = \frac{\partial}{\partial a} \left[\frac{\partial \psi_2}{\partial \eta_{21}} + \left(\frac{\partial u_2}{\partial a} \frac{\partial \psi_2}{\partial \eta_{11}} + \frac{\partial \psi_3}{\partial \eta_{21}} \right) \right], \tag{14}$$

where

$$\psi_2 = \frac{1}{2} c_{IJKL} \eta_{IJ} \eta_{KL}, \tag{15a}$$

$$\psi_3 = \frac{1}{6} c_{IJKLMN} \eta_{IJ} \eta_{KL} \eta_{MN}, \tag{15b}$$

are, respectively, the second-order and third-order state functions.

It is seen that the second-order elastic constants may enter K_3 through the terms $\frac{\partial \psi_2}{\partial \eta_{21}}$ and $\frac{\partial u_2}{\partial a}\frac{\partial \psi_2}{\partial \eta_{11}}$. For a purely polarized wave direction, the term $\eta_{11}(\eta_{12}+\eta_{21})$ must not appear in the expression of ψ_2, as pointed out above. Thus, the term $\frac{\partial \psi_2}{\partial \eta_{21}} \propto \eta_{21}$ will not generate quadratic term of $u_{2,1}$ as there is no nonlinear term in η_{21}. Because $\frac{\partial \psi_2}{\partial \eta_{11}} \propto \eta_{11} = \frac{1}{2}u_{2,1}^2$, $\frac{\partial u_2}{\partial a}\frac{\partial \psi_2}{\partial \eta_{11}}$ will not generate square terms but only cubic terms of $u_{2,1}$; no second-order elastic constants can be involved in K_3 for a purely polarized shear wave.

$$[\alpha] = \begin{bmatrix} \cos\theta\cos\varphi & \cos\theta\sin\varphi & \sin\theta \\ -\cos\mu\sin\varphi - \sin\theta\cos\varphi\sin\mu & \cos\mu\cos\varphi - \sin\theta\sin\varphi\sin\mu & \sin\mu\cos\theta \\ \sin\mu\sin\varphi - \sin\theta\cos\varphi\cos\mu & -\sin\mu\cos\varphi - \sin\theta\sin\varphi\cos\mu & \cos\mu\cos\theta \end{bmatrix}. \tag{10}$$

This distinctive feature makes the SHG of shear waves a straight manifestation of material nonlinear properties.

The solution of (14) can be obtained by using the perturbation method of successive approximation [9], i.e., let

$$u_2 = \varepsilon u_2^{(0)} + \varepsilon^2 u_2^{(1)} + \cdots, \tag{16}$$

where ε is a constant (<1) indicating the order of the magnitude of the successive terms in (16), and put it into (14). The equation of motion can be separated as:

$$\rho_o \ddot{u}_2^{(0)} - K_2 u_{2,22}^{(0)} = 0 \qquad \text{for order } \varepsilon, \tag{17a}$$

$$\rho_0 \ddot{u}_2^{(1)} - K_2 u_{2,22}^{(1)} = K_3 u_{2,2}^{(0)} u_{2,22}^{(0)} \qquad \text{for order } \varepsilon^2. \tag{17b}$$

Thus,

$$u_2 = u_2^{(0)} + u_2^{(1)} = A_1 \sin(\omega t - ka) - A_2 \cos 2(\omega t - ka), \tag{18}$$

where k is wave-number and A_2 is the amplitude of the second harmonic wave:

$$A_2 = \frac{1}{8} k^2 a A_1^2 \beta_T, \tag{19}$$

where β_T is the nonlinearity parameter defined by:

$$\beta_T = -\frac{K_3}{K_2}. \tag{20}$$

The nonlinearity parameter β_T characterizes the SHG of shear waves and is a description of the shear nonlinear property of the material.

We have calculated K_2, K_3, and the nonlinear parameter of SHG of shear waves for crystals with cubic symmetry group (432, 43m m$\overline{3}$m) and (23, m3), hexagonal (622, 6mm, $\overline{6}$m2, 6/mm), and trigonal (32, 3m, $\overline{3}$m). To find the expression of K_3 for a purely polarized shear wave, one simply calculates the differentiation $\frac{\partial\psi_3}{\partial\eta_{21}}$ or $\frac{\partial\psi_3}{\partial\eta_{31}}$. It is found that $K_3 = 0$ for many purely polarized shear waves, as mentioned above. The nonzero K_3 and corresponding K_2 along with their propagation and polarization directions are listed in Table I for these crystals.

