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Piezoelectric stiffening in the thickness direction of c-plane ZnO single crystal measured by Brillouin scattering

Brillouin 散乱法を用いた c 面 ZnO 単結晶板における音響電気効果による音速変化測定

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1. Introduction

In the applications of thickness mode FBARs, the electromechanical coupling coefficient k_{33} is important property. The electromechanical coupling coefficient can be estimated from piezoelectric stiffened and unstiffened acoustic wave velocities [1]. However, in semiconductive materials which have the low-resistivity, this stiffening effect occurs in the high-frequency (GHz) range. It is then difficult to observe this stiffening by conventional ultrasonic methods in the MHz range.

We have reported the piezoelectric stiffening in m- or a-plane semiconductive materials (e.g. ZnO and GaN single crystals) by Brillouin scattering (BRS), which can measure the acoustic wave velocity in GHz range. However, piezoelectric stiffening in c-plane semiconductive materials has not been measured yet because BRS needs the refractive index to determine the acoustic wave velocity in the thickness direction. For this problem, we have recently reported a BRS measurement technique to estimate the refractive index and the acoustic wave velocity in the thickness direction at once [2].

In this study, we investigated the acoustic wave velocity change due to piezoelectric stiffening in the thickness direction of c-plane ZnO single crystals by BRS.

2. Measurement of piezoelectric stiffening

Piezoelectric stiffening depends on the frequency and the sample resistivity which changes due to the temperature. This frequency-temperature characteristics of piezoelectricity including the effect of carrier diffusion can be estimated by Hutson and White's equation [3]. **Fig.1** shows theoretically estimated resistivity and frequency dependence of the longitudinal wave velocity change in a ZnO single crystal due to the piezoelectric stiffening. In this estimation, Smith's constants [4] were used for c^E_{33}, ε^S_{33} and e_{33}. Carrier diffusion was estimated for the case in which carrier mobility was constant at 100 cm^2/V. From this figure, it is clear that the piezoelectric stiffening in a ZnO single crystal whose resistivity is less than 1 Ωm is observed in GHz range. In addition, the velocity change becomes unclear by the effect of carrier diffusion if the frequency and the mobility become higher. The electromechanical coupling coefficient k_{33} is obtained from the following equation:

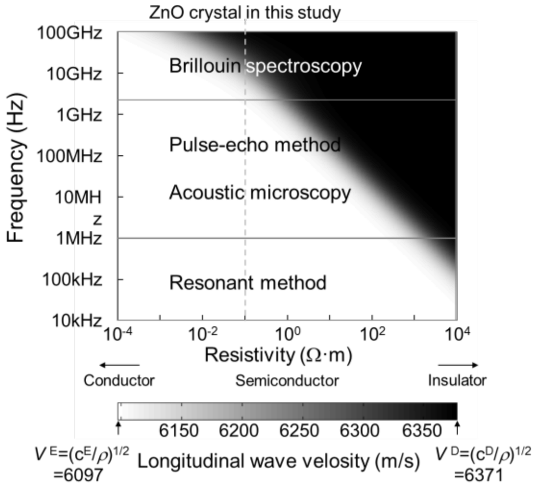

Fig. 1 Theoretically estimated longitudinal wave velocities propagating along c-axis in a ZnO crystal as functions of resistivity and frequency. Hutson and White's equation was used [1,3,4]. The carrier diffusion was estimated by assuming that carrier mobility was constant (100 cm^2/V) and temperature was 27ºC.

$$k_{33}^2 = 1 - \left(\frac{V^E}{V^D} \right)^2 \tag{1}$$

Here, V^E is the unstiffened acoustic wave velocity and V^D is the stiffened acoustic wave velocity.

3. Measurement sample

Four c-plane ZnO single crystals (Zeniya Industrial Inc., sample size: 10×10×0.5 mm^3) were prepared for this study. Sample temperature was controlled from -190 to 50ºC on a heat stage (LK-600PM, Linkam) to change the sample resistivity. To prevent the frost on the sample at low-temperatures, the samples were set in the vacuum chamber during the measurement and laser beam was irradiated through a silica glass window. The resistivity of the samples was measured by using liquid Gallium alloy as an electrode. From this measurement, the resistivity change of sample A was 0.007-0.03 Ωm, Sample B was 0.05-0.4 Ωm, Sample C was 0.1-0.4 Ωm, and Sample D was 0.3-10 Ωm in the range of -190-50ºC.

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4. Measurement system

BRS measurements were performed by a tandem Fabry-Pérot interferometer (JRS Instruments) using a solid laser (mpc3000, Laser Quantum, 532 nm). The actual diameter of the focused laser beam in the sample was approximately 50 μm. The laser power near the sample was 240 mW.

