Available online at www.sciencedirect.com
SCIENCE DIRECT

Journal of the European Ceramic Society 24 (2004) 1161–1164
www.elsevier.com/locate/jeurceramsoc

Possibility of crystalline axis determination in cubic BSO crystal by Brillouin scattering experiment

Y.S. Yu*, B.H. Min

Research Center for Electronic Ceramics, Research Institute of Basic Science, Dongeui University, Busan 614-714, South Korea

Abstract

The single crystal BSO of high quality was grown in air by Czochriaski method. We prepared one cubic shape sample of which the crystal axes were unknown. We have confirmed the directions of crystal axes and the elastic properties in the prepared BSO crystal by Brillouin scattering experiment. In the case of cubic symmetry crystals it is impossible to observe the conoscope image in the polarizing microscope. Determination of crystal axes by XRD in the bulk crystal is a very tedious work. So we propose the least squares fit method to determine the cubic crystal axes by Brillouin scattering experiment.

Keywords: BSO; Mechanical property; Non-destructive evaluation; Optical properties; Spectroscopy

1. Introduction

$Bi_{12}SiO_{20}$(BSO) has noncentro-symmetric sillenite structure and belongs to the space group I23.[1,2] This material is very attractive for practical applications due to its pronounced piezoelectric, electro-optic, elasto-optic, and photoconductive properties.[3–7] BSO is a piezoelectric semiconductor with a large band gap energy[3] and is one of the most sensitive photorefractive semiconductors with a fast response time.[8] Therefore BSO is a potential material for real-time optical information processing, optical computing, real-time interferometry[7] and image amplification.[9]

BSO crystal has the body center cubic structure and the corners of the unit cell are occupied by the silicon–oxygen tetrahedra. The unit cell contains two formula units of $Bi_{12}SiO_{20}$. Si atoms are positioned in the center and at the corners of the cubic unit cell. Four oxygen atoms are the nearest neighbours of the Si atom and form a tetrahedron, SiO_4. The Si–O bond length is 0.165 nm, and the lattice constant of BSO is 1.0104 nm.[10]

In this work, we have observed the acoustic phonons in the BSO crystal through the sandercock type (3+3) pass tandem Fabry–Perot interferometer. In the case of cubic symmetry crystals it is impossible to confirm the crystal axes through the polarizing microscope, because the conoscope image cannot be observed. Therefore, we applied the least squares fit method to the results of Brillouin scattering experiments to confirm both the directions of the crystal axes and the elastic constants in the cubic shaped BSO crystal.

2. Theory

The relations of wave vectors and angular frequencies among the incident light, the scattered light, and the acoustic phonon in Brillouin inelastic light scattering are as follows,

$$k_s = \vec{k}_i \pm \vec{q} \tag{1}$$

$$\omega_s = \omega_i \pm \Omega \tag{2}$$

where the subscripts i and s represent the incident and the scattered light, respectively. Ω is the Brillouin shift and $\vec{q}$ is a wave vector of the elastic wave. From Eq. (1) the wave vector of the elastic wave is represented by the following equation,[11]

$$q = \left|\vec{k}_s - \vec{k}_i\right| = \frac{2\pi}{\lambda}\left(n_i^2 + n_s^2 - 2n_i^2 n_s^2 \cos\theta\right)^{1/2} \tag{3}$$

where λ is the wavelength of the incident light in vacuum; n_i and n_s are the refractive indices of the

* Corresponding author.
E-mail address: ysyu@dongeui.ac.kr (Y.S. Yu).

0955-2219/$ - see front matter
doi:10.1016/S0955-2219(03)00589-2

incident and the scattered light in the medium, respectively; and θ is the scattering angle between the incident and the scattered light. In Brillouin scattering the scattered light from the thermally excited acoustic phonons shows a frequency shift with respect to the incident light. The frequency shift is proportional to the phase velocity of the acoustic wave which is usually in a hypersonic range. The phase velocity of the acoustic wave in the medium is given by the relation

$$v = \frac{\Omega}{q}. \tag{4}$$

An analysis of the elastic wave propagation in crystals begins with the equation of motion[12]

$$\rho \frac{\partial^2 u_i}{\partial t^2} = C_{ijkl} \frac{\partial^2 u_l}{\partial x_i \partial x_k} \tag{5}$$

where u is displacement and ρ is the density of the material. C_{ijkl} is the tensor components of the elastic constants and the summation convention is always valid.

