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Optical characteristics of nanocrystallized glass fiber with second-order optical nonlinearity

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Second harmonic generation (SHG) and propagation loss have been measured on nanocrystallized optical fibers of tellurite-based glass, $15K_2O \cdot 15Nb_2O_5 \cdot 70TeO_2$. The result of the angular dependence of the SHG intensity is discussed based on the difference in the transmission factor varying with the polarization direction. Propagation-loss measurements on the fiber samples by a cut-back method showed low loss compared with that of a typical $LiNbO_3$ waveguide.

Key-words : Optical fiber, Glass ceramics, Tellurite

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1. Introduction

Fiber-type devices with active functions for optical signal processing, such as switching, modulation, and routing, are some of the most promising components in the new era of fiber network systems. Optical fibers exhibiting electro-optic or nonlinear effects are attractive candidates because these effects allow realization of the above-mentioned active functions. Although considerable efforts to fabricate fibers with a crystalline phase, i.e., single-crystal fibers using optically nonlinear crystal materials ($LiNbO_3$, etc.),[1),2)] have been made for developing active fiber-type components, it is difficult to form fibers that maintain the ordered structure of the crystalline phase in a glass matrix. Although a tellurite glass fiber containing small crystals of nonlinear $KNbO_3$ was fabricated for the same purpose,[3)] the homogeneity of the refractive index in the core section was low due to segregation of the Te component, which caused light scattering.

Crystallized glass having nanoscale nonlinear optical crystallites is one of the best materials for fiber-type functional devices because it shows high transparency in addition to typical advantages of conventional crystallized glasses, such as high formability and permanent nonlinearity resulting from the crystallites. Nanocrystallization with optical nonlinearity has been reported in several glass systems. We have selected a tellurite-based glass composition, $15K_2O \cdot 15Nb_2O_5 \cdot 70TeO_2$, because it shows advantageous optical properties such as a high refractive index and infrared transmissivity. Furthermore, crystallized glass fabricated using this composition shows nanocrystallization with a high level of transparency and second harmonic generation (SHG).[4),5)] In a previous report, the fabrication of nanocrystallized glass fiber was achieved using the same tellurite composition as reported here by a pertinent thermal treatment, and nanoscale crystals with an average diameter of 50 nm and random orientations were observed in the obtained crystallized glass fibers.[6)]

In this paper, we discuss optical nonlinearity and propagation loss in the fabricated crystallized glass fibers. In the tellurite-based crystallized glass, SHG has been experimentally confirmed in a bulk sample.[7)] However, the mechanism of SHG from this crystallized glass has not been clarified, and therefore, the optical nonlinearity of crystallized glass fibers is unclear. One of the objectives of this study is to clarify the optical nonlinearity in the obtained crystallized glass fibers. A new experimental setup was built for this purpose because existing measurement methods, such as the Maker fringe, does not apply for fiber samples. Propagation-loss measurement was performed to show the practical level of propagation loss in crystallized glass fibers. The high transparency of crystallized glass was discussed in a previous report,[4)] but quantitative estimation has not been attempted. In this study, quantitative estimation of propagation loss was performed by the cut-buck method, and the loss was compared with a typical value of an optical waveguide fabricated from $LiNbO_3$ for the same purpose.

2. Experimental procedure

2.1 Fiber fabrication and nanocrystallization in glass fibers

Crystallized glass fiber samples were prepared by fiber drawing from a preform rod fabricated from the glass composition, $15K_2O \cdot 15Nb_2O_5 \cdot 70TeO_2$ (in mol%), and following the crystallization process using the two-step heating technique. In the fiber drawing process, the glass fibers were obtained with diameters in the range from 230 μm to 320 μm, 80% of which were in the range from 260 μm to 280 μm. Fibers in the diameter range between 270 μm and 280 μm were selected for use in the next process to reduce the effect of diameter fluctuation along the length. Then, the obtained glass fibers were cut into 30 mm segments for ease of post-heating treatment and measurements. In the crystallization process, fiber samples were treated in a muffle furnace using boron nitride powder. The sample fibers were imbedded in the boron nitride powder. The two-step heating conditions (the first step at 375°C for 5 h and the second step at 425°C for 1 h) were applied for the heat treatment.