In this measurement, 180º and 90R scattering geometries shown in **Fig.2** were used to investigate the refractive index and the longitudinal wave velocity in the thickness direction of the samples. In these scattering geometries, the acoustic wave velocity are obtained using the following equations:

$$v^{180} = f^{180}\frac{\lambda_i}{2n} \quad (2)$$

$$v_L^{90R} = f_L^{90R}\frac{\lambda_i}{2\left[n^2 - \left\{\sin\left(\frac{\Theta'}{2}\right)\right\}^2\right]^{\frac{1}{2}}} \quad (3)$$

Here, λ_i is the wavelength of the incident light, n is refractive index, $\Theta'/2$ is scattering angle, f^{180} is the shift frequency of 180º Brillouin scattering peak owing to the longitudinal wave and f_L^{90R} is the shift frequency of 90R Brillouin scattering peaks owing to the longitudinal wave.

In this measurement, the refractive index is treated as the ordinary refractive index $n_\perp$ because the crystalline axis of c-plane ZnO single crystal is perpendicular to the polarization of the incident light. Therefore, the ordinary refractive index $n_\perp$ is obtained from the following equation:

$$n_\perp = \frac{\sin(\Theta'/2)}{\sqrt{1-\left(f_L^{90R}/f^{180}\right)^2}} \quad (4)$$

However, the ordinary refractive index measured by this technique has large uncertainty. Therefore, the value was estimated by linear fitting of the temperature characteristics of the indices.

From this ordinary refractive index and equation (2), the longitudinal wave velocity was estimated.

5. Results and discussion

Figure 3 shows the longitudinal wave velocity of c-plane ZnO single crystal measured in the range of 0.007-10 Ωm. A linear velocity changes due to heating was subtracted from experimental data by using temperature coefficient of $\Delta c_{33}/\Delta T$= 1.23×10^{-2} GPa/ºC [5]. In addition, the theoretical curve calculated by Spector's equation [6] with the effect of carrier diffusion was also plotted. From the experimental data, the longitudinal wave velocity increased as the resistivity increased showing an empirical curve. However, in comparision with the theoritecal curve, the measured curve showed a characteristic shift. This difference seems to result from the properties used in the theoretical estimation, especially dielectric property.

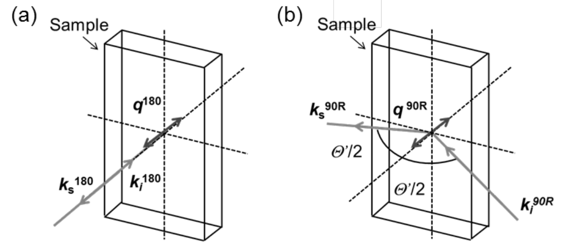

Fig. 2 (a) 180º scattering geometry and (b) 90R scattering geometry: Wave vector of incident light. k_s^{180}: Wave vector of scattered light. q^{180}: Wave vector of phonon. k_i^{90R}: Wave vector of incident light. k_s^{90R}: Wave vector of scattered light. q^{90R}: Wave vector of phonon. Θ': Scattering angle which is defined by the incident light and the scattered light.

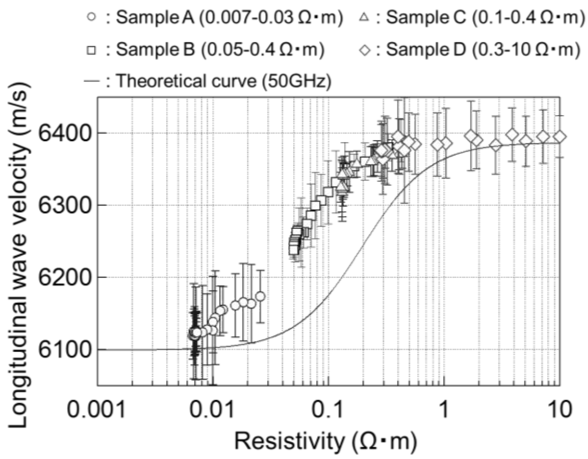

Fig. 3 Longitudinal wave velocity of the ZnO single crystal measured in the range of 0.007-10 Ωm. The resistivity of the crystals changed by heating. The change of the elastic constant c_{33} due to heating was excluded [5]. Also described is the theoretical curve at 50 GHz, calculated by Spector's equation [6].

6. Conclusion

We have succeeded in the observation of the longitudinal wave velocity change due to the piezoelectric stiffening in c-plane ZnO single crystales. However, we could not determine the electromechanical coupling coefficient k_{33} because the resistivity range of velocity dispersion becomes wider in the low-resistive materials.

This result suggests that the electromechanical coupling coefficient k_{33} can be estimated by using the combination of 180º and 90R scattering geometries if the sample resistivity changes widely.

References

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