3. Experimental

Single crystal BSO was grown from melt by the Czochralski method using a RF furnace in air. The density of BSO is 9.2 g/cm^3 and the refractive index is 2.615 at a wavelength of 514.5 nm.[4] The spectrum of the scattered light was analyzed by the use of a high contrast (3+3)-pass tandem Fabry–Perot interferometer. The light source was the 514.5 nm line of a single frequency Ar-ion laser. The incident power of 100 mW was focused into the sample by a lens of 50 mm focal length and the scattered light was collected by the same lens in a back-scattering geometry. The free spectral range was 50 GHz in our experiment and P-polarized light was incident by 70° with respect to the normal direction of the sample surface. The sample was cubic shape with dimensions 10×10×10 mm which are not parallel to the crystal axes. The series of Brillouin spectra are obtained by rotating the cubic sample from 0 to 360° with respect to the normal direction of the sample surface. A more detailed explanations of both the sample geometry and related formula can be also founded in the literature.[13,14]

4. Results and discussions

An arbitrary cubic sample was prepared to confirm the crystal axes and the elastic constants of the single crystal BSO. Brillouin scattering experiments and the least squares fit method were carried out. Fig. 1 shows the Brillouin spectrum of the single crystal BSO on c′-plane, where c′ means that the crystal axis c does not coincide with the edge of the cubic sample. In Fig. 1, the central peak is the Rayleigh line and the ghost peaks appear on both of the end parts which give the free spectral range (FSR) of 50 GHz. In general there are three acoustic components, as expected from the elastic continum, which are one longitudinal acoustic mode (L) and two transverse acoustic modes (T1, T2). In Fig. 1 the light is incident on the c′-plane with the incident angle 70° with respect to the c′-axis. Fig. 1, shows that all three of the acoustic components appear. Fig. 2(a) and (b) shows the variations of the Brillouin peaks which are measured from 0 to 360° at intervals of 10° with respect to the c′ and a′ axes, respectively.

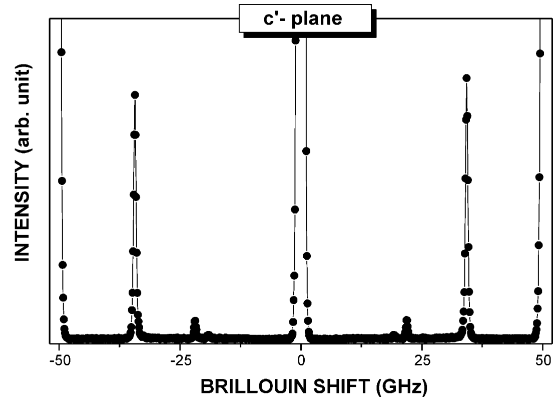


Fig. 1. Brillouin spectra of BSO about c′-plane.

If the prepared cubic sample is cut along the crystal axes, the variations of the Brillouin peaks in Fig. 2(a) and (b) will show a fourfold symmetry with a period of 90°. However, Fig. 2(a) and (b) do not coincide with each other, and neither does the periodicity, so the edges of the prepared cubic sample are not parallel with the crystal axes. Therefore, it is possible to find the crystal axes and the elastic constants from the data in Fig. 2(a) and (b) by using the rotational transformation and Christoffel equation.[11]

