

# Microphotoluminescence and Microphotorefectance Analyses of CO<sub>2</sub> Laser Rapid-Thermal-Annealed SiO<sub>x</sub> Surface With Buried Si Nanocrystals

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**Abstract**—The optical properties of a SiO<sub>x</sub> film rapid-thermal-annealed (RTA) by CO<sub>2</sub> laser are primarily investigated. The microphotoluminescence ( $\mu$ -PL) and high-resolution transmission electron microscopy (HRTEM) analyses indicate that the precipitation of random-oriented Si nanocrystals can be initiated when laser intensity ( $P_{\text{laser}}$ ) is larger than 4.5 kW/cm<sup>2</sup>. At  $P_{\text{laser}}$  of 6 kW/cm<sup>2</sup>, the Si nanocrystal exhibits a largest diameter of 8 nm and a highest density of  $4.5 \times 10^{16}$  cm<sup>-3</sup>, which emits strong PL at 790–825 nm. The microphotorefectance of the CO<sub>2</sub> laser RTA SiO<sub>x</sub> film reveals a volume density product dependent refractive index increasing from 1.57 to 1.87 as  $P_{\text{laser}}$  increases from 1.5 to 7.5 kW/cm<sup>2</sup>. Nonetheless, the laser ablation of the SiO<sub>x</sub> film occurs with a linear ablation slope of 35 nm/kW/cm<sup>2</sup> at beyond 7.5 kW/cm<sup>2</sup>, which terminates the enlargement of Si nanocrystals, degrades the near-infrared PL, and slightly reduces the refractive index of the CO<sub>2</sub> laser RTA SiO<sub>x</sub> film.

**Index Terms**—CO<sub>2</sub> laser annealing, microphotoluminescence, nanotechnology, Si nanocrystal, SiO<sub>x</sub>.

## I. INTRODUCTION

THE CO<sub>2</sub> LASER based zone annealing (or zone drawing) technique has previously emerged to modify the morphology or structural properties of different materials including polymers [1], [2], metallic thin films [3]–[5], superconductors [6], and dielectrics [7] etc. Particularly, such a laser heating process was also found to initiate the recrystallization and sintering of ceramic powders [8], or to enhance the surface crystallinity and the specific phase of an optical nonlinear crystal (beta-BBO) [9]. Optical microscopy has shown that the crystallite surface exhibits same morphology as those observed after traditional furnace processing; however, the effect of CO<sub>2</sub> laser annealing on the growth rate and the crystallite size is more pronounced. Not long ago, the CO<sub>2</sub> laser annealing was primarily employed to improve the properties of a liquid-phase deposited, fluorinated silicon oxide film, which helps to concentrate the fluorinated silicon oxide film and

reduce the effective surface charge density caused by surface defect states [7]. Nonetheless, the CO<sub>2</sub> laser annealing induced modifications are intensity ( $P_{\text{laser}}$ ) dependent and usually becomes prominent at  $P_{\text{laser}} > 10$  kW/cm<sup>2</sup>. Recently, the high-temperature (> 1000 °C) furnace annealing is employed to precipitate the Si nanocrystals in SiO<sub>2</sub> film [10]. However, such a high-temperature heat treatment could seriously damage the whole integrated circuits (ICs) on the same Si wafer, which constrains the monolithic integration of the Si nanocrystal doped SiO<sub>x</sub> layer with the Si-based ICs. Owing to the large absorption coefficient as high as  $1.2 \times 10^3$  cm<sup>-1</sup> of the oxide material at wavelength of 10.6  $\mu$ m, a CO<sub>2</sub> laser annealing of SiO<sub>x</sub> film on quartz substrate is investigated for the first time. In this paper, the optical properties and the size/density of localized precipitated Si nanocrystals embedded in the CO<sub>2</sub>-laser rapid-thermal-annealed (RTA) SiO<sub>x</sub> film are analyzed by high-resolution transmission electron microscopy (HRTEM), microphotoluminescence ( $\mu$ -PL), and microphotorefectance ( $\mu$ -PR).

