Elastic Properties of Lithium Germanate Glasses Studied by Brillouin Scattering

ブリルアン散乱法によるリチウムゲルマン酸塩ガラスの弾性 的性質

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1. Introduction

The relation between microscopic structure and physical properties of amorphous or glassy materials is the current topics in materials science. Lithium germanate glasses are utilized as the optical fibers and infrared transmitting glasses. Some physical properties of lithium germanate glasses, such as density and refractive index, are known to pass through maxima or minima with increasing Li₂O content. Such behaviors are called the "germanate anomaly", which is related to the conversion of germanium ions from four-fold to six-fold coordination and the formation of non-bridging oxygen [1,2].

To investigate the relationship between the structural changes and the elastic properties, we measured the elastic properties such as sound velocity and elastic moduli. The elastic properties are very sensitive to the structural changes. Consequently, they also show unique behaviors against their compositions.

In this experiment the only thin and fragile samples are obtained by using the plate quenching method. Only Brillouin spectroscopy enables us to investigate the elastic properties of such samples by using a finely focused laser beam without any contact with a sample and breaking a sample [3].

2. Experimental

Lithium germanate glasses, $xLi_2O \cdot (100-x)GeO_2$ represented as a function of Li_2O mole fraction, containing 6-36 mol% Li_2O were prepared to investigate the structural change originated from their compositions. The starting materials were powdered GeO_2 and $LiOH \cdot H_2O$. They were first made to react in an aqueous solution and dry in dryer for a week and heated at approximately $1050^{\circ}C$ in the electric furnace [4]. Germanate glasses have a strong tendency to crystallize; therefore, the glass samples were made by using the plate quenching technique with rapid cooling.

The Brillouin spectra were measured by a Sandercock-type 3+3 pass tandem multipass Fabry-Perot interferometer (FPI) combined with a optical microscope. In order to measure not only a

transverse acoustic (TA) mode but also a longitudinal acoustic (LA) mode, the Brillouin spectra were measured at the right-angle (90°A) scattering geometry [3].

3. Results and Discussion

Figure 1 shows the observed Brillouin spectra of lithium gernanate glasses (x=6, 18, 30) containing LA and TA modes. The longitudinal (V_L) and transverse (V_T) sound velocities are given by

$$V_{i} = \frac{\Delta v^{i}_{90^{o} A} \lambda}{\sqrt{2}} \quad (i = L, T) , \qquad (1)$$

where λ is the wavelength of the incident laser beam (532nm) and $\Delta V_{90^{\circ}A}$ is the Brillouin shift. The 90°A-scattering geometry is an effective method, since it needs no refractive index in determination of the sound velocities [3]. Figure 2 shows the composition dependence of the V_L and V_T at room temperature. The values of V_L and V_T gradually increase with increasing Li₂O content up to about 22 mol% and then keep nearly constant.

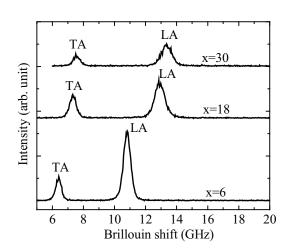


Fig.1 Brillouin spectra of lithium germanate glasses, xLi₂O·(100-x)GeO₂

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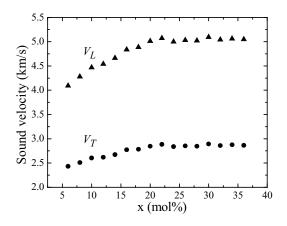


Fig.2 Composition dependences of the sound velocity V_L and V_T .

The elastic constants such as longitudinal modulus (L), shear modulus (G), bulk modulus (K), Young's modulus (E) and Poisson's ratio (σ) are given by

$$L = \rho V_L^2, \quad G = \rho V_T^2, \quad K = L - \frac{4}{3}G,$$

$$E = \frac{G(3L - 4G)}{L - G}, \quad \sigma = \frac{L - 2G}{2(L - G)},$$
(2)

where ρ is the density of the sample [3]. The ρ values are gained from the literature reported by Shelby et al, [2]. Figure 3 shows the composition dependence of L, G, K, and E. All these elastic constants show similar behaviors and increase gradually with increasing Li₂O content. They show maxima at about 22 mol% and then decrease with increasing Li₂O content. It is thought that these behaviors are due to the result of the conversion of germanium ions from four-fold to six-fold coordination and the formation of non-bridging oxygen that destroys the glass network [1,2,5]. The composition dependence of Poisson's ratio is shown in Figure 4 also shows a unique behavior like V_L and V_T . Generally Poisson's ratio is more sensitive to the structure units forming the germanate network than other elastic moduli such as L, G, K, and E [3]. The glass structure has been investigated by Raman scattering. The relationship between Poisson's ratio and the glass structure has been discussed.

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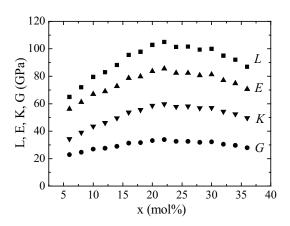


Fig.3 Composition dependences of the longitudinal modulus (L), Young's modulus (E), bulk modulus (K), shear modulus (G).

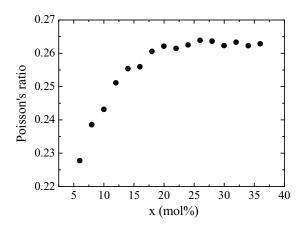


Fig.4 Composition dependence of Poisson's ratio.