Two classes of cubic crystals (432, 43m, m3m) and (23, m3) have the same second-order elastic constants. But their numbers of independent TOECs are different with the former to be 6 and the later to be 8, respectively. For cubic crystals, the three principle axes directions, the face diagonal and body diagonal directions are purely polarized wave directions. But SHG is permitted only for the shear wave propagating along body diagonals. The nonlinearity parameter of the shear wave propagating along the three-fold axis exhibits three-fold symmetry when the polarization direction is rotated. The shear wave propagating in a plane passing through a surface diagonal and polarizing normal to the plane is also a pure polarized shear wave. But the SHG of this purely polarized wave is permitted only for the second cubic crystal class for which $c_{112} \neq c_{113}$ and $c_{155} \neq c_{166}$.

In a hexagonal crystal (622, 6mm, $\overline{6}$m2, 6/mm), the shear wave with polarization perpendicular to the six-fold symmetric axis (Z-axis) is always a purely polarized wave irrespective of the propagation direction. For this shear wave, it is found that K_2 is independent of φ. Therefore, the velocity of the shear wave exhibits shear isotropy. Whereas K_3, i.e., the nonlinearity parameter is zero when $\varphi = n\pi/3$ ($n = 1, 2, \ldots, 6$) exhibiting a six-fold rotational symmetry. When $\varphi \neq n\pi/3$, K_3 exhibits a two-fold symmetry, or $K_3(\varphi) = K_3(\varphi + \pi)$. From this we conclude that, for materials with Curie symmetry ∞m (Curie symmetry refers to shear isotropic symmetry) [12], for example piezoelectric ceramics, there must exist the relation $c_{111} = c_{222}$ (or $c_{166} = c_{266}$ because $c_{111} - c_{222} = c_{166} - c_{266}$ for hexagonal crystal). The reason is that $K_3 = 0$ when the wave propagates along the X-axis or in the direction at an angle $\varphi = n\pi/3$ with respect to the X-axis and polarizes normal to the six-fold or Z-axis of a hexagonal crystal. Because one cannot identify the X- from Y-axis for materials with symmetry ∞m, K_3 should be zero for all φ, which demands that $c_{111} = c_{222}$ or $c_{166} = c_{266}$. Thus the independent TOECs is 9 for materials with ∞m symmetry but 10 for 6mm crystals. The conclusion is in agreement with that given by Fritz [13] who derived the same results by using the method of rotating the sixth-order tensor based on group theoretical arguments. In a hexagonal crystal, the shear wave with polarization parallel to the six-fold (Z) axis is always a purely polarized wave when propagating in the XY plane. But the SHG is prohibited for those waves. The SHG is permitted only when the polarization deviates from the six-fold axis by an angle $\theta_p \neq 0$.

In trigonal crystals (32, 3m, $\overline{3}$m), the shear wave with polarization normal to the three-fold axis is always a pure mode. It is seen from Table I that the nonlinearity parameter of the wave exhibits a three-fold rotational symmetry with respect to the rotation of the polarization direction. The SHG is permitted only when $\varphi \neq (2n+1)\pi/6$. When the wave propagation direction is in the YZ plane of trigonal crystals and by an angle $\theta_r \neq 0$ with respect to the Y-axis, the shear wave polarized at the direction by the same angle θ_r with respect to the Z-axis is a purely polarized shear wave. Its nonlinearity parameter involves 10 of the 14 independent TOECs.

TABLE I

K_2 AND K_3 FOR CUBIC (432, 43M, M$\overline{3}$M) AND (23, M3), HEXAGONAL (622, 6MM, $\overline{6}$M2, 6/MM), AND TRIGONAL (32, 3M, $\overline{3}$M) CRYSTALS WITHOUT PIEZOELECTRICITY.

