Cavity Q Measurements of Silica Microspheres with Nanocluster Silicon Active Layer

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Abstract—In this paper, the effect of the nanocluster-silicon (nc-Si) active layer on the cavity Q of silica microspheres is investigated. The silicon-rich silicon oxide (SRSO) (140 ± 10 nm thick) films with excess Si content ranging from 5 to 14 at.% were deposited on the silica microspheres formed by the CO₂ laser melting of an optical fiber, and subsequently annealed at temperatures ranging from 650 °C to 1100 °C. The cavity Q of the spheres with the active layer was measured at 1.56 μm using a tunable external cavity coupled laser diode and a tapered fiber coupling. We find that the presence of the nc-Si active layer reduces the Q value of the microsphere from ≥2 × 10⁷ to (2–5) × 10⁵. However, we found no correlation between the formation, size, and density of the nc-Si and the cavity Q-factor, indicating that the scattering by the nc-Si does not present the dominant optical loss mechanism in the SRSO film.

Index Terms—Loss mechanism, microsphere, nanocluster Si.

I. INTRODUCTION

The ever-increasing need for faster information transfer at a lower cost, both over the long-distance optical fiber network and the interchip/intrachip connections, has led to the emergence of the silicon photonics as a possible key field of the scientific and technological development. By merging the proven information carrying capacity of the silicon photonics with the infrastructure of the enormously successful silicon industry, silicon photonics could enable a mass production of the reliable and inexpensive optical components and systems. An interesting material for the silicon photonics is nanocluster Si (nc-Si). They can be used, either by themselves [1], [2] or when doped with rare-earth ions [3]–[5] to develop the Si-based light sources. Furthermore, nonlinear optical properties of the nc-Si are strongly enhanced over those of the bulk Si [6], [7]. Since the presence of nc-Si raises the refractive index of the dielectric host, thereby forming a natural waveguiding core, these results indicate that nc-Si may be used as the material basis for realization of efficient, optically active devices that are based entirely on Si.

But the presence of nc-Si in a dielectric such as SiO₂ leads to scattering due to the high refractive index contrast. Furthermore, the enhanced optical activity that make nc-Si interesting can exacerbate this problem, since properties such as polarizability and oscillator strength that lead to scattering are also enhanced. As this loss mechanism is intrinsic to the nc-Si themselves, its accurate characterization is critical for the development of any scheme for an optical device that relies on nc-Si, as well as an important part of the investigation of the fundamental optical properties of nc-Si.

So far, however, a systematic investigation of the nc-Si induced optical losses has not been reported, in part because the loss measurements usually require the formation of the waveguide whose fabrication can introduce scattering losses that are difficult to control accurately. In this paper, we report the measuring of the cavity Q-factor of silica microspheres with a silicon-rich silicon oxide (SRSO) consisting of nc-Si embedded inside a SiO₂ matrix, and the active surface layer formed by the plasma-enhanced chemical vapor deposition (PECVD). By taking an advantage of the ultrahigh-Q whispering gallery modes of a silica microsphere [8], [9], accurate measurements of even very small scattering losses are possible, and by depositing the SRSO layer on the surface, we can obtain a good overlap of the mode using a film that is likely to be used in an eventual nc-Si-based optical device. We find that the presence of the SRSO layer reduces the cavity Q-factor from ≥2 × 10⁷ to (2–5) × 10⁵. However, we find no correlation between nc-Si size, density, or even the presence of nc-Si with the cavity Q-factor, indicating that scattering by nc-Si does not contribute significantly to the optical loss. We will also discuss the implications of the result on the feasibility of realizing nc-Si-based optical devices.

II. EXPERIMENTAL CONDITIONS

A. Sample Preparation

Silica microspheres with a diameter of 150 μm were formed at the tip of tapered optical fibers by melting the tip with a CO₂ laser and taking advantage of the surface tension to form spherical droplets with an atomically smooth surface. After microsphere formation, SiO₂ (x < 2) layers of thickness 140 ± 10 nm and varying values of excess Si content were deposited using inductively coupled PECVD of SiH₄ and O₂ with Ar plasma. The microspheres were rotated along the fiber axis during the deposition process to ensure a uniform film deposition. Henceforth, they will be referred to as SiXX, with XX referring to the excess Si content in the SRSO film. High-temperature, post-deposition
annealing for 1 h in the flowing Ar environment was used to precipitate nc-Si. Two sets of SRSO-coated microspheres were fabricated. In one case, the anneal temperature was fixed at 1100 °C, while the excess Si content was varied from 5 to 14 at.% In the other case, the excess Si content was fixed at 10 ± 1 at.% while the anneal temperature was varied from 650 °C to 1100 °C. All samples that were annealed at 1100 °C were also hydrogenated by annealing for 1 h at 650 °C in the forming gas (90% N₂:10% H₂). For the comparison, a microsphere was deposited with a pure SiO₂ layer and also annealed at 1100 °C. Finally, in all cases, a piece of Si wafer was also placed in the deposition chamber during the deposition and subjected to the identical processing steps for the film analysis.