$$|\Gamma'_{il} - \rho v'^2 \delta_{il}| = 0 \tag{6}$$

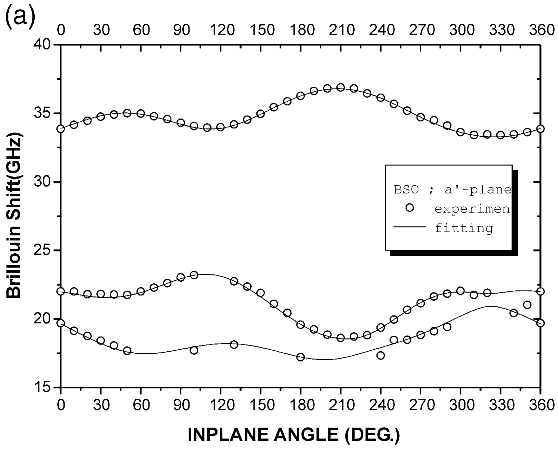
where

$$\Gamma'_{il} = c'_{ijkl} Q'_j Q'_k \tag{7}$$

and

$$c'_{ijkl} = R_{im} R_{jn} R_{kp} R_{lq} c_{mnpq}. \tag{8}$$

In the above equations prime represents rotation and R is the rotational transformation matrix. According to the transformation theory and the Christoffel equation we construct the least squares fit program to obtain the elastic constants and the crystal axes of the sample. In Fig. 2(a) and (b) the circles are the measured values and the lines are the results of the fit. As you can see, the expermental values and the values fitted by the theory are almost the same as each other. Therefore, it is possible to find the crystal axes and the elastic constants



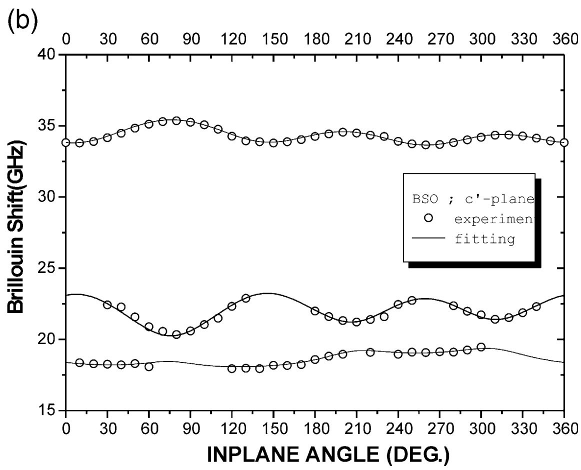


Fig. 2. Brillouin shifts of the acoustic phonons according to the propagation direction about (a) a′-plane; and (b) c′-plane.

Table 1
Elastic constants of BSO crystal obtained from the least squares fit (unit: $10^{10}N/m^2$)

c_{11}	c_{12}	c_{44}
13.43	3.06	2.64

of the sample from the least squares fit. The results of the obtained elastic constants are shown in Table 1. The elastic constants obtained in our work and the measured data by the ultrasonic technique are similar. The ultrasonic method is applied in BSO crystals to obtain the elastic constants by Stepanov et al.[4] From the view point of the crystal axes determination, Brillouin scattering is a very simple and convenient method to confirm the crystal axes of the cubic symmetry bulk crystal compared with XRD method. The obtained Euler angles are $\alpha = 39.58$, $\beta = 123.62$, and $\gamma = 69.38°$ in our sample.

From the elastic constants obtained it is possible to construct the slowness curves of the acoustic phonons in the BSO crystal. Fig. 3 shows the slowness curves of the elastic waves of the single crystal BSO in the cube

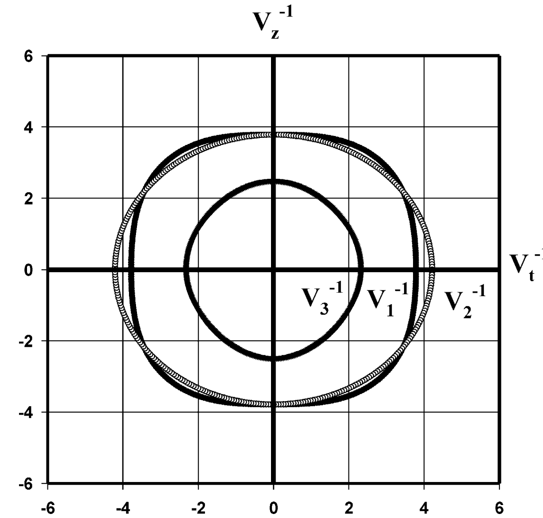


Fig. 3. Slowness curves of the acoustic phonons in the cube diagonal face of BSO. (unit: 10^{-4}s/m).

diagonal face. As you can see, in Fig. 3 the slowness curves have a twofold symmetry and show degenerate transverse modes in the directions of the crystal axes. In this plane there are six directions of degenerate transverse modes. The anisotropy factor for a cubic symmetry crystal is defined as follows

$$A = \frac{2c_{44}}{c_{11} - c_{12}} \tag{9}$$

and the inverse of the longitudinal wave velocity can be written in terms of the anisotropy factor as

$$\frac{1}{V_L} = \left[\rho / \left\{C_{11} - C_{44}(1 - 1/A)\right\}\right]^{1/2}. \tag{10}$$

In the case of our sample the anisotropy factor is 0.509, that is, this value is less than unity. Therefore as you can see in Fig. 3 the quasishear slowness curve bulges in and the quasilongitudinal slowness curve bulges out.