## II. EXPERIMENTAL

The 280-nm-thick SiO<sub>x</sub> films were deposited on both-side-polished quartz substrates by using high-density plasma enhanced chemical vapor deposition (PECVD) with a gas mixture of SiH<sub>4</sub> and N<sub>2</sub>O. The substrate temperature was kept at 150 °C for 15 min to balance the temperature of the quartz substrate before deposition. The fluence ratio of SiH<sub>4</sub> to N<sub>2</sub>O, the rf power, and the reaction gas pressure were 1 : 6, 50 W, and 120 mtorr, respectively. Afterwards, the CO<sub>2</sub> laser RTA was performed in atmosphere using a CW CO<sub>2</sub> laser with  $P_{\text{laser}}$  ranging from 1.5 to 13.5 kW/cm<sup>2</sup>. The laser spot was focused within 0.2 mm<sup>2</sup> using a hemispherical lens. The CO<sub>2</sub> laser illuminating time was as short as 1 ms. The ablation thickness of SiO<sub>x</sub> film was measured by  $\alpha$ -step with a resolution of 1 nm. The  $\mu$ -PL (or  $\mu$ -PR) of CO<sub>2</sub> laser RTA SiO<sub>x</sub> pumped by a HeCd laser at 5 W/cm<sup>2</sup> and 325 nm was detected by a fluorescence spectrophotometer (Jobin Yvon, TRIAX-320). The  $\mu$ -PR analysis is demonstrated using a similar system with a HeNe laser at wavelength and power of 632.8 nm and 2 mW, respectively. A HRTEM (JEOL, 4000EX TEM) with a point-to-point resolution of 0.17 nm was used to characterize the orientation, lattice constant, size, and density of the precipitated Si nanocrystals in SiO<sub>x</sub> film.

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### III. RESULTS AND DISCUSSION

#### A. Surface Temperature Evaluation

The surface temperature ( $T_{\text{surface}}$ ) of the  $\text{SiO}_x$  film under the  $\text{CO}_2$  laser RTA process was determined using a previously developed thermal-physical model [11]–[13]. Neglecting the heat transport due to convection, the thermal radiation, and thermal expansion, the absorbed thermal energy per unit volume and per unit time of the  $\text{SiO}_x$  film annealed using a  $\text{CO}_2$  laser with a Gaussian-like power envelope can be described as  $g_{\text{laser}}(r, z) = 4P_{\text{laser}}(1 - R) \exp(-4r^2/D^2) \exp(-\alpha z)/\pi D^2 d_{\text{absorb}}$  [11], where  $P_{\text{laser}}$  is the illuminating intensity of the  $\text{CO}_2$  laser,  $R$  is the optical reflectivity of  $R = [(n - 1)^2 + k^2]/[(n + 1)^2 + k^2]$ ,  $r$  is the distance to the center of the focused laser spot,  $D$  is the beam diameter at  $1/e$  intensity of the Gaussian distribution,  $\alpha$  is the optical absorption coefficient of the  $\text{SiO}_x$  film, ( $\alpha = 4\pi k/\lambda$ ),  $\lambda$  is the laser wavelength, and  $d_{\text{absorb}}$  is the penetrating depth of the  $\text{CO}_2$  laser. The real ( $n$ ) and imaginary ( $k$ ) parts of the refractive index of the  $\text{SiO}_x$  film at the wavelength of  $10.6 \mu\text{m}$  in the room temperature are approximately 2.224 and 0.102, respectively. As a result, the temperature  $T(r, z)$  and surface temperature  $T_{\text{surface}}$  of the  $\text{SiO}_x$  film are expressed by the following equations [11], [12]:

$$T(r, z) = \frac{g_{\text{Laser}}(r, z)\tau}{\rho C_p} = \frac{4(1 - R)}{\rho C_p} \times \frac{P_{\text{Laser}}\tau}{\pi D^2 d_{\text{absorb}}} \times \exp\left(\frac{-4r^2}{D^2}\right) \times \exp(-\alpha|z|) \quad (1)$$

$$T_{\text{surface}}(r) = \frac{4(1 - R)}{\rho C_p} \times \frac{P_{\text{Laser}}\tau}{\pi D^2 d_{\text{absorb}}} \times \exp\left(\frac{-4r^2}{D^2}\right) \quad (2)$$

