Nanowatt threshold, alumina sensitized neodymium laser integrated on silicon

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Abstract: Low threshold lasers based on rare-earth elements have enabled numerous scientific discoveries and innovations in industry. However, pushing the threshold into the sub-microwatt regime has been stymied by a fundamental material phenomenon. Specifically, rare earth dopants form clusters which quench emission and reduce efficiency. Here, we fabricate resonant cavity lasers from neodymium-doped silica films containing alumina. The alumina prevents the clustering of the Neodymium, enabling the lasers to achieve thresholds of 530 nanoWatts at room temperature.

OCIS codes: (130.0130) Integrated optics; (140.3530) Lasers, neodymium; (140.4780) Optical resonators; (160.5690) Rare-earth-doped materials; (160.6060) Solgel.

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1. Introduction

High efficiency, low threshold fiber lasers based on rare-earth metal doped glasses have enabled numerous industries and applications, ranging from defense to biological investigations [1–3]. While there are several motivations for designing an optically-pumped laser system based on a rare earth element, two reasons stand out. First, using a single pump source, it is possible to generate a wide range of emission wavelengths due to the multiple excitation and decay pathways of rare earth ions. For example, when neodymium is pumped near 800nm, lasing near 900-940nm, 1050-1150nm, and ~1330nm can be observed [4–6]. Second, rare earth ions have very large absorption cross sections for excitation. Because the laser threshold is inversely proportional to the gain coefficient, they are an ideal gain medium.

However, the field of optoelectronics is moving towards the development of integrated photonic devices, in which all of the individual components are fabricated directly on a silicon substrate. This approach requires the transformation of fiber laser systems into integrated microscale devices. One straightforward approach is to use integrated waveguides in lieu of optical fiber [5, 7, 8]. However, the optical loss of integrated waveguides, which affects the laser threshold, is significantly higher than optical fiber.

To compensate for this performance decrease, there are two other parameters related to the laser threshold which can be modified: 1) the concentration of the gain media and 2) the optical field intensity inside the waveguide. However, rare earth dopants have limited solubility in silica and will begin to cluster at high concentrations, quenching the emission. As such, assuming a simple silica matrix, there is a maximum concentration which can be used. Additionally, the primary method for tuning the optical field intensity in fiber-based lasers is to change the refractive index of the fiber. However, this approach is not able to provide the orders of magnitude enhancement in optical field intensity necessary to decrease the thresholds from mW to μW-levels.

An alternative approach to increase the optical field intensity and improve the laser performance is to transition from a simple waveguide platform to an optical resonant cavity. In a fiber laser, photons only have a single opportunity to interact with the gain media. In contrast, resonant cavities confine light in circulating orbits, increasing the number of interactions in proportion to the photon lifetime or quality factor of the cavity, which in turn is proportional to the circulating intensity. This improvement in excitation efficiency can be analytically described by the expression for the enhancement factor (E) [9]:

$$E = \frac{(1 - \gamma_p)(1 - \kappa)}{(1 - (1 - \gamma_p)\frac{\kappa}{\pi\alpha_p}e^{-\alpha_p\pi D}/2)} \frac{1 - e^{-\alpha_p D}}{\alpha_p\pi D}$$

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where $D$ is the resonator's diameter, $\kappa$ is the coupling efficiency, $\gamma_p$ is the coupling loss and $\alpha_p$ is the total cavity loss, which is inversely related to the intrinsic cavity $Q$. At critical coupling, when $\kappa$ is at its ideal value, Eq. (1) can be simplified to Eq. (2). From these expressions, the direct dependence of the enhancement factor on the coupling efficiency and the quality factor is evident.

The threshold power ($P_{\text{thres}}$) is defined as the power at which the signal loss equals the gain. As such, in cavity-based rare-earth lasers, it is dependent on the balance between the gain concentration and the cavity $Q$, and a simple pair of expressions can be written to establish the lasing condition. The first expression defines the balance between the $Q$ of the cavity (cavity loss) and the dopant concentration ($N_T$) [9, 10]:

$$\alpha_p < \Gamma \sigma_s' N_T.$$  

where $\Gamma_s$ is the overlap factor which describes the interaction strength between the optical intensity and the rare earth distribution in the material, and $\sigma_s'(\sigma_s')$ describes the optical absorption (emission) cross sections of the rare earth material used. Using Eq. (3), a lower bound on the $Q$ of the signal can be determined (using $Q = \frac{2\pi n}{\lambda \alpha_p}$):

$$Q > \frac{2\pi n}{A \Gamma_s \sigma_s' N_T}.$$  

From these conditions, an expression for the threshold power can be written [9]:

$$P_{\text{th}} = \frac{1}{\Gamma_s} \frac{\sigma_s'}{\sigma_s} e^{-\alpha_p} \frac{\alpha_p}{\Gamma_s \sigma_s' N_T} \frac{1}{E} \frac{A}{E}$$  

where $A$ is the cross section of the resonator. One important aspect of this expression is that the quality factor appears in both the enhancement factor ($E$) and in the cavity loss ($\alpha_p$) terms. However, as seen in Eqs. (1) and (5), both terms are also dependent on other factors, such as coupling and gain concentration. As such, with low dopant concentrations or by operating in the critical coupling regime, the dependence of $P_{\text{thres}}$ on $Q$ can be simplified to $P-Q^2$; however, in other regimes of operation, the behavior can be significantly more complex.