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2.2 Second-harmonic generation measurement

The SHG intensity from the crystallized glass fibers was measured using an experimental setup comprising a Q-switch Nd–YAG laser (1064 nm), as shown in **Fig. 1**. Note that in this geometry, the excitation and the output beams are at a right angle with the fiber samples. This method was employed to use a short light path in the fiber samples and induce rotation-angle dependence of the SHG intensity from the fiber samples. Observation of a significant SHG intensity from the end face of the fiber samples is difficult because phase matching methods for optical fibers containing nano-sized nonlinear optical crystals have not been established yet. In this measurement, the significance of SHG intensity is shown by comparing the rotation angle dependence of the measured SHG intensity resulting from the curved surface of fiber samples with the calculated values. The crystallized fiber samples were placed in a row in parallel on a slide glass, and a polarized beam from an Nd–YAG laser was introduced to the side faces of the fibers. The induced SHG light was detected by a photomultiplier through an IR filter. The sample fibers were rotated against the beam direction to measure the variation in the induced SHG intensity (see Fig. 1).

2.3 Propagation-loss measurement

Propagation loss was measured by a cut-back method. **Figure 2** shows the measurement system. Fiber samples with lengths of 5, 10, and 15 mm were selected as length to obtain enough measurement accuracy. Each data point of loss is an average value on 10 fiber samples cut from a fiber drawn from the same preform; the sampling position was randomly selected. After coating the fiber samples with a low refractive-index polymer to provide a clad structure, we polished both the endfaces optically. A laser beam from a semiconductor laser with 1550 nm wavelength and 1 mW output power was introduced into the measurement sample through a single-mode optical fiber (core diameter 9 μm) connected directly. An aperture with a 300 μm hole was placed between the sample and detector to remove scattered light.

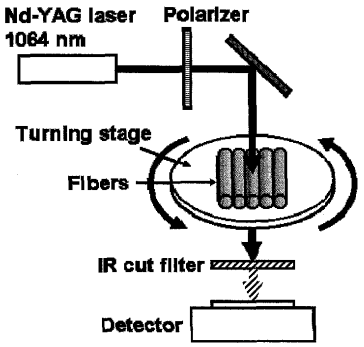

Fig. 1. Experimental setup for SHG measurement on fiber samples.

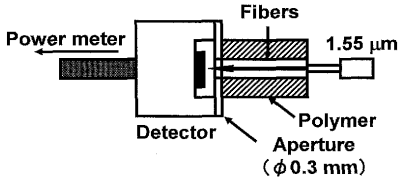

Fig. 2. Experimental setup for propagation-loss measurement by the cut-back method.

3. Results and discussion

3.1 SHG measurement

Figure 3 shows the result of SHG measurements on nanocrystallized glass fiber samples compared with that on y-cut quartz. The result shows that the average value of the SHG intensity observed from the fiber sample was about 1/400 of the maximum intensity on y-cut quartz. In this measurement, higher-order harmonic generation by the involvement of multiple crystals is not considered because the obtained SHG intensity was comparatively small. The significance of this result can be explained by a discussion on rotation-angle dependence of the SHG intensity obtained by this measurement.