B. Sample Characterization

The composition and structure of the deposited films were analyzed with the Rutherford backscattering spectroscopy (RBS) and X-ray photoelectron spectroscopy (XPS) using the Al Kα line at 1486.6 eV. The photoluminescence (PL) spectra were obtained at room temperature using the 488-nm line of an Ar-ion laser, the 325-nm line of a HeCd laser, a grating monochromator, and by employing the standard lock-in technique. The PL spectra were corrected for the system response. The cavity Q-factors of the microspheres were obtained by coupling in the signal near 1.56 µm from a tunable external cavity laser with a linewidth of <300 kHz into the whispering gallery mode of the microsphere using a tapered fiber and measuring the transmitted intensity.

Fig. 1 shows the schematic of the measurement setup. The upper inset shows an optical microscope image of a microsphere with an SRSO active layer on the surface, showing the uniformity of the deposited layer. The lower inset shows a cross section SEM image of the deposited film, showing smooth sample surface and thickness uniformity. Since the spheres were rotated along the axis during deposition, and the width of the mode along the equator is only a few micrometers wide, scattering of light due to the variation of thickness along the equator is expected to be negligible.

III. RESULTS AND DISCUSSION

A. Film Properties

Fig. 2(a) shows the effect of annealing on the Si 2p core-level XPS spectra of the Si10 films (Si10a, Si10b, Si10c). The as-deposited film shows a peak at 103.9 eV due to the Si–O bonds as well as strong signals at 99.7 eV due to the Si–Si bond and at energies in between, indicating the presence of the amorphous Si (a-Si) nanoclusters and suboxides. Annealing at 1100 °C results in a clear separation of the Si–O and Si–Si peaks and a fivefold increase in the Si–Si bond signal, indicating a phase separation of Si and SiO₂. Such phase separation is accompanied by the emergence of nc-Si, as demonstrated by the high-resolution TEM image shown in the inset that shows nc-Si with a diameter of about 3 nm. Fig. 2(b) shows the effect of the excess Si content on the XPS spectra of the films that were annealed at 1100 °C. Note that the Si–Si bond signal at 99.7 eV increases continuously with increasing excess Si content, as expected.

Fig. 3 shows the PL spectra of the deposited films. As shown in Fig. 3(a), the formation of nc-Si is accompanied by a strong increase in the nc-Si PL intensity, as reported in [10] and [11]. Increasing the excess Si content increases the nc-Si size, as demonstrated by the monotonic redshift of nc-Si PL with increasing excess Si content shown in Fig. 3(b). From the PL spectra, we estimate the nc-Si radius to range from 0.7 to 1.9 nm [10], which is in good agreement with the TEM result shown in Fig. 2(A). Note that the PL energies and the size-induced redshifts are much lower than what would be expected for porous
Si or hydrogen-passivated nc-Si of the same size [12], but agree well with oxygen-passivated nc-Si [10]–[13], indicating that the nc-Si PL originate from the oxygen-related surface states.

Given the estimated nc-Si size and the excess Si content, we can calculate the nc-Si density and the refractive index of the SRSO active layer using the Maxwell–Garnett theory. Table I summarizes the results of such calculations. We estimate the mode-overlap to be about 2%–3% for all the samples based on the calculation of the radial component of the intensity.