Cubic (432, 43m, m$\overline{3}$m)	
(N_1, N_2, N_3)	$\left(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}\right)$
(U_1, U_2, U_3)	$\left(\frac{1}{\sqrt{2}}\left(-\frac{1}{\sqrt{3}}\cos\mu + \sin\mu\right), -\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{3}}\cos\mu + \sin\mu\right), \sqrt{\frac{2}{3}}\cos\mu\right)$
K_2	$\frac{1}{3}(c_{11} - c_{12} + c_{44})$
K_3	$\frac{\sqrt{2}}{18}(c_{111} - 3c_{112} + 3c_{144} - 3c_{166} + 3c_{123} - 2c_{456})\cos(3\mu)$
Cubic (23, m3)	
(N_1, N_2, N_3)	1) $\left(\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}\right)$ 2) $\left(\frac{1}{\sqrt{2}}\cos\theta, \frac{1}{\sqrt{2}}\cos\theta, \sin\theta\right)$
(U_1, U_2, U_3)	1) $\left(\frac{1}{\sqrt{2}}\left(-\frac{1}{\sqrt{3}}\cos\mu + \sin\mu\right), -\frac{1}{\sqrt{2}}\left(\frac{1}{\sqrt{3}}\cos\mu + \sin\mu\right), \sqrt{\frac{2}{3}}\cos\mu\right)$ 2) $\left(-\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0\right)$
K_2	1) $\frac{1}{3}(c_{11} - c_{12} + c_{44})$ 2) $\frac{1}{2}(c_{11} - c_{12})\cos^2\theta + c_{44}\sin^2\theta$
K_3	1) $\frac{\sqrt{2}}{8}\left[c_{111} - \frac{3}{2}(c_{112} + c_{113}) - \frac{3}{2}(c_{115} + c_{116}) + 3c_{144} + 3c_{123} - 2c_{456}\right]\cos(3\mu)$ 2) $\frac{3}{8}\left[(c_{112} - c_{113})\cos^3\theta - (c_{155} - c_{166})\right]\sin\theta\cos(2\theta)$
Hexagonal (622, 6mm, $\overline{6}$m2, 6/mm)	
(N_1, N_2, N_3)	1) $(\cos\theta\cos\varphi, \cos\theta\sin\varphi, \sin\theta)$ 2) $(\cos\theta_p\cos\varphi, \cos\theta_p\sin\varphi, \sin\theta_p)$
(U_1, U_2, U_3)	1) $(-\sin\varphi, \cos\varphi, 0)$ 2) $(-\sin\theta_p\cos\varphi, -\sin\theta_p\sin\varphi, \cos\theta_p)$
K_2	1) $\frac{1}{2}(c_{11} - c_{12})\cos^2\theta + c_{44}\sin^2\theta$ 2) $\frac{1}{4}(c_{11} + c_{33} - 2c_{13})\sin^2(2\theta_p) + c_{44}\cos^2(2\theta_p)$
K_3	1) $\frac{1}{2}(c_{111} - c_{222})\cos^3\theta\sin(2\varphi)\left[\sin^2(2\varphi) - 3\cos^2(2\varphi)\right]$ 2) $\frac{1}{8}(-c_{111} + 3c_{113} - 3c_{133} + c_{333})\sin^3(2\theta_p)$ $+\frac{1}{8}(c_{111} - c_{222})\sin^3(2\theta_p)\sin^2\varphi\left(\sin^2\varphi - 3\cos^2\varphi\right)^2$ $+\frac{3}{2}(c_{344} - c_{155})\cos^2(2\theta_p)\sin(2\theta_p)$
$\theta_p = \tan^{-1}\left(\frac{c_{11} - c_{13} - 2c_{44}}{c_{33} - c_{13} - 2c_{44}}\right)^{1/2}$	
Trigonal (32, 3m, $\overline{3}$m)	
(N_1, N_2, N_3)	1) $(0, 0, 1)$ 2) $(0\cos\theta_r, \sin\theta_r)$
(U_1, U_2, U_3)	1) $(-\sin\varphi, \cos\varphi, 0)$ 2) $(0, -\sin\theta_r, \cos\theta_r)$
K_2	1) c_{44} 2) $\frac{1}{4}(c_{11} + c_{33} - 2c_{13})\sin^2(2\theta_r) + c_{44}\cos^2(2\theta_r) + \frac{1}{2}c_{14}\sin(4\theta_p)$
K_3	1) $c_{444}\cos(3\varphi)$ 2) $\left[c_{444} + \frac{3}{2}(c_{344} - c_{155})\tan 2\theta_r - \frac{3}{4}(c_{114} + 2c_{124} - 2c_{134})\tan^2(2\theta_r)\right.$ $\left. + \frac{1}{8}(c_{333} + 3c_{113} - 3c_{133} - c_{222})\tan^3(2\theta_r)\right]\cos^2(2\theta_r)$