$$\frac{\partial T_{\text{surface}}(r)}{\partial r} = \left(\frac{-4r^2}{D^2}\right) \times \frac{4(1 - R)}{\rho C_p} \times \frac{P_{\text{Laser}}\tau}{\pi D^2 d_{\text{absorb}}} \times \exp\left(\frac{-4r^2}{D^2}\right), \quad r \geq 0 \quad (3)$$

where  $\tau$ ,  $\rho$  and  $C_p$  are the illuminating time, the density, and the specific heat of the  $\text{SiO}_x$  film, respectively. The optical absorption coefficient ( $\alpha$ ), the optical reflectivity ( $R$ ), the Gaussian beam diameter at  $1/e$  intensity ( $D$ ), the radial distance ( $r$ ), the illumination time ( $\tau$ ), the density ( $\rho$ ), and the specific heat ( $C_p$ ) of the  $\text{SiO}_x$  film are set as  $1209 \text{ cm}^{-1}$ , 0.145,  $370 \mu\text{m}$ ,  $21 \mu\text{m}$ , 1 ms,  $2800 \text{ kg/m}^3$ , and  $1270 \text{ J/kg/K}$ , respectively. According to these parameters, the simulated  $T_{\text{surface}}$  of the  $\text{SiO}_x$  film can be increasing from  $130 \text{ }^\circ\text{C}$  and  $3350 \text{ }^\circ\text{C}$  as the  $\text{CO}_2$  laser  $P_{\text{laser}}$  enlarges from  $1.5$  to  $13.5 \text{ kW/cm}^2$ . Therefore, the  $T_{\text{surface}}$  of the  $\text{SiO}_x$  film proportional to the  $\text{CO}_2$  laser  $P_{\text{laser}}$  is estimated from (2). For example, the  $\text{SiO}_x$  surface temperature profile around the annealed zone under illuminating with the  $P_{\text{laser}}$  of  $6 \text{ kW/cm}^2$  is shown in Fig. 1. The surface temperature at the central part of a Gaussian-beam illuminated zone is about  $1349 \text{ }^\circ\text{C}$ . The temperature gradient around the annealed zone of the  $\text{SiO}_x$  film is also plotted in Fig. 1. Within an illuminating spot of  $400 \mu\text{m}$  diameter, the maximum temperature gradient is only  $4.5 \text{ }^\circ\text{C}/\mu\text{m}$ .

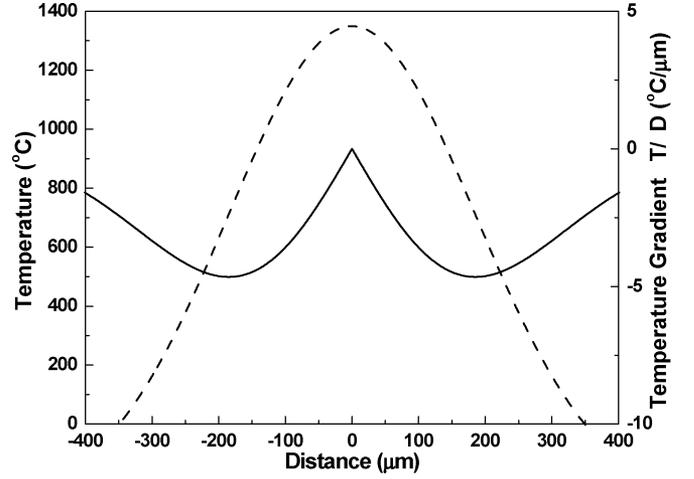


Fig. 1. Surface temperature and temperature gradient in the annealing zone of  $\text{SiO}_x$  illuminated by  $\text{CO}_2$  laser at  $P_{\text{laser}} = 6 \text{ kW/cm}^2$ .