### B. Estimation of Scattering Loss due to nc-Si

As the diameters of nc-Si are $\ll 1.56 \, \mu m$ and are randomly distributed, the scattering is well within the Rayleigh limit and is given by [14]

$$
\sigma = N \frac{8\pi n_{\text{silica}}^2}{3} \left( \frac{2\pi n_{\text{silica}}}{\lambda_0} \right)^4 \rho^6 \left( \frac{m^2 - 1}{m^2 + 2} \right)^2$

(1)

where $N$ is the nc-Si density, $\lambda_0$ the vacuum wavelength ($1.56 \, \mu m$), $a$ the nc-Si radius, and $m = n_{\text{nc-Si}}/n_{\text{silica}}$ is the ratio of the refractive index of the nc-Si to that of the surrounding medium (e.g., silica). Using the values given in Table I, we can estimate the contribution of nc-Si scattering loss to the reduction of the $Q$-factor of such nc-Si-coated microspheres. Table II summarizes the results of such calculations. We find that in all the cases, the Rayleigh scattering loss due to the nc-Si is $\ll 0.05 \, \text{dB/cm}$. Since the mode-overlap is about 2%–3% for all the samples, we estimate that the scattering by the nc-Si film can reduce the microsphere $Q$-factor to at most $2\times10^8$. Furthermore, since the scattering cross section is sensitively dependent on the radius of the scatter, the $Q$-factor should increase strongly with decreasing nc-Si size such that the Si14 sphere has a $Q$-factor that is 55 times smaller than that of the Si05 sphere, and the microsphere $Q$-factor should be dominated by nc-Si scattering.

#### C. $Q$-Factor of Microspheres with nc-Si Active Layer

Built-in differences between different microspheres and the inevitable variations in the microsphere fabrication make it impossible to compare the $Q$-factors of the identical optical modes between different spheres. Therefore, the highest $Q$-factors observed from each sphere were compared, under the assumption that they represent the high-$Q$ equatorial modes. The results are summarized in Fig. 4. The inset shows the typical transmission trace used to derive the $Q$-factor. The highest $Q$ value of $2 \times 10^7$ (corresponding to a modal propagation loss of 0.01 dB/cm of the excited mode) is observed from the microsphere that was deposited with the pure silica. The $Q$ values of the spheres deposited with the SRSO are much lower—however, they all remain within $(2–5) \pm 0.5 \times 10^5$ nearly independent of the excess Si content. These values for $Q$ translate to a modal propagation loss of 0.6–1.5 dB/cm.

The fact that the SRSO-coated spheres have much lower $Q$-factors than that of the silica-coated sphere indicates that the presence of the excess Si does lead to strong optical losses. On the other hand, the experimental results are in stark contrast to the theoretical estimates obtained in the previous section. First, the $Q$-factors of the SRSO-coated microspheres are lower by nearly three orders of magnitude. Second, they hardly show any dependence on the excess Si content, even though Fig. 3(b) convincingly demonstrates that nc-Si size increases with increasing excess Si content. Finally, the $Q$-factor of the as-deposited Si10 sphere changes only from $3 \times 10^5$ to $4 \times 10^5$ following the annealing at $1100^\circ$C and attendant formation of nc-Si.
D. Effect of nc-Si Size Distribution on nc-Si Scattering Loss

However, since the scattering cross section increases as \( a^6 \), the overall scattering loss can be dominated by the presence of a few, large nanoclusters. Therefore, in this section, we investigate the effect of the presence of the large clusters due to the nc-Si size distribution on the total scattering loss.

Based on the reports that the nc-Si size distribution is well described by a Gaussian function [10], we model the density of nc-Si with the radius varying from \( x \) to \( x + dx \) as

\[
N(x) \, dx = A \exp \left[ -\frac{1}{2} \left( \frac{x - a}{\delta} \right)^2 \right] \, dx \tag{2}
\]

where \( a \) is the mean radius of nc-Si (Table I), \( \delta \) the standard deviation of the nc-Si size distribution, and \( A \) is a constant. It is important to note, however, that \( A \) is not free, but is constrained by the requirement that the integrated volume of the nc-Si remain the same for a given film for all the values of \( \delta \). In other words, we require

\[
V_{\text{nc-Si}} = \int_{0}^{\infty} N(x) v(x) \, dx \tag{3}
\]

where \( v(x) = 4\pi x^3 / 3 \) is the volume of one nc-Si with a radius \( x \) and \( V_{\text{nc-Si}} \) is the total nc-Si volume fraction obtained from Table I. The total scattering cross section due to nc-Si with size distribution is then given by

\[
\sigma_{s} = \frac{8\pi}{3} \left( \frac{2\pi n_{\text{silica}}}{\lambda_0} \right)^4 \left( \frac{m^2 - 1}{m^2 + 2} \right)^2 \int_{0}^{\infty} N(x) x^6 \, dx. \tag{4}
\]

Fig. 5 shows the calculated effect of nc-Si distribution on the microsphere \( Q \)-factor, under the assumption that the optical loss is dominated by nc-Si scattering. We find that \( Q \)-factors decrease as the standard deviation increases. However, the decrease is much smaller than what would be expected by the simple \( a^6 \) dependence, since a wide size distribution leads to a strong reduction in the nc-Si density. Consequently, the \( Q \)-factor remains \( >3 \times 10^6 \) even for a \( \delta \) of 3 nm (e.g., with a significant number of nc-Si with \( >10 \) nm diameter). Furthermore, Fig. 5 indicates that the \( Q \)-factor should decrease monotonically with increasing excess Si, even with such a wide size distribution.