The rotation angle dependence of the SHG intensity on the fiber samples can be described only by the variation in the transmission factor of the incident laser beam to the measurement sample. The polarization direction of the crystals has to be discussed with the transmission factor in the case of single crystal fibers or crystallized glass fibers with oriented crystallites, but such a discussion is unnecessary at this time because the sample fibers have nano-sized and randomly oriented crystals. The transmission factors of the p and s polarizations are changed by the incident angle ϕ due to the curved surface of the fiber samples. The variation in the transmission factors from the center to the edge of a sample fiber was calculated as power transmission factors using Fresnel's formulas. In this calculation, 1.00 and 2.11 were used as refractive indices of air and crystallized glass, respectively. **Figure 4**(a) shows the results of the calculation of the relation between incident angle ϕ_1 and transmission factors for both polarizations. The transmission factors for p polarization T_p and s polarization T_s show different variations with the average values calculated as 0.913 and 0.780, respectively. The variation in the total transmission factor with θ, the rotation angle of the sample, arises from the difference in the transmission factor between p and s polarizations. Figure 4(b) shows the variation in the transmission factor for each polarization together with the total value. Since the transmission factors for the p and s polarizations have a period of π, $\pi/2$ out of phase with each other and the amplitudes are also different, the total transmission factor varies with a period of $\pi/2$. The variation in the total transmission factor by the rotation of fiber samples directly affects the SHG intensity. The variation in the SHG intensity was calculated, as indicated by a broken line in Fig. 4(b), where the SHG intensity should increase in proportion to the square of the incident beam intensity. The amplitude of the variation fits with the experimental results, and the good agreement between theory

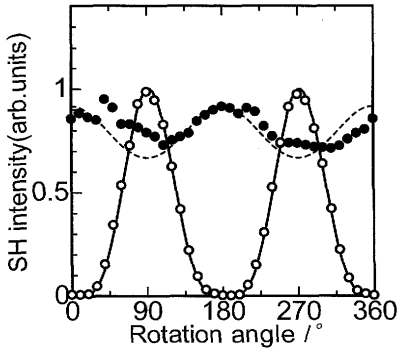

Fig. 3. Result of SHG measurement. Open circles and closed circles designate data on y-cut quartz and crystallized fiber, respectively.

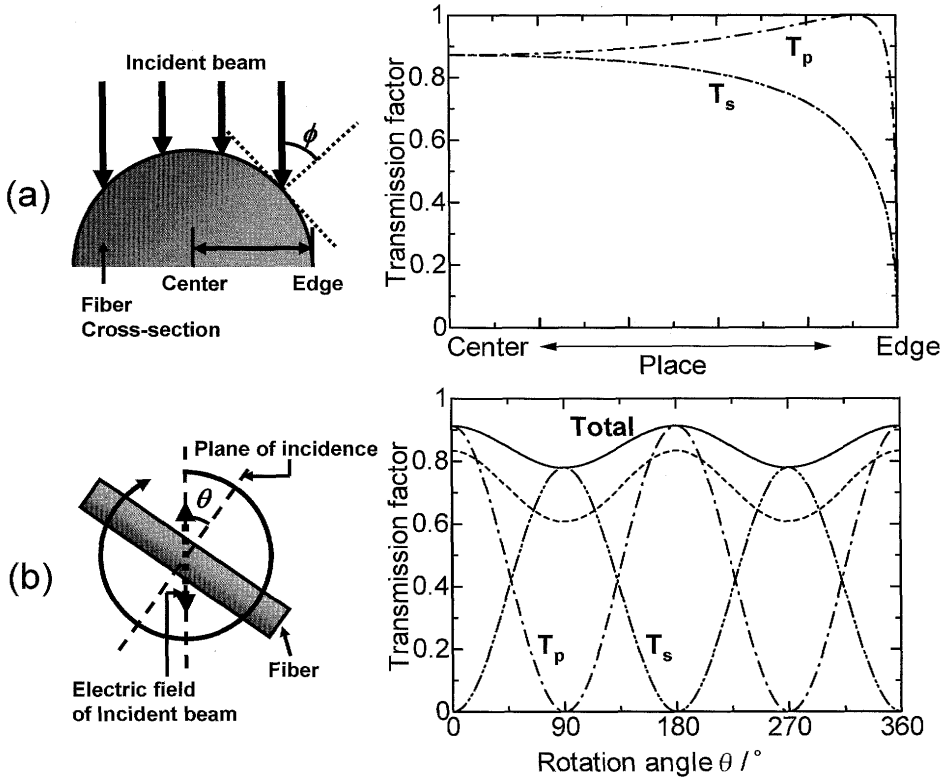

Fig. 4. (a) Variation in the transmission factor with an irradiated region. (b) Variation in the transmission factors with the rotation angle. T_p and T_s are transmission factors for *p*-polarization and *s*-polarization, respectively. The solid black line indicates the sum of T_s and T_p, and the black broken line indicates the square of the sum.

