Non-Resonant Optical Nonlinearity of Germano-Silicate Optical Fiber Incorporated with Si Nanocrystals

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The non-resonant optical nonlinearity of a germano-silicate glass optical fiber with silicon (Si) nanocrystals incorporated, fabricated by using a modified chemical vapor deposition process, was investigated by the continuous-wave self-phase modulation method. The measured non-resonant nonlinear refractive index, \(n_2 = 5.70 \times 10^{-20}\) m\(^2\)/W, of the fiber was found to be \(~2\) times larger than that of the fiber without Si incorporation.

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I. INTRODUCTION

Optical signal processing technologies based on nonlinear optical phenomena are attracting much attention for future optical communication systems. All-optical switching and all-optical signal processing devices, such as optical wavelength converters [1], optical clock recovery systems [2], data regenerators [3] and pulse generators [4], have been intensely investigated. The Kerr effect is caused by the nonlinear response of the bound electrons of a host glass under an intense optical field, so optical devices based on the Kerr effect have great advantages for ultrafast operation [5]. Among the materials having the Kerr effect, highly nonlinear optical fibers are exceedingly effective for all-optical applications due to their comparability with currently-used silica-glass-based communications systems. Great efforts to fabricate various optical fibers with large Kerr nonlinearity have been made, such as erbium doped Bi\(_2\)O\(_3\)-based optical fibers [6], multi-step index Bi\(_2\)O\(_3\)-based optical fibers [7] and SF57-glass holey fibers [8].

In this paper, we report on the non-resonant optical nonlinearity of a germano-silicate optical fiber with quantum dots of silicon nanocrystals (Si-nc fiber) incorporated, which was fabricated by using a modified chemical vapor deposition (MCVD) process and a solution doping technique. To increase the non-resonant optical nonlinearity of the optical fiber, we have tried to incorporate Si nanocrystals having a large optical nonlinearity in the core of the silica glass fiber and not doping too much Ge in the core as with the well-known highly nonlinear optical fiber of Sumitomo. The non-resonant optical nonlinearity of the fiber was measured by using the continuous-wave self-phase modulation (cw-SPM) method [9-11].

II. EXPERIMENTS

The germano-silicate optical fiber with silicon nanocrystals (Si-nc fiber) incorporated was fabricated by using the MCVD and the fiber drawing processes. During the MCVD process, an ethanol solution containing a 0.3-g fine silicon powder (Kojundo, SIE17PB) less than 4 \(\mu\)m in size was prepared and used to incorporate silicon nanocrystals into the core of the fiber by using the solution doping method [12, 13]. Figure 1 shows the optical absorption spectrum of the fiber with Si-nc incorporated using the conventional cut back method. As shown in the figure, a broad absorption band appeared from 500 nm to 1200 nm, peaking at 740 nm, which originates from the Si-nc incorporated in the core of the fiber [13, 14]. Several spiky absorption peaks, 650, 850 and 1240 nm, were found in the Si-nc fiber. The spiky peaks are known to originate from the presence of hydroxyl groups in the fiber [14, 15]. The optical absorption peak near 1315 nm is due to a cut off of the fundamental mode in the fiber.

The photoluminescence (PL) spectrum of the Si-nc fiber upon pumping at 514 nm by an Ar-ion laser was
Fig. 1. Optical absorption spectrum of the germano-silicate optical fiber with silicon nanocrystals incorporated.

Fig. 2. Photoluminescence spectrum of the germano-silicate optical fiber with silicon nanocrystals incorporated.

Fig. 3. Experimental setup for non-resonant nonlinearity measurements by using the cw-SPM method [12, 13, 15]: TLS = tunable laser source, PC = polarization controller, BPF = band-pass filter, FUT = fiber under test, VOA = variable optical attenuator and OSA = optical spectrum analyzer.

also investigated [25, 26]. Figure 2 shows the PL spectrum of the Si-nc fiber for different pump powers of 30 and 100 mW. A broad PL band with the peak at 770 nm was observed and it also originated from the incorporated Si-nc [13]. In previous reports [13, 14], an overlaid PL peak was observed near 680 nm and was known to be induced by a drawing induced defect [14, 15]. In the present result shown in Figure 2, on the other hand, the PL peak was not clearly found at the corresponding region because lower pump powers, 30 and 100 mW, were used. The PL spectra shown in Figure 2 also had larger spectral noise in comparison with the previous results [13, 14]. The formation of nano-sized silicon crystals was also confirmed by using the optical emission spectrum and transmission electron microscopy [15].

The non-resonant optical nonlinearity of the Si-nc fiber was measured by using the continuous wave SPM method [9-11]. The experimental setup for the measurement is shown in Figure 3. In order to compare the non-resonant nonlinearities between the Si-nc fiber and the conventional germano-silicate single-mode optical fiber (SMF), a reference germano-silicate fiber (Ref. fiber) without incorporation of Si-nc was also made using the same MCVD and fiber drawing processes. For the examination, the nonlinearity of the SMF was also measured using the cw-SPM method.

