

ULTRASONIC INVESTIGATION OF THE NONLINEAR
ELECTROACOUSTICAL PROPERTIES OF THE
 $\text{Bi}_{12}\text{GeO}_{20}$ ¹

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In our contribution we investigate a high frequency (h.f.) electric field by a nonlinear mixing of two, in opposite directions travelling, ultrasonic waves in a $\text{Bi}_{12}\text{GeO}_{20}$ (BGO) single crystal. We report on the nonlinear mixing of shear ultrasonic waves propagating in $[1,0,0]$ directions of the BGO crystal. Using experimental results we calculate the nonlinear elastoelectric coefficient $G = \partial C_{66}/\partial E_2$. Data on velocity and attenuation measured at room temperature are presented.

1. INTRODUCTION

The generation of high frequency (h.f.) electric field by a nonlinear mixing of two, in opposite direction travelling ultrasonic waves can be described as a two-phonon photon process, where two annihilating phonons (\vec{q}, ω) and ($-\vec{q}, \omega$) create a photon ($\vec{\gamma}, 2\omega$); \vec{q} is the wavevector of the ultrasonic wave, ω it is the angular frequency and $\vec{\gamma}$ is the wavevector of the h.f. electric field. This interaction is also known as the convolution of two signals and is mainly studied for two reasons:

1. The study of the two phonon-photon process yields information on elastoelectric nonlinear properties of the investigated material [1,13,14].
2. It is used widely in order to produce microwave signal convolvers and signal correlators [2].

In our contribution we shall deal with the generation of the h.f. electric field by the nonlinear mixing of two in opposite directions travelling ultrasonic waves in $\text{Bi}_{12}\text{GeO}_{20}$ (BGO) single crystals. We report on the nonlinear mixing of shear ultrasonic waves propagating in $[1,0,0]$ directions of the BGO crystal. Using experimental results we calculated the nonlinear elastoelectric coefficient $G = \partial C_{66}/\partial E_2$.

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The BGO single crystal was investigated with ultrasonic waves long time ago [3,4,7,8,9]. Electroacoustic two-pulse echo was also investigated [10,11,15], but there is a lack of informations on the elastoelectric nonlinearity in this crystal.

II. THEORETICAL DISCUSSION

First we present a theoretical discussion which is necessary to describe the experimental method and results. We choose the Cartesian coordinate system $(x_\alpha, x_\beta, x_\gamma)$ so that x_α axis is perpendicular to the surface of the sample and x_β and x_γ are so oriented on the surface that the transformation of all tensor properties of the investigated sample from the basic crystallographic system to the system $(x_\alpha, x_\beta, x_\gamma)$ may be as simple as possible.

Then applying Christoffel's equation we calculated velocities and polarizations of the acoustic modes propagated in [1,0,0] direction in the BGO single crystal, which belongs to the 2 3 cubic group of symmetry. From that we know that in [1,0,0] in direction of BGO there exist one purely longitudinal and two equivalent purely shear modes of ultrasonic waves.

Since the BGO is piezoelectric the surface excitation can be used to generate ultrasonic modes. According to [6] the optimum direction of the external electric field $E_0^{(ex)}$ for the excitation of the u.w. (q_1, ω_1) is given by the direction cosines n_i , where

$$n_i = \sum_j \frac{k_j e_{ij\alpha}}{|e^{(ex)}|} \quad (1)$$

where j runs over α, β, γ and the components $e_i^{(ex)}$ are given as

$$e_i^{(ex)} = \sum_j k_j e_{ij\alpha}^* \quad (2)$$

Here the modified piezoelectric constants e_{ijk}^* are

$$e_{\alpha j \alpha}^* = e_{\alpha j \alpha} \frac{E_0}{E_0^s} \quad (3)$$

and

$$e_{ij\alpha}^* = e_{ij\alpha} - e_{\alpha j \alpha} \frac{e_{ij\alpha}^*}{e_{\alpha\alpha}^*}, \quad i \neq \alpha. \quad (4)$$

The meaning of the symbols is: e_{ij}^* - the components of the dielectric constants tensor at a constant strain, k_i - the components of the unit vector \vec{u}_0/u_0 , u_0 - the amplitude of the excited ultrasonic wave.