and experimental results confirms that the SHG was induced in the sample fibers.

Since the experimental value matched the calculated value, it was clarified that the nanocrystallized glass fibers have optical nonlinearity. However, measurement of the actual value of nonlinear optical constants on the fiber sample is currently difficult. Thus, improvement of the measurement method is necessary for this purpose.

3.2 Propagation loss measurement

The result of the propagation-loss measurement is shown in **Fig. 5**(a). The broken lines are least-mean-square fittings, and their slopes indicate the propagation loss per unit length. The propagation-loss values on the untreated glass fibers and crystallized glass fibers are 0.02 dB/cm and 0.15 dB/cm, respectively. The initial objective of this study was to obtain a value comparable to that of a light waveguide fabricated using $LiNbO_3$, and the obtained loss value is sufficiently low such that an actual device application can be considered. The significance of this result is a propagation-loss level that has been achieved on the thickly crystallized glass fiber with crystal particles occupying most of its volume. A lower propagation loss of about 100 dB/km (0.01 dB/cm) has been reported on nanocrystallized glass fiber for a laser host material based on another glass composition, $30SiO_2 \cdot 15AlO_{3/2} \cdot 29CdF_2 \cdot 17PbF_2 \cdot 4YF_3$.[8] The remarkably low propagation loss reported in Ref. 8 is a result of the low volume density of about 1–10 vol% of the crystallites. In the crystallized fiber sample obtained in this study, it was supposed that the volume fraction was more than 90% from the transmission electron microscopy (TEM) image, as shown in **Fig. 6**.

To further reduce propagation loss in the crystallized glass

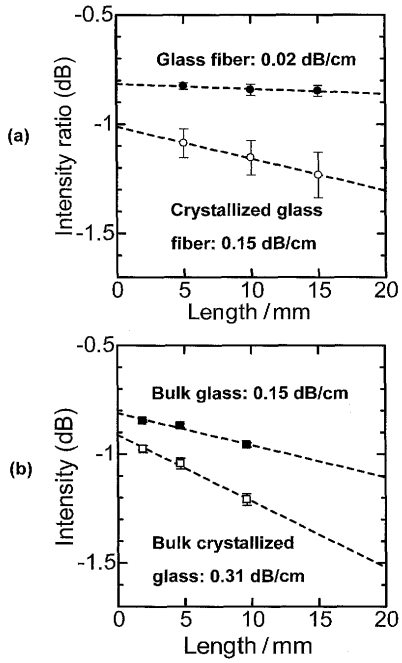

Fig. 5. Results of propagation-loss measurement: (a) fiber sample and (b) bulk sample.

fibers, it is important to understand the cause of propagation loss. We discuss here about what has an influence on the propagation loss of crystallized glass fibers. In the fiber samples, the increment of propagation loss after crystallization was 0.13 dB/cm.

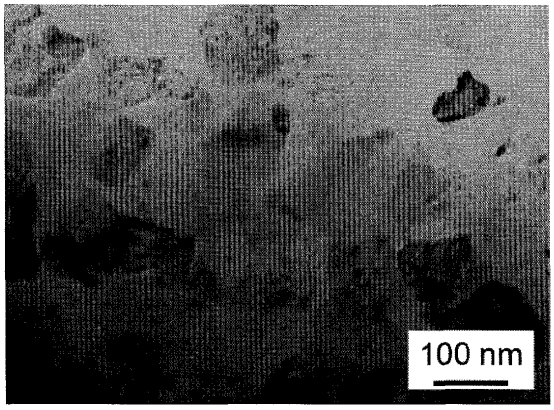

Fig. 6. Transmission electron microscopy (TEM) image of crystallized glass fiber sample.