III. RESULTS AND DISCUSSION

As shown in the cw-SPM experiment (Figure 3), the polarization states of the input light signals with wavelengths of $\lambda_1$ and $\lambda_2$ became parallel after passing through two polarization controllers (PCs). A band pass filter with a 3-dB bandwidth of 3 nm was used to reduce the amplified spontaneous emission (ASE). The coupled signals through the band pass filter were amplified by using an Er-doped fiber amplifier (EDFA) and were launched into the fiber under test (FUT). The zeroth and the first order harmonic intensities, $I_0$ and $I_1$, of the output signals were measured by using an optical spectrum analyzer (OSA) while changing the electric current of the EDFA. An optical attenuator (ATT) was used to protect the OSA from damage.

To determine the non-resonant nonlinear coefficient of the fiber, we measured the SPM nonlinear phase shift, $\varphi_{SPM}$ and the corresponding average power, $P_{AVG}$. Experimentally, the nonlinear phase shift was measured in the spectral domain. The ratio of the spectral components in terms of the Bessel function is given by

$$\frac{I_0}{I_1} = \frac{J_0^2(\varphi_{SPM}) + J_1^2(\varphi_{SPM})}{J_0^2(\varphi_{SPM}) + J_1^2(\varphi_{SPM})},$$

(1)
Table 1. Nonlinear optical parameters of the fiber with silicon nanocrystals incorporated.

<table>
<thead>
<tr>
<th>Fiber</th>
<th>Refractive index difference</th>
<th>Effective length $L_{eff}$</th>
<th>Effective area $A_{eff}$</th>
<th>Slope coefficient $\kappa_{ac}$</th>
<th>Nonlinear refractive index $n_2$</th>
<th>Effective nonlinearity $\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Symbol</td>
<td>Unit</td>
<td>$\Delta$</td>
<td>$L_{eff}$</td>
<td>$A_{eff}$</td>
<td>rad/W</td>
<td>$n_2$</td>
</tr>
<tr>
<td>SMF [16]</td>
<td></td>
<td>0.50</td>
<td>155.1</td>
<td>108.0</td>
<td>0.354</td>
<td>3.03</td>
</tr>
<tr>
<td>Ref. fiber [16]</td>
<td></td>
<td>1.00</td>
<td>153.0</td>
<td>63.05</td>
<td>0.472</td>
<td>2.30</td>
</tr>
<tr>
<td>Si-nc fiber</td>
<td></td>
<td>1.12</td>
<td>18.80</td>
<td>58.55</td>
<td>0.0147</td>
<td>5.70</td>
</tr>
</tbody>
</table>

where $I_0$ and $I_1$ are the intensities of the zeroth- and the first-order harmonics, respectively and $J_n$ is the Bessel function of $n$-th order. The non-resonant nonlinear refractive index $n_2$ and the nonlinear parameter $\gamma$ were estimated by using [11]

$$n_2 = \frac{\lambda A_{eff}}{4\pi L_{eff}} \left[ \frac{\varphi_{SPM}}{P_{AVG}} \right] = \frac{\lambda A_{eff}}{4\pi L_{eff}} \kappa_{ac},$$  \hspace{1cm} (2)

$$\gamma = \frac{2\pi n_2}{\lambda A_{eff}} = \frac{\varphi_{SPM}}{P_{AVG}} \frac{1}{2L_{eff}} = \frac{\kappa_{ac}}{2L_{eff}},$$  \hspace{1cm} (3)

where $A_{eff}$ is the effective area, $L_{eff}$ is the effective length of the fiber, $\lambda$ is the central wavelength of the two light signals ($\lambda_1 + \lambda_2$)/2 (in this experiment, $\lambda_1 = 1549.70$ nm and $\lambda_2 = 1550.25$ nm) and $\kappa_{ac}$ is the slope coefficient from the linear curve obtained by plotting $\varphi_{SPM}/P_{AVG}$.

The cw-SPM spectrum of the fiber with Si-nc incorporated is shown in Figure 4(a). The zeroth-order harmonic signals with intensity $I_0$ appeared at 1549.70 nm and 1550.25 nm and the first-order harmonic signals with intensity $I_1$ appeared clearly at 1549.14 nm and 1550.80 nm. $I_0$ and $I_1$ were found to increase linearly with increasing input pumping power of the EDFA. The average wavelength difference between $I_0$ and $I_1$ was 0.56 nm. Figure 4(b) shows the calculated nonlinear phase shift values of the three fibers vs. the input pumping power with the slopes. As shown in the figure, the nonlinear phase shift was found to increase linearly proportional with increasing input pumping power, regardless of the dopants inside the fiber, showing the accuracy and the stability of this cw-SPM measurement. The slopes of the Ref. fiber and the SMF were about 4 times and 2.5 times larger than that of Si-nc fiber, respectively. Even though the slope coefficient $\kappa_{ac}$ of the Si-nc fiber was smallest among the fibers, the Si-nc fiber showed the largest nonlinear refractive index $n_2$ and nonlinear parameter $\gamma$, which were more than twice those of the Ref. fiber.