N: wave propagation direction

U: wave polarization direction

$(c_{33} - c_{13} - 2c_{14})\tan^3\theta_r + 3c_{14}\tan^2\theta_r - (c_{11} - c_{13} - 2c_{14})\tan\theta_r - c_{14} = 0$

IV. The Influence of Piezoelectric Effect

The electromechanical coupling effect in a piezoelectric crystal usually enhances the nonlinearity, which in turn modifies the nonlinearity parameter of SHG [14]. The hexagonal and trigonal crystals calculated above are piezoelectric. It is known that, for symmetry class 622, 6mm, $\overline{6}$m2, 6/mm of hexagonal crystals, the numbers of independent second- and third-order elastic constants are the same. But the numbers of independent second- and third-order piezoelectric constants are different. The same is also true for 32, 3m, $\overline{3}$m trigonal crystals. Here we estimate the influence of piezoelectric effect only for 6mm and 3m crystals as paradigms.

We start from the electro-elastic and dielectric terms in state function (3). First, the scale function given in constitutive coordinate system of the crystals is transformed into the state function in working system through (8) and (9). Then, the differentiations of $\frac{\partial \psi}{\partial \eta'_{kl}}$ and $-\frac{\partial \psi}{\partial W'_1}$ are calculated, and W'_1 is eliminated by using the quasistatic electric equation (4b).

For a 6mm symmetry crystal, the shear wave with polarization normal to the six-fold axis has no piezoelectric coupling. The same is true for the shear wave with polarization normal to the three-fold axis for 3m crystals. Thus, the piezoelectric effects are calculated for the other two directions, and the results are listed in Table II. It is seen that the piezoelectricity can affect both the sound velocity and the nonlinearity parameter. The former becomes the stiffened velocity and the latter becomes the effective nonlinearity parameter. For 6mm crystals, the piezoelectric coupling terms in K_3 do not involve the angle φ; therefore, its symmetry does not change. Also, it is observed that the second-order piezoelectric constants are not involved in K_3, which is different from purely polarized longitudinal wave [14]. The quadratic nonlinearity of the shear waves in piezoelectric crystals comes from material and electromechanical nonlinearity, but there is no contribution from the finite strain.

V. Experimental Setup and Results

It is seen from (18) and (19) that, if the amplitudes of the fundamental (A_1) and second harmonic (A_2) waves can be experimentally measured, the ultrasonic nonlinearity parameter of shear waves β_T can be calculated by:

$$\beta_T = \frac{8}{k^2 a}\left(\frac{A_2}{A_1^2}\right). \tag{21}$$

The related TOECs or combination of TOECs can be determined by:

$$K_3 = -\beta_T K_2, \tag{22}$$

where the K_2 is determined by ultrasonic velocity measurement. There are several methods to measure the absolute value of acoustic wave amplitude [2]. But the capacitive detector and laser probe can respond only to the mechanical displacement out of the plane. They cannot be used to detect particle displacement of shear waves, which has only in-plane displacement. By using calibrated piezoelectric transducer [15], we have successfully measured the absolute amplitude of the longitudinal waves [16]. Because the principle is based on the receiving current, it should be applicable to measure the particle displacement of shear waves when shear transducers are used.

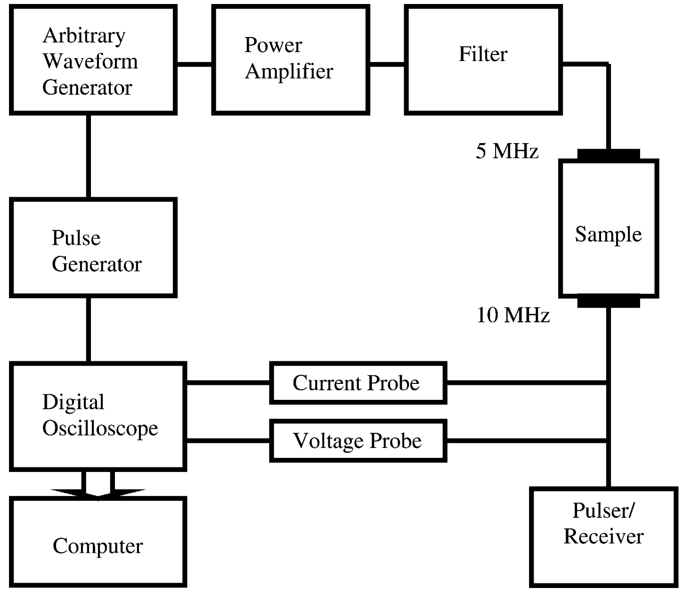

Fig. 2. The experimental setup for the measurement of ultrasonic nonlinearity parameter of shear waves.