The amplitude of the excited ultrasonic wave is

$$u_0 = \frac{|e^{(ex)}|}{\rho v \omega} E_0^{(ex)} \quad (5)$$

with the following meaning of the symbols: ρ - mass density, v - velocity of the ultrasonic wave, ω - angular frequency, $E_0^{(ex)}$ - the amplitude of the external electric field.

Using equation (1), where x_α is in the [1,0,0] direction of the BGO single crystal, we get the following statements:

1. The longitudinal mode will not be excited.
2. The pure shear mode propagating in the [1,0,0] direction and polarized in [0,1,0] is excited by the external electric field oriented in the [0,0,1] direction.
3. Similarly, the shear mode propagating along [1,0,0] and polarized in [0,0,1] is excited by the electric field oriented in [0,1,0].

The ultrasonic wave propagating in the piezoelectric crystal in the direction given by the unit vector \vec{m} is accompanied by an electric field with components

$$E_i = \frac{m_\alpha e_{ijk} m_j}{m_\alpha \epsilon_{rs} m_s} S_{jk} \quad (6)$$

S_{jk} are components of the strain tensor of the ultrasonic wave.

According to relation (4) we conclude that only shear modes S_5 and S_6 are electrically active in the [1,0,0] direction of the BGO.

III. EXPERIMENTAL PROCEDURE AND RESULTS

The sample of BGO had the shape of a parallelepiped which edges $L_x = 2,99$ cm, $L_y = L_z = 1$ cm oriented along the crystallographic axis x_1, x_2, x_3 , respectively. We provided our sample with electrodes according to Fig.1. The electrodes 1, 1' serve for the application of the electric field along the axis x_2 to excite a shear ultrasonic wave polarized along the axis x_3 . The electrodes 2, 2' serve to generate the shear mode polarized along the axis x_2 . The electrodes 3, 3' and 4, 4' have the same function as the electrodes 1, 1' and 2, 2'. The electrodes 5, 5' serve to detect an electric field generated by mixing the ultrasonic waves. The experimental setup is shown in Fig.2.

The pulse generator generates, through the surface excitation, two ultrasonic pulses which travel in opposite directions. The ultrasonic pulses interact in the middle of the sample and generate an h.f. electric field which is received with electrodes 5, 5'. The received 2ω h.f. pulse has been amplified and then, after detection, displayed on the screen of the synchroscope. The amplitude of the pulse has been measured using the generator of gauge signals. Using this generator we also measured the amplitude of the pulses that generated the ultrasonic wave. The amplitude E_2 of the electric field 2ω is according to [5] equal to

$$E_2 = \sqrt{\frac{L_x L_y L_z}{\pi}} \frac{G \omega^4 u_0^2}{\epsilon_0 c_0^2 v^2} \quad (7)$$

We have used these denotations: L_x, L_y, L_z - dimensions of the electrodes 5, 5', L_x - their distance between them, c_0 - the velocity of light in vacuum, $G = \partial \epsilon_{66} / \partial E_2$ - the nonlinear coefficient.

In order to evaluate the nonlinear coefficient G we need the value of the amplitude u_0 of the ultrasonic wave in the interaction volume. This value has been obtained as follows:

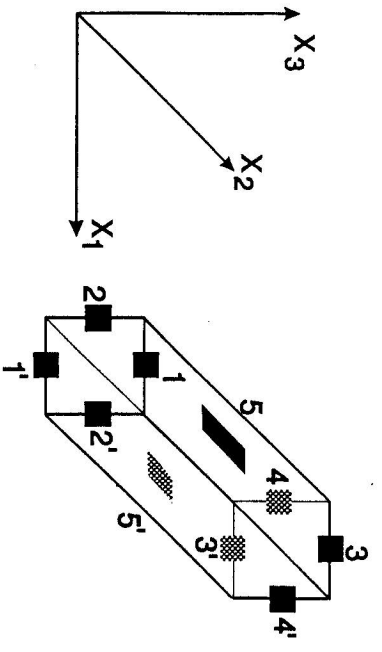


Fig. 1. Sample with electrodes.