The measured nonlinear optical parameters of the Si-nc fiber, together with those of the other fibers without Si incorporation, are summarized in Table 1. The refractive index difference of the SMF was more than twice smaller than that of the Ref. fiber and the Si-nc fiber, which means the germanium concentration of the SMF was about half those of the Ref. fiber and the Si-nc fiber.

On the other hand, the effective area of the SMF was much larger than those of the high-refractive-index fibers (Ref. fiber and Si-nc fiber). Due to the strong absorption by the incorporated Si-nc, the effective length of the Si-nc fiber was much larger than those of the germano-silicate fibers (SMF and Ref. fiber).

The non-resonant-type optical nonlinearity in oxide glasses is known to originate from the hyper-
polarizability of the glass constituents, such as bridging oxygens (BOs) and non-bridging oxygens (NBOs) [9, 16]. The BOs forms strongly covalent bonding with Si/Ge atoms and the bonds with the BOs are difficult to distort when they are subjected to an optical field. Therefore, only a very weak nonlinear effect arises from the structure related to BOs. On the other hand, the non-bridging oxygens (NBOs) have higher ionicity and are easily distorted by an applied optical field; thus, the glass with the NBOs has high optical nonlinearity [10, 12]. Various imperfect structures, such as NBOs and related defects (non-bridging oxygen hole centers (NBO-HCs), peroxy radicals, silicon oxugen deficient centers), have been reported to be created in silicon-rich silica thin films made by using plasma-enhanced chemical vapor deposition (PECVD) and Si-ion-implanted silica glass [18-20].

When germanium (Ge) ions are introduced into oxide glass, they are known to act as a network former and the proportion of NBOs in the silica glass network decreases [21]. Since the NBO concentration in the Ref. fiber with a higher Ge concentration (larger refractive index difference) was smaller than that of the commercial silica-glass-based single-mode fiber (SMF) with the lower Ge concentration (smaller refractive index difference), the non-resonant nonlinear refractive index of the Ref. fiber was smaller than that of the SMF. On the other hand, in the case of the Si-nc fiber, the Ge concentration was higher than that of the Ref. fiber; thus, the concentration of created NBOs was less. However, by incorporation of excessive Si into the fiber core during the fiber fabrication, a silicon-rich region in the fiber-core's glass composition might be introduced; thus, other NBOs and defects might be created. The increased optical nonlinearity of the Si-nc fiber, 5.70 × 10^{-20} \text{ m}^2/\text{W}, about 2 times of that of the SMF can also be explained by the contribution from the Si nanoparticles incorporated in the fiber. A large optical nonlinearity was also observed in semiconductors (GaAs, Ge, Si and InAs) and in heavy-metal-ion (Pb, Ti and Bi)-doped glasses due to their larger nonlinear polarizability [22, 23].

The nonlinearity is well known to be enhanced by increasing the concentration of Si-nc in glass. We were able to retain a small portion of Si particles during the collapsing process for making the fiber preform, as evident from Figures 1 and 2. However, it was very difficult to avoid the oxidation of Si particles during the process because of the high processing temperature, ~2000 °C. Even though the nonlinearity was increased, the enhanced nonlinearity was not very high in comparison with those observed in other semiconductors [22, 23]. In the other hand, our group found that the Si incorporation in the fiber had a strong effect on the enhancement of the resonant nonlinearity [24]. In summary, the large optical nonlinearity of the Si-nc fiber was found to be induced by the Si nanocrystals incorporated and by the polarizable structures, such as NBOs and defects, in a silicon-rich region and in the vicinity of Si nanocrystals.

IV. CONCLUSION

We have successfully fabricated germano-silicate optical fibers with Si nanocrystals incorporated for nonlinear optical applications by using MCVD and solution doping processes. By using the continuous-wave self-phase modulation method, we estimated the non-resonant nonlinear refractive index of the fiber to be 5.70 × 10^{-20} \text{ m}^2/\text{W}, about 2 times larger than that of the reference fiber without Si incorporation. Si nanocrystals were found to be effective for enhancing the non-resonant optical nonlinearity of the silica-based fiber, which may be due to the formation non-bridging oxygens induced by the incorporation of Si nanocrystals into the originally homogeneous germano-silicate tetrahedral structure.

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