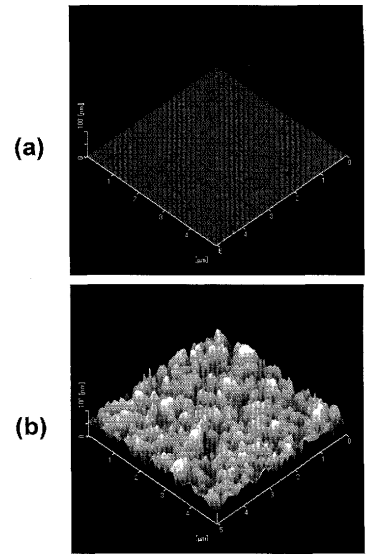

Fig. 7. AFM images of fiber samples: (a) glass fiber and (b) crystallized glass fiber.

The following are believed to be the causes of this increment: crystal particles, roughness of the core–clad boundary, and the difference in the refractive index between the core and clad regions.

The crystal particle is the cause of the inner region of crystallized glass fibers, and the roughness and refractive-index difference are the causes of the core–clad boundary. The contribution to propagation loss of the inner region was estimated by propagation-loss measurement on a bulk sample that has no core–clad boundary. The bulk sample was treated for crystallization under the same conditions as fiber samples. Figure 5(b) shows the result of propagation-loss measurement. The propagation losses of glass and crystallized bulk samples were 0.15 dB/cm and 0.31 dB/cm, respectively, and they showed that the contribution of crystal particles for propagation loss is 0.16 d/cm.

The existence of the two causes of propagation loss on the core–clad boundary (roughness and refractive-index difference) was examined by atomic force microscope (AFM) measurement and refractive index measurement using a wavelength of 1.55 μm. **Figure 7** shows the AFM images of the surface of glass and crystallized fiber samples. It clearly shows an increase in the surface roughness after crystallization. In addition, refractive index measurement on glass, crystallized glass, and the clad region (polymer) shows values of 1.98, 2.07, and 1.45, respectively, and it was seen that the refractive index difference between the core and clad regions increases by 0.09 after crystallization. The estimation of the contribution of each cause for propagation loss is difficult. However, it is believed that the sum of these causes, the total contribution of the boundary region, can be estimated from the difference between the increment of the propagation loss on the fiber sample by crystallization and contribution of an intrinsic cause. By employing this method, the total contribution of the boundary region was estimated to be –0.03 dB/cm. This means that the boundary region function decreases the propagation loss by crystallization. This result suggests that the increase in the refractive-index difference between the core and clad regions decreases the radiant light to the clad region.

The above result showed that the contribution of the inner region (crystal particle) strongly influences the propagation loss of fiber samples after crystallization. Therefore, the decrease in light scattering by crystal particles is effective in decreasing propagation loss. It is expected that the longer nucleation process in the two-step heat treatment will decrease the crystal size and block the light scattering.

4. Conclusions

The SHG intensity and propagation loss have been measured on transparent nanocrystallized glass fibers fabricated using a tellurite-based glass system. We have observed an SHG intensity of 1/400 of z-cut quartz on the crystallized glass fibers. The angular dependence of the SHG intensity was compared with, and explained by, an analysis of the angular dependence of the transmission factor of the fiber. The propagation loss of the crystallized glass fiber was 0.15 dB/cm, and this value was comparable to that of a typical $LiNbO_3$ waveguide. Discussion about the cause of propagation loss showed a large contribution of inner crystal particles for propagation loss, and it was suggested that the longer nucleation time on the crystallization process is effective in decreasing light scattering by crystal particles.

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