The experimental setup is shown in Fig. 2. The setup is basically the same as that given in [17], except that shear wave transducers are used. Before doing the nonlinear measurement, the receiving transducer is calibrated. As described in [16], the calibration procedure gives the response function of the receiving transducer $H(\omega)$, and the absolute amplitude of the ultrasonic wave at frequency ω can be determined by:

$$A(\omega) = |I(\omega)| * |H(\omega)|. \tag{23}$$

After obtaining $H(\omega)$, the nonlinear measurement is simply the determination of the output currents of the receiving transducer $I(\omega)$ and $I(2\omega)$, corresponding to the fundamental and the second harmonic waves, respectively.

To demonstrate the validity of the method, the third-order elastic constant c_{444} of a Z-cut $LiNbO_3$ sample is measured. From Table I, it is seen that K_3 of the shear wave propagating along the Z-direction and polarized in the X-direction for a 3m crystal involves only c_{444}. The sample (Valpey-Fisher, Hopkington, MA) is a Z-cut $LiNbO_3$ cylinder with a diameter of 1 cm and a length of 2 cm. The PZT-4 shear wave transducers (5 MHz for the fundamental and 10 MHz for the second harmonic) are used. To test the possible nonlinearity of the measurement system itself, an aluminum sample is tested first. It is observed that the second harmonic generated in an aluminum sample is practically undetectable, which confirms that the

TABLE II

Piezoelectric corrections to K_2 and K_3 for 6mm and 3m symmetry crystals.

Hexagonal (6mm)	
(N_1, N_2, N_3)	$(\cos\theta_p \cos\varphi, \cos\theta_p \sin\varphi, \sin\theta_p)$
(U_1, U_2, U_3)	$(-\sin\theta_p \cos\varphi, -\sin\theta_p \sin\varphi, \cos\theta_p)$
K_2	$\frac{1}{4}\left[(c_{11}+c_{33}-2c_{13})+\frac{(e_{33}-e_{31})\left[(e_{33}-e_{31})\sin^2\theta_p+2e_{15}\cos(2\theta_p)\right]}{\varepsilon_{11}\cos^2\theta_p+\varepsilon_{33}\sin^2\theta_p}\right]$ $\cdot\sin^2(2\theta_p)+\left(c_{44}+\frac{e_{15}^2\cos^2\theta_p}{\varepsilon_{11}\cos^2\theta_p+\varepsilon_{33}\sin^2\theta_p}\right)\cos^2(2\theta_p)$
K_3	$\frac{1}{2}(-c_{111}+3c_{113}-3c_{133}+c_{333})\sin^3(2\theta_p)$ $+\frac{1}{2}(c_{111}-c_{222})\sin^3(2\theta_p)\sin^2\varphi\left(\sin^2\varphi-3\cos^2\varphi\right)^2$ $+\frac{3}{2}(c_{344}-c_{155})\cos^2(2\theta_p)\sin(2\theta_p)+c_{444}\cos^3(2\theta_p)$ $+\left(\frac{\tilde{e}}{\tilde{\varepsilon}}\right)\Big\{\left[(e_{135}-e_{115})\cos^2\theta_p+e_{344}\cos^2(2\theta_p)\right]\cos(2\theta_p)$ $+\frac{1}{8}(e_{333}+e_{311}-e_{313})\sin^2(2\theta_p)\Big\}\sin\theta_p$ $-\frac{1}{2}\left(\frac{\tilde{e}}{\tilde{\varepsilon}}\right)^2\left[(b_{33}-b_{31})\sin^2\theta_p-(b_{11}+b_{13})\cos^2\theta_p+2b_{44}\cos(2\theta_p)\right]\sin(2\theta_p)$ $+\left(\frac{\tilde{e}}{\tilde{\varepsilon}}\right)^3\left(6\chi_{311}\cos^2\theta_p+\chi_{333}\sin^2\theta_p\right)\sin\theta_p$

θ_p satisfies

$(B+\varepsilon_{33}D)\tan^4\theta+(C+\varepsilon_{11}D+\varepsilon_{33}E)\tan^2\theta+(A+\varepsilon_{11}E)=0$