Such a wide size distribution, however, is inconsistent with the TEM results shown in Fig. 2(a) as well as with the previous investigations, which have reported \( \delta \) value of only about 0.5 nm [10]. On the other hand, it is possible that the radius of the nc-Si is underestimated, as it had been reported that there could be an amorphous Si shell covering the crystalline core that does not show up in the conventional high-resolution TEM [11]. Therefore, the effect of increasing the average nc-Si diameter while maintaining a \( \delta \) of 0.5 nm was investigated, as shown in Fig. 6. Again, we find that while the scattering loss due to nc-Si increases with larger radii, the overall effect is quite limited due to the strong concomitant decrease in the overall nc-Si density. Consequently, the \( Q \)-factor still remains \( >1 \times 10^7 \), even with an average nc-Si radius of 3.5 nm.

E. Possible Loss Mechanisms for Lowering of \( Q \) by nc-Si Active Layer

None of the above-mentioned results, however, are consistent with the experimentally observed \( Q \)-factors of (2–5) \( \pm 0.5 \times 10^5 \) that are nearly independent of the excess Si content. Furthermore, as Si10 sample shows, even the formation of the nc-Si has little effect on the \( Q \)-factor. Thus, the data suggest that nc-Si scattering is not responsible for the lowering of the microsphere \( Q \)-factor by the nc-Si active layer, and that the lowering of the \( Q \)-factor is dominated by another extrinsic loss mechanism in the SRSO. At this moment, it is not clear what such a loss mechanism could be, since the absorption of the 1.56-\( \mu \)m light by nanocrystal Si or pure silica is expected to be negligible. Furthermore, as the high \( Q \)-factor of the silica-coated...
sphere shows, the gross physical damage during processing is unlikely to be the main loss mechanism. It is possible that the presence of Si clusters, either amorphous or crystalline, produces nanometer-scale roughness at the air/SRSO and/or SRSO/silica interfaces that can lead to the modal losses of a few decibels per centimeter [15]. It is also possible that there are extrinsic defects in the SRSO that absorb the 1.56-µm light. For instance, it has been reported that a substantial fraction of the excess Si can remain in a-Si phase even after annealing at 1100 °C [11], [16], and that the absorption loss for the 1.5-µm light in the non-hydrogenated a-Si can be as high as 500 dB/cm [17]. Such an absorption by the remaining a-Si would be consistent with Fig. 4, as SRSO with smaller excess Si content is likely to have a larger fraction of the excess Si in the a-Si phase due to the increasing difficulty of nc-Si to crystallize with decreasing size [18].

If the Q-factor of the SRSO-deposited microspheres is dominated by the interfacial scattering, then the bulk optical loss of the SRSO would be expected to remain at 0.6–1.5 dB/cm, indicating the feasibility of the efficient, nanocrystal Si based optical devices. On the other hand, if the lowering of the Q-factor was due to absorption, then the bulk optical loss of the SRSO film would be more than 20–50 dB/cm, which is too large for an optical device. We note, however, that such absorption is not intrinsic to the presence of the nanocrystal Si, and can be reduced (e.g., by recrystallization of the remaining a-Si by laser annealing [16]). Therefore, the efficient, practical nanocrystal Si based optical devices would still be feasible, provided that care is taken to engineer the film structure to consist of the well-defined nanocrystal Si and pure silica only.

IV. CONCLUSION

In conclusion, we have investigated the effect of depositing an active layer of nanocrystal Si on silica microspheres. We observed strong reduction of the cavity Q-factor by the active layer. However, we found no correlation between the formation, size, and density of nc-Si and cavity Q-factor, indicating that scattering by nc-Si does not present a significant loss mechanism, and demonstrating the feasibility of the efficient, practical optical devices based on nanocrystal Si.

REFERENCES

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