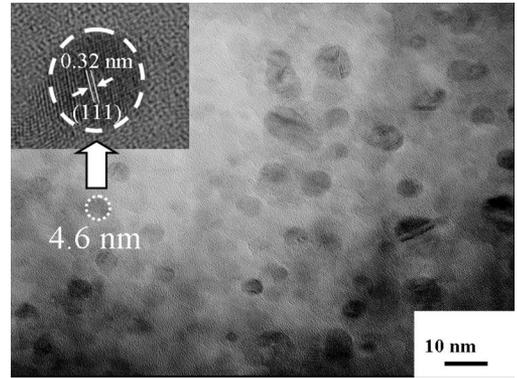


Fig. 2. HRTEM cross-sectional image of the laser RTA  $\text{SiO}_x$  film by  $\text{CO}_2$  laser at  $P_{\text{laser}} = 5.8 \text{ kW/cm}^2$ .

#### B. HRTEM Analysis

As a result, the simulated  $T_{\text{surface}}$  of the  $\text{SiO}_x$  film can increase from  $130 \text{ }^\circ\text{C}$  to  $3350 \text{ }^\circ\text{C}$  as the  $\text{CO}_2$  laser  $P_{\text{laser}}$  enlarges from  $1.5$  to  $13.5 \text{ kW/cm}^2$ . At  $P_{\text{laser}} = 5.8 \text{ kW/cm}^2$  (or  $T_{\text{surface}} = 1285 \text{ }^\circ\text{C}$ ), the cross-sectional HRTEM image reveals that the diameters of Si nanocrystals precipitated in the  $\text{SiO}_x$  matrix range from  $3$  to  $8 \text{ nm}$ , as shown in Fig. 2. The orientation of Si nanocrystals embedded in the  $\text{SiO}_x$  film is random, including the (111)-orientation with a lattice spacing of about  $0.32 \text{ nm}$ , as shown in the inset of Fig. 2. The HRTEM estimated density of the Si nanocrystals in the  $\text{CO}_2$  laser RTA  $\text{SiO}_x$  film at  $P_{\text{laser}} = 5.8 \text{ kW/cm}^2$  is about  $4.5 \times 10^{16} \text{ cm}^{-3}$ . Similar laser recrystallization was previously demonstrated by using a tightly focused continuous-wave  $\text{Ar}^+$  laser ( $\lambda = 514.5 \text{ nm}$ ) [14], which helps to synthesize Si nanocrystals in the hydrogenated amorphous  $\text{SiO}_x$  ( $\text{a-SiO}_x : \text{H}$ ) film. It was found that the diameter of the Si nanocrystals increases from  $2.5$  to  $12 \text{ nm}$  under an extremely high  $P_{\text{laser}}$  of ranging from  $600 \text{ kW/cm}^2$  to  $2.6 \text{ MW/cm}^2$ . However, the surface damage of the  $\text{a-SiO}_x : \text{H}$  film was also evidenced at such high intensities even with a short irradiating time. A later experiment showed similar results by using a frequency-tripled Nd : YAG laser at wavelength and pulsewidth of  $355 \text{ nm}$  and  $8 \text{ ns}$ , respectively [15]. By increasing

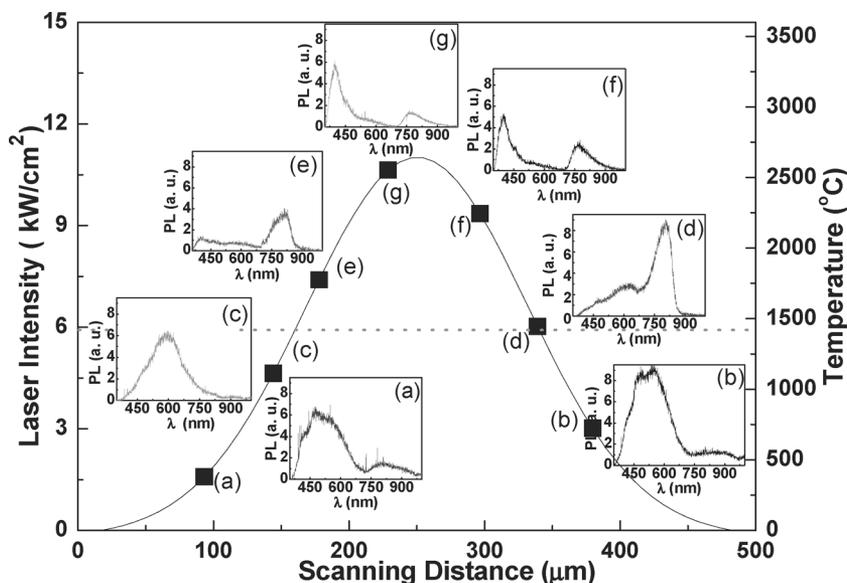


Fig. 3.  $\mu$ -PL spectra of CO<sub>2</sub> laser annealed SiO<sub>x</sub> at different site of the annealed spot area.