$A=e_{15}(2e_{15}+e_{31})$ $B=e_{33}(e_{33}-e_{31}-e_{15})$ $C=(e_{33}-e_{31})(e_{31}+3e_{15})-2e_{15}^2$ $D=c_{33}-c_{13}-2c_{44}$

$E=-c_{11}+c_{13}+2c_{44}$, $\tilde{\varepsilon}=\varepsilon_{11}\cos^2\theta_p+\varepsilon_{33}\sin^2\theta_p$,

$\tilde{e}=\left[e_{15}\cos(2\theta_p)+(e_{33}-e_{31})\sin^2\theta_p\right]\cos\theta_p$

Trigonal (3m)	
(N_1, N_2, N_3)	$(0, \cos\theta_r, \sin\theta_r)$
(U_1, U_2, U_3)	$(0, -\sin\theta_r, \cos\theta_r)$
K_2	$\frac{1}{4}\left(c_{11}+c_{33}-2c_{13}+\frac{\left[(e_{33}-e_{31})\sin\theta_r-e_{22}\cos\theta_r\right]^2}{\varepsilon'_{11}}\right)\sin^2(2\theta_r)$ $+\left(c_{44}+\frac{e_{15}^2\cos^2\theta_r}{\varepsilon'_{11}}\right)\cos^2 2\theta_p$ $+\frac{1}{2}\left(c_{14}+\frac{e_{15}\left[(e_{33}-e_{31})\sin\theta_r-e_{22}\cos\theta_r\right]\cos\theta_r}{\varepsilon'_{11}}\right)\sin(4\theta_r)$
K_3	$\Big[c_{444}+\frac{3}{2}(c_{444}-c_{155})\tan(2\theta_r)-\frac{3}{4}(c_{114}+2c_{124}-2c_{134})\tan^2(2\theta_r)$ $+\frac{1}{8}(c_{333}+3c_{113}-3c_{133}-c_{222})\tan^3(2\theta_r)\Big]\cos^2(2\theta_r)$ $+\left(\frac{e'_{15}}{\varepsilon'_{11}}\right)\Big\{\frac{1}{2}\left[(e_{222}-2e_{223})\cos\theta_r+(e_{333}+e_{322}-e_{323})\sin\theta_r\right]\sin^2(2\theta_r)$ $+2(e_{244}\cos\theta_r+e_{344}\sin\theta_r)\cos^2(2\theta_r)+\left[(e_{234}-e_{244})\cos\theta_r-2e_{324}\right]\sin(4\theta_r)\Big\}$ $+\left(\frac{e'_{15}}{\varepsilon'_{11}}\right)^2\Big[b_{14}+(b_{11}-b_{13}-b_{44})\tan\theta_r-(b_{44}+b_{14}+2b_{41})\tan^2\theta_r$ $+(b_{31}-b_{33})\tan^3\theta_r\Big]\cos^4\theta_r$ $+\left(\frac{e'_{15}}{\varepsilon'_{11}}\right)^3\left(\chi_{222}\cos^3\theta_r+\chi_{333}\sin^3\theta_r+3\chi_{311}\cos^2\theta_r\sin\theta_r\right)$

N: wave propagation direction; **U**: wave polarization direction

θ_r satisfies

$(B_5+\varepsilon_{33}A_3)\tan^5\theta_r+(B_4+\varepsilon_{33}A_2)\tan^4\theta_r+(B_3+\varepsilon_{33}A_1+\varepsilon_{11}A_3)\tan^3\theta_r$
$+(B_2+\varepsilon_{11}A_2+\varepsilon_{33}A_0)\tan^2\theta_r+(B_1+\varepsilon_{11}A_1)\tan\theta_r+B_0+\varepsilon_{11}A_0=0$

$A_0=-c_{14}$, $A_1=-(c_{11}-c_{13}-2c_{44})$, $A_2=3c_{14}$, $A_3=c_{33}-c_{13}-2c_{44}$, $B_0=e_{22}e_{15}$,

$B_1=e_{15}(e_{31}+2e_{15})-e_{22}^2$, $B_2=e_{22}(e_{33}-e_{31}-e_{15})$, $B_3=(e_{31}+2e_{15})(e_{33}-e_{31}-e_{15})$,

$B_4=-e_{22}e_{33}$, $B_5=e_{33}(e_{33}-e_{31}-e_{15})$, $\varepsilon'_{11}=\varepsilon_{11}\cos^2\theta_r+\varepsilon_{33}\sin^2\theta_r$