5. Conclusions

BSO single crystals were grown by the Czochralski method in air. We prepared one cubic shape sample with unknown crystal axes in the BSO crystal. Brillouin scattering experiments performed on two faces of the prepared sample and the Brillouin shift data obtained were processed by the least squares fit program. As a result it was possible to confirm both the crystal axes and the elastic constants of the cubic BSO crystal. It is usually very time consuming to determine the crystal axes in the bulk crystal by XRD, so in this paper we propose a method using the least squares fit from Brillouin scattering experiments. The accuracy of the axes determination in our study is comparable to values obtained by XRD and the optical interference method. Our results for the elastic constants are very similar to the results of Stepanov's[4] using the ultrasonic method. From the elastic constants obtained the slowness and velocity curves can be constructed, and the anisotropy factor is determined to be 0.509.

Acknowledgements

The authors would like to express their gratitude for financial support from the Research Center for Electronic Ceramics (RCEC) of Dongeui University founded by Korea Science and Engineering Foundation (KOSEF), Ministry of Science and Technology (MOST) and Busan Metropolitan City Government.

References

1. Xuewu, Xu., Jingying, Liao., Bingfu, Shen., Peifa, Shao., Xiangiu, Chen and Chongfan, He., $Bi_{12}SiO_{20}$ single crystal growth by the Bridgman method. *J. Crystal Growth*, 1993, **133**, 267–272.
2. Xuewu, Xu. and Jingying, Liao., Polar habits of the $Bi_{12}Sio_{20}$ crystals grown by the Czochralski method. *J. Crystal Growth*, 1993, **134**, 337–431.
3. Aldrich, R. E., Hou, S. L. and Harvill, M. L., Electrical and optical properties of $Bi_{12}SiO_{20}$. *J. Appl. Phys.*, 1971, **42**, 493–494.
4. Stepanov, S. I., Shandarov, S. M. and Khat'kov, N. D., Photoelastic contribution to the photorefractive effect in cubic crystals. *Sov. Phys. Solid State*, 1987, **29**, 1754–1756.
5. Peltier, M. and Micheron, F., Volume hologram recording and charge transfer process in $Bi_{12}SiO_{20}$ and $Bi_{12}GeO_{20}$. *J. Appl. Phys.*, 1977, **48**, 3683–3690.
6. Grousson, R., Henry, M. and Mallick, S., Transport properties of photoelectrons in $Bi_{12}SiO_{20}$. *J. Appl. Phys.*, 1984, **56**, 224–229.
7. Huignard, J. P., Herriau, J. P. and Valentin, T., Time average holographic interferometry with photoconductive electrooptic $Bi_{12}SiO_{20}$ crystals. *Applied Optics*, 1977, **16**, 2796–2798.
8. Huignand, J. P. and Micheron, F., High sensitivity read-write volume holographic storage in $Bi_{12}SiO_{20}$ and $Bi_{12}GeO_{20}$ crystals. *Appl. Phys. Lett.*, 1976, **29**, 591–593.
9. Lipson, S. G. and Nisenson, P., Imaging characteristics of the itek PROM. *Applied Optics*, 1974, **13**, 2052–2060.
10. Wojdowski, W., Vibrational modes in $Bi_{12}GeO_{20}$ and $Bi_{12}Sio_{20}$ crystals. *Phys. Stat. Sol. (b)*, 1985, **130**, 121–130.
11. Auld, B. A., *Acoustic Fields and Waves in Solids I*. John Wiley and Sons, New York, 1973.
12. Vacher, R. and Boyer, L., Brillouin scattering: a tool for the measurement of elastic and photoelastic constants. *Phys. Rev. B*, 1972, **6**, 639–673.
13. Yu, Y. S. et al., A study of elastic properties of Cr-doped $KTiOPO_4$ single crystals by Brillouin scattering. *J. Korean Phys. Soc.*, 1998, **32**, S518–S520.
14. Jung, H. R. et al., Elastic properties of $Li_2B_4O_7$ single crystal by means of Brillouin scattering. *J.Korean Phys. Soc.*, 1998, **32**, S521–S523.