the peak energy density of the Nd : YAG laser up to 350 mJ/cm<sup>2</sup>, the size of Si nanocrystals can be enlarged to 200 nm. Such a process exhibits a similar surface damage problem, since the peak  $P_{\text{laser}}$  of 4.4 MW/cm<sup>2</sup> on the sample surface is far beyond the ablation threshold. Gallas *et al.* [16] then observed that the threshold energy density of a 248-nm KrF pulsed excimer laser for annealing SiO<sub>x</sub> without any ablation is only 85 mJ/cm<sup>2</sup>. Nonetheless, only a few Si nanocrystals can be precipitated in SiO<sub>x</sub> under such low  $P_{\text{laser}}$ , since few laser energies are absorbed and transferred to heat by the SiO<sub>x</sub> film with infinitely small absorption coefficient at such short wavelengths (for example,  $\alpha < 1 \times 10^{-6} \text{ cm}^{-1}$  at <532 nm) [13]. In contrast, the CO<sub>2</sub> laser crystallization can precipitate Si nanocrystals at a  $P_{\text{laser}}$  of at least two orders of magnitude smaller than that required for visible or UV lasers, which simultaneously eliminates the laser-ablation induced surface damage effect. The phase separation between Si and oxygen atoms can be initiated when sufficient energy is absorbed by the SiO<sub>x</sub> film; however, the annealing temperature for the precipitation of Si nanocrystals should be higher than 900 °C. Nesheva *et al.* [17] have observed the formation of amorphous Si nanoparticles in films annealed at 700 °C for 1 h. To format the crystallite Si nanocrystals in SiO<sub>x</sub> films, a furnace annealing at 1030 °C for 1 h is mandatory, as confirmed by Raman scattering analysis. In addition, Yi *et al.* [18] have also demonstrated that the amorphous Si clusters can be formatted at annealing temperature ranging between 300 °C and 900 °C, but the crystallization of these amorphous Si clusters is only observed by annealing in a nitrogen atmosphere at > 900 °C for 1 h. That is, the annealing temperature of lower than < 900 °C could not activate the crystallization process for the amorphous Si clusters, even if the annealing duration is lengthened. Under CO<sub>2</sub> laser annealing, the surface temperature of the SiO<sub>x</sub> film is dependent on the CO<sub>2</sub> laser intensity, the lower CO<sub>2</sub> laser intensity will reduce the surface temperature of the SiO<sub>x</sub> film. Our simulation has also interpreted a threshold annealing intensity of 4.5 kW/cm<sup>2</sup>, which corresponds to a sur-

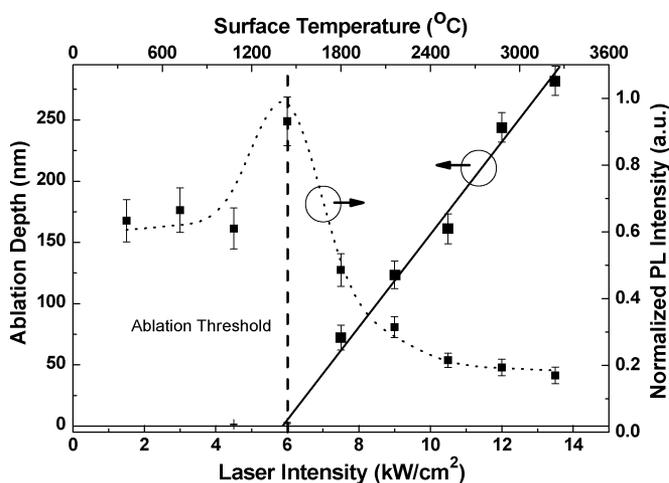


Fig. 4. Laser ablated depth and normalized PL intensity of the laser RTA SiO<sub>x</sub> film as a function of  $P_{\text{laser}}$ .

face temperature of 1100 °C for initiating the crystallization of the Si nanocrystals. Similarly, the Si nanocrystals are difficult to be precipitated and the size of the Si nanocrystals could not be larger at a lower CO<sub>2</sub> laser intensity even with longer exposure times.