$e'_{15}=(e_{33}-e_{31})\sin^2\theta_r\cos\theta_r-e_{22}\sin\theta_r\cos^2\theta_r+e_{15}\cos\theta_r\cos 2\theta_r$

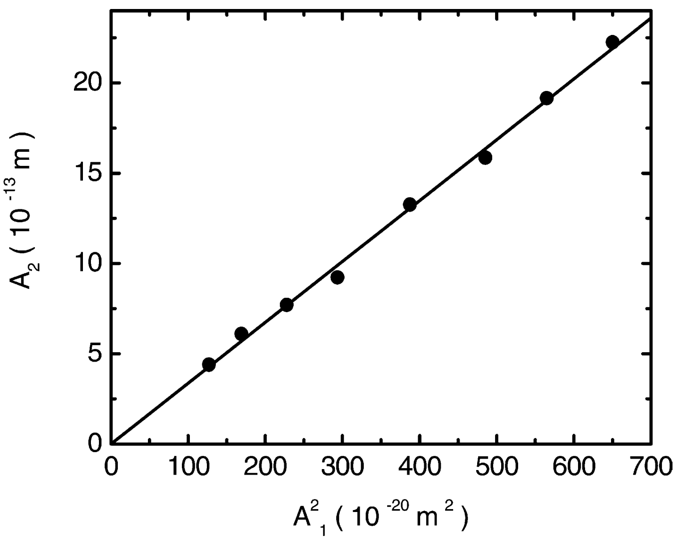

Fig. 3. The variation of the second harmonic amplitude A_1 with the square of the fundamental wave amplitude A_1^2 for $LiNbO_3$ single crystal.

TABLE III

Measured shear nonlinearity parameter and the third order elastic constant c_{444} of $LiNbO_3$ single crystal.

		c_{444}	
β	c_{44}	This work	[15]
1.7	0.6	−1.02	−0.3

Unit of c_{44} and c_{444}: 10^{11} N/m^2

SHG of shear waves cannot take place in isotropic solids. Fig. 3 gives a variation of the amplitude of the second harmonic wave with the square of the fundamental amplitude for the $LiNbO_3$ sample. The straight line is expected from (19). From the slope of the straight line, the nonlinearity parameter β_T, and hence, the shear elastic constant c_{444} of the sample are determined. The results are listed in Table III. Also listed in Table III is the c_{444} value given by Cho and Yamanouchi [17]. Our result is larger than that reported in [17] in which the measurement was made using pressure dependence of the ultrasonic velocity. But a standard error of 0.4 was reported in [17], which means an uncertainty of more than 130% for Cho's value.

VI. Summary and Conclusions

The nonlinear process of self-interaction of a shear wave, which generates its own second harmonic wave, has been investigated theoretically based on rotationally invariant scalar state function of an electro-elastic crystal. As examples, the calculations are conducted for cubic, hexagonal, and trigonal crystals. The calculations were performed without including the piezoelectricity at the beginning because most of the SHG of shear waves do not involve piezoelectricity, then the influence of piezoelectricity is considered for 6mm and 3m crystals. The nonlinear self-interaction of shear waves has some special characteristics, which make the SHG of shear waves extremely interesting to study both in terms of fundamental principles and in practice. First, the process of self-interaction of a shear wave to generate its own second harmonic wave is permitted only in some special wave propagation directions. Second, the geometrical nonlinearity originated from finite strain does not contribute to the SHG of shear waves. Therefore, unlike the case of longitudinal waves, the second-order elastic constants of the material are not involved in the nonlinear parameter of SHG in the shear waves. Third, the nonlinear parameter exhibits anisotropy directly related to the symmetry of the crystal. Such symmetry property does not exist in SHG of longitudinal waves [14]. The SHG resulting from the nonlinear self-interaction of shear waves can be used to determine third-order shear elastic constants of materials. In order to do so, it is suggested to use the calibrated shear transducer to detect the amplitudes of the fundamental and second harmonic waves. The method has been used to determine the shear third-order elastic constant c_{444} for the Z-cut $LiNbO_3$ single crystal. Our result is larger than the earlier reported result but the accuracy of the earlier result is difficult to evaluate.

In summary, we have derived the fundamental principles of the SHG method for shear waves and demonstrated the feasibility of the method, which can add a new method for the determination of the TOECs of solids.

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