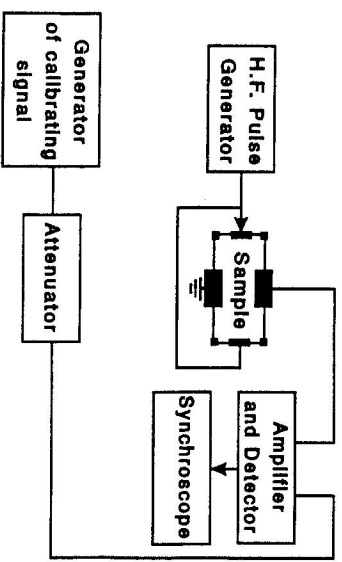


Fig. 2. Block diagram of the experimental arrangement.

1. We measured the amplitude of the exciting electric field E_0 and then according to relation (6) we calculated the amplitude of the ultrasonic wave at the face surfaces of the sample. To calculate this, we used values of material constants of the BGO obtained by Abrahams and Bernstein [12].
2. We measured the frequency dependence of the attenuation (Fig. 3), which shows the quadratic dependence of the attenuation coefficient, $\alpha = K \cdot f^2$, where $K = 1,358 \cdot 10^{-16} \text{ dB cm}^{-1} \text{ s}^2$. Then we measured velocity of the shear ultrasonic wave $[1,0,0]$, $[0,1,0]$. We have obtained $v = (1,75 \pm 0,01) \times 10^3 \text{ ms}^{-1}$, which is in very good agreement with the calculated value $v = 1,754 \cdot 10^3 \text{ ms}^{-1}$.
3. Using the results for the attenuation we calculated the amplitude of the ultrasonic wave in the middle of the sample.

4. Then we measured the amplitude of the 2ω electric field. Performing the procedures given above we have got all values necessary for the evaluation of the nonlinear coefficient $G = \partial C_{66} / \partial E_2$.

Our experiment has given the following data: $f = 19,6 \text{ MHz}$, $E_0 = (220 \pm 10) \cdot 10^{-4} \text{ Vm}^{-1}$, $E_2 = (4 \pm 1) \text{ Vm}^{-1}$, $G = (130 \pm 40) \text{ NV}^{-1} \text{ m}^{-1}$. The lower accuracy of the calculation results from inaccuracy in the measurement of very low amplitudes of the electric field E_2 .

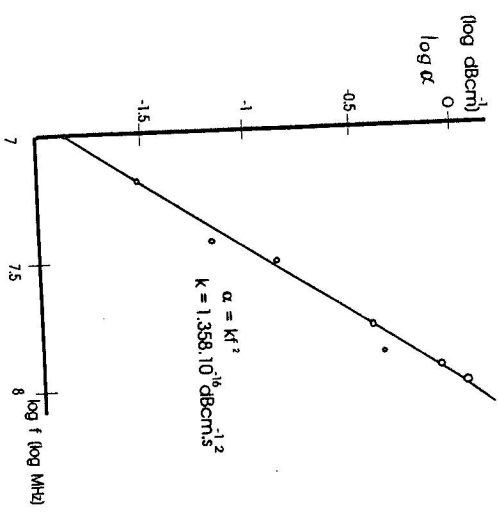


Fig. 3. Frequency dependence of the attenuation on shear u.w. in $[1,0,0]$ direction of BGO.

IV. CONCLUSIONS

1. The frequency dependence of the shear mode propagating along the $[1,0,0]$ direction of BGO crystal shows that $\alpha = k \cdot f^2$, where $k = 1,358 \cdot 10^{-16} \text{ dB cm}^{-1} \text{ s}^2$.
 2. The measured value of the velocity of the shear mode in the $[1,0,0]$ direction of BGO $v = (1,75 \pm 0,01) \cdot 10^3 \text{ ms}^{-1}$ is in a very good agreement with the calculated one. No significant dispersion was observed in the region $10 - 100 \text{ MHz}$.
 3. The value of the nonlinear coefficient $G = (130 \pm 40) \text{ NV}^{-1} \text{ m}^{-1}$, obtained for BGO is comparable with the values of the nonlinear coefficient obtained for LiNbO_3 by Thomson and Quate[1] who measured G in the range from 11 to $77 \text{ NV}^{-1} \text{ m}^{-1}$.
- From this point of view the BGO single crystal is also very suitable for fabrication of signal-processing acousto-electric devices.