### C. $\mu$ -PL and $\mu$ -PR Analyses

Subsequently, the  $\mu$ -PL and  $\mu$ -PR are employed to characterize the localized optical properties of the CO<sub>2</sub> laser RTA SiO<sub>x</sub> film. The power-dependent  $\mu$ -PL analysis reveals different luminescent features at annealing and ablation regions. The diameter and area of the CO<sub>2</sub> laser annealed zone are 500  $\mu\text{m}$  and 0.2 mm<sup>2</sup>, respectively. A tightly focused He-Cd laser beam, with a spot size much smaller than that of the CO<sub>2</sub> laser zone, transversely scans across the CO<sub>2</sub> laser annealed SiO<sub>x</sub> sample. In the outer area of the Gaussian laser spot, the SiO<sub>x</sub> matrix is not recrystallized and the Si nanocrystal is unable to

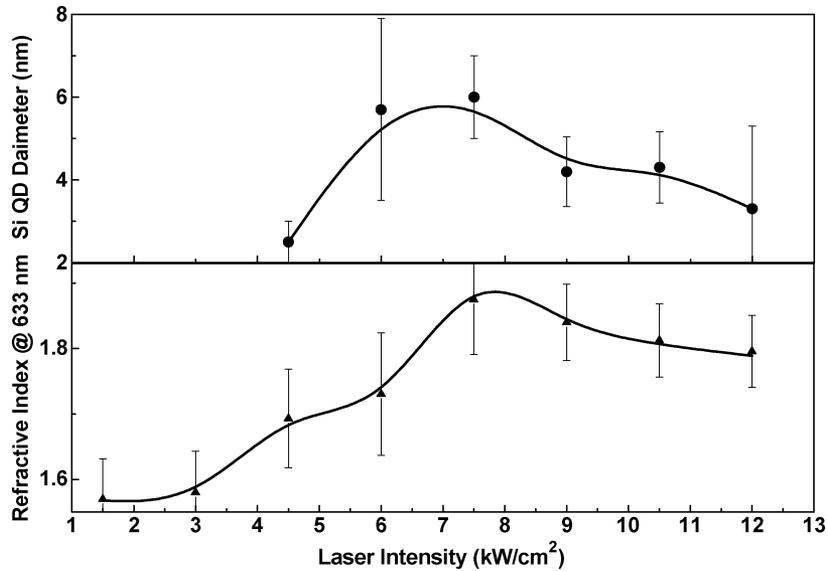


Fig. 5. The size of Si nanocrystal (upper part) and the refractive index of CO<sub>2</sub> laser RTA SiO<sub>x</sub> film (lower part) at different laser intensities.

be precipitated, since the  $P_{\text{laser}}$  as well as the equivalent  $T_{\text{surface}}$  is relatively low. A broadband blue–green PL contributed by the radiative defects such as  $E'_\delta$  centers [19] at 520 nm and the neutral oxygen vacancies centers [20] at 455 nm are observed (see Fig. 3). The PL peak red-shifts to 520 nm as  $P_{\text{laser}}$  slightly increases to 3 kW/cm<sup>2</sup> (equivalent to a surface temperature of 900 °C), indicating a more pronounced activation of the  $E'_\delta$  defects than the oxygen vacancies (see inset (b) of Fig. 3). Near the central region of the annealing spot, the precipitation of small-size Si nanocrystals (with a size of 0.8–1 nm) is initialized as the PL has further red-shifted to 600–620 nm at  $P_{\text{laser}} = 4.5$  kW/cm<sup>2</sup> (or a corresponding surface temperature of 1100 °C). The average size of Si nanocrystals persistently enlarges as the significant PL at 806 nm with spectral width of 106 nm is obtained at  $P_{\text{laser}} = 5.8$  kW/cm<sup>2</sup> (see inset (d) of Fig. 3). The maximum PL peak wavelength is observed at 825 nm at  $P_{\text{laser}} = 7.5$  kW/cm<sup>2</sup>. Nonetheless, the density of Si nanocrystals in SiO<sub>x</sub> also decreases at higher annealing laser intensities as the PL intensity at corresponding wavelength is greatly attenuated by an order of magnitude. The effect of  $P_{\text{laser}}$  on the laser ablation of the SiO<sub>x</sub> film clearly shows a linear ablation slope of 35 nm/kW/cm<sup>2</sup> at  $P_{\text{laser}}$  beyond ablation threshold (6 kW/cm<sup>2</sup>), as shown in Fig. 4. The corresponding temperature on the SiO<sub>x</sub> surface is up to 1902 °C as  $P_{\text{laser}}$  becomes > 7.5 kW/cm<sup>2</sup>, which has already exceeded over the melting temperature of fused silica. Moreover, a CO<sub>2</sub> laser annealing process at  $P_{\text{laser}} > 7.5$  kW/cm<sup>2</sup> not only anneals the SiO<sub>x</sub> film and precipitates Si nanocrystals locally, but also leads to an increasing of structural damage related PL at 410 nm by at least one order of magnitude. The SiO<sub>x</sub> matrix could be promptly compressed during such a rapid laser ablation procedure, where numerous oxygen-dependent defects such as weak oxygen bond (O–O) [21], oxygen vacancy, and ionized oxygen molecule (O<sub>2</sub><sup>-</sup>) [22], [23] with PL wavelengths at 410–455 nm are generated by the damaged bonds of the SiO<sub>2</sub> matrix (see inset (f)–(g) of Fig. 3). Such a phenomenon was never observed in furnace annealed SiO<sub>x</sub> film under a similar condition, as

the furnace annealing usually causes a gradual recovery on the compressing strain of SiO<sub>2</sub> matrix nearby Si nanocrystals.

The refractive indexes of CO<sub>2</sub> laser RTA SiO<sub>x</sub> films measured by  $\mu$ -PR greatly increase from 1.57 to 1.87 as  $P_{\text{laser}}$  increases from 1.5 to 7.5 kW/cm<sup>2</sup> (lower part of Fig. 5). The as-grown SiO<sub>x</sub> deposited under a N<sub>2</sub>O/SiH<sub>4</sub> flow ratio of 6 exhibits comparable refractive index with that reported by Ueno *et al.* [24]. The decrease in the N<sub>2</sub>O/SiH<sub>4</sub> flow ratio from 7 to 0.2 leads to an increasing refractive index of SiO<sub>x</sub> from 1.48 to 2.1. Such a variation is correlated with the precipitation of Si nanocrystals. At a surface temperature below 600 °C (or  $P_{\text{laser}} < 3$  kW/cm<sup>2</sup>), the change in refractive index is less than 0.6%, since the precipitation of Si nanocrystals has not yet been initiated. The refractive index of SiO<sub>x</sub> reaches a maximum of 1.87 as the surface temperature increases to 1300 °C, while the average diameter of Si nanocrystal is also largest. As evidence, Parakash *et al.* [25] have previously demonstrated that the enlargement of Si nanocrystals can increase the refractive indexes of the furnace-annealed PECVD-grown SiO<sub>x</sub> film. The density of the Si nanocrystal is also increased under higher annealing temperature and longer annealing time. A similar refractive index was also observed for the Si-ion-implanted SiO<sub>2</sub> doped by Si nanocrystals with size of 1–3.5 nm [26]. Obviously, the more Si atoms segregate from the SiO<sub>x</sub> matrix, the higher density of Si nanocrystals with uniform size can be obtained. From  $\mu$ -PL, the precipitation of Si nanocrystals in the SiO<sub>x</sub> film are initiated at  $P_{\text{laser}} > 4.5$  kW/cm<sup>2</sup> (or a corresponding surface temperature of 1100 °C), and the estimated size of Si nanocrystals is increased from 2.8 to 6 nm as  $P_{\text{laser}}$  increases from 4.5 to 7.5 kW/cm<sup>2</sup>, as shown in the upper part of Fig. 5. In contrast, the refractive indexes of the CO<sub>2</sub> annealed SiO<sub>x</sub> films slightly decrease from 1.87 to 1.79 as  $P_{\text{laser}}$  increases from 7.5 to 12 kW/cm<sup>2</sup>. Further increasing  $P_{\text{laser}}$  not only increases the annealing temperature but also activates the segregation of Si atoms and increases the density of the Si nanocrystal concurrently. From this point of view, it is not appropriate to attribute the change in refractive index of SiO<sub>x</sub> film only by the

size variation of Si nanocrystals. From HRTEM analysis, it is observed that both the size of the Si nanocrystal is decreased when annealing the SiO<sub>x</sub> films at  $P_{\text{laser}} > 7.5 \text{ kW/cm}^2$  due to the reoxidation of the Si nanocrystals. Moreover, the dramatically decreasing intensity of PL emitting from the Si nanocrystals also at higher annealing laser intensities corroborates well with a significant reduction in the density of the Si nanocrystals. This eventually results in a slightly decreasing value of the volume–density product for Si nanocrystals buried in the SiO<sub>x</sub>, which evidently elucidates the effect of size and density of Si nanocrystals on the variation in refractive index or absorption coefficient of SiO<sub>x</sub> films according to Mie theory [27]. Nonetheless, the effect of the dense oxygen dependent defects on the aforementioned optical constants should also take into account.

#### IV. CONCLUSION

In conclusion, the structural and optical properties of the PECVD-grown Si-rich SiO<sub>x</sub> film RTA by CO<sub>2</sub> laser are primarily characterized by using  $\mu$ -PL,  $\mu$ -PR and HRTEM in this work. The CO<sub>2</sub> laser RTA process performs the *in situ* and localized temperature control of the SiO<sub>x</sub> film, which thus facilitates precipitating Si nanocrystals from damaging the nearby devices. The equivalent temperature of the SiO<sub>x</sub> surface is increasing from 130 °C to 3350 °C as the CO<sub>2</sub> laser  $P_{\text{laser}}$  enlarges from 1.5 to 13.5 kW/cm<sup>2</sup>. The Si nanocrystals with maximum diameter and density of 8 nm and  $4.5 \times 10^{16} \text{ cm}^{-3}$ , respectively, can be locally precipitated within the CO<sub>2</sub> laser RTA SiO<sub>x</sub> film, giving rise to a near-infrared PL at 790–806 nm. These are obtained at just below ablation-threshold intensity ( $6 \text{ kW/cm}^2$ ), which is at least two orders of magnitude smaller than that required for visible or UV lasers. The power-dependent  $\mu$ -PL analysis indicates that the precipitation of small-size Si nanocrystals is initialized when  $P_{\text{laser}} > 4.5 \text{ kW/cm}^2$  and a maximum PL peak wavelength of 825 nm can be observed at  $P_{\text{laser}} = 7.5 \text{ kW/cm}^2$ . Nonetheless, the SiO<sub>x</sub> film is ablated with a linear ablation slope of  $35 \text{ nm/kW/cm}^2$  at beyond threshold  $P_{\text{laser}}$  of  $6 \text{ kW/cm}^2$ . The  $\mu$ -PR results indicate that the refractive index of the CO<sub>2</sub> laser RTA SiO<sub>x</sub> film varies from 1.57 to 1.87 as the  $P_{\text{laser}}$  increases from 1.5 to  $7.5 \text{ kW/cm}^2$ . At  $P_{\text{laser}} < 3 \text{ kW/cm}^2$ , the change in refractive index is less than 0.6%, since the precipitation of Si nanocrystals has not yet been initiated. The refractive index of SiO<sub>x</sub> reaches maximum as the surface temperature increases to 1285 °C, while the average diameter of the Si nanocrystal is also the largest. Annealing at higher intensities not only damages the SiO<sub>x</sub> structure, but also constrains the precipitation of Si nanocrystals and decreases the refractive index of the SiO<sub>x</sub>. This eventually degrades the near-infrared PL and reduces the refractive index of the CO<sub>2</sub> laser RTA SiO<sub>x</sub> film.

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