Given the relations in Eqs. (1)–(5), high $Q$ devices containing dopants with large absorption cross sections are desirable for laser applications. For example, in a silica cavity with a quality factor ($Q$) of 100 million, the increase in circulating intensity is approximately 100,000 times. Using this approach, lasing thresholds as low as tens to hundreds of nanowatts have been achieved in rare earth-doped spheres [11, 12]. However, silica microspheres are not integrated on silicon wafers; therefore, they face many of the same challenges as fiber lasers.

Combining the high $Q$ factors of the microsphere cavity and the requisite on-chip fabrication, silica toroid resonators are an especially promising platform for integrated rare earth lasers [13]. Recent research has demonstrated rare earth-doped silica toroid microlasers with microwatt thresholds. For example, a 4.2 $\mu$W lasing threshold has been achieved in a ytterbium and erbium codoped silica toroid [14], and a neodymium-doped toroid integrated on silicon recently demonstrated a 69$\mu$W lasing threshold [15]. While these results are promising, integrated silica devices have not been able to achieve sub-microwatt lasing thresholds. One reason for this limitation is the limited solubility of rare earth dopants in silica. This causes the dopants to form clusters which quench emission, even at moderate concentrations [16].
One solution for reducing dopant clustering and improving laser performance is to incorporate metal ions, such as aluminum, into the silica host matrix. Aluminum ions have been observed to selectively coordinate around neodymium ions, preventing clustering [6, 17–19]. In particular, the Nd³⁺ ions tend to coordinate favorably with Al-O bonds in the silica matrix. This stabilizes the Nd³⁺ ions and disperses them more uniformly throughout the silica [20]. As a result of this coordination and the decrease in clustering, the fluorescence behavior of Nd³⁺ ions improves significantly [18–21].

In the present work, we demonstrate an alumina-sensitized neodymium-doped silica toroid microlaser integrated on a silicon wafer (Fig. 1(a)). Specifically, we synthesize a series of silica sol-gels which are co-doped with 0.1 mol% Nd³⁺ and 0 to 2 mol% alumina. Then, we fabricate toroid microlaser devices from the films and characterize the dependence of lasing threshold and emission behavior on the alumina concentration. Since the alumina reduces clustering and creates a more favorable local environment around the Nd³⁺ ions, the lasing threshold and efficiency are significantly enhanced, enabling sub-microwatt lasing thresholds in an integrated silica device.

2. Material synthesis and device fabrication
The Nd³⁺ and alumina-doped silica is synthesized using the following sol-gel method, in which the silica precursor tetraethoxysilane (TEOS) undergoes an acid catalyzed hydrolysis-condensation reaction. Tetraethoxysilane (TEOS, Alfa Aesar, 99.999%), aluminum isopropoxide (Sigma Aldrich, 99.99%), ethanol, deionized water, and nitric acid (HNO₃, EMD, 68%), are combined in a glass vial, stirring 5 minutes between each addition. The molar ratios of (TEOS: ethanol:water:HNO₃) are set to (1:4:4:0.05), and the molar ratio of aluminum isopropoxide is varied between 0 to 0.02 to control the resulting concentration of alumina. After the addition of HNO₃, the mixture is stirred for 1 hour at 70°C, allowing the TEOS and aluminum isopropoxide to undergo hydrolysis and condensation reactions. While the sol gel is stirring, neodymium nitrate (Alfa Aesar, 99.99%) is dissolved in deionized water in a 1:10 molar ratio. When the TEOS solution has finished stirring, the neodymium solution is added to the TEOS solution and stirred for 30 minutes. Afterwards, the solution is aged for 60 hours in ambient conditions.

Silica films are created by spin-coating the aged sol gels onto bare silicon wafers at 7000rpm for 30 seconds. The coated wafers are dried on a 75°C hot plate for 5 minutes, and then annealed in a tube furnace at 900°C for 2 hours with a ramp rate of 1°C/minute. This process creates a 350nm thick film, and is repeated once more to achieve a ~700nm thick film, as measured by ellipsometry. Once the annealing steps are complete, 40 μm diameter toroidal microring devices are fabricated from the sol gel silica films using a combination of photolithography and etching processes followed by a laser reflow step (Fig. 1(a)) [13, 22].

3. Device and material characterization
To characterize the quality factor and the lasing behavior of the toroid lasers, we couple light into the cavity from a 765-781nm tunable narrow linewidth laser (Velocity series, Newport) using a tapered optical fiber waveguide (Fig. 1(b)–1(d)). The tapered fiber’s output containing both the pump light and the toroid laser’s emitted light is sent to a 90:10 splitter. The 90% end goes to an optical spectrum analyzer (Agilent 86142B) and the remaining 10% to a detector and high speed digitizer/oscilloscope (National Instruments). This allows simultaneous monitoring of the toroid’s emitted light on the OSA and the amount of light coupled into the toroid laser on the digitizer/oscilloscope.
To measure the resonators’ quality factors (Q), the transmission spectrum is recorded using the oscilloscope and is fit to a Lorentzian. The linewidth is determined from the full-width-half-maximum of the fit, and from this value, the Q of the device is determined (Q = λ/δλ). During the Q measurement, the scan range and rate of the laser are optimized to minimize any non-linear effects which might distort the linewidth (δλ). In laser characterization experiments, the toroid laser’s emission intensity is measured with different amounts of power coupled into the toroid. Then, the emission versus coupled power is plotted and a line is fit to the data. The lasing threshold is given by the x-intercept of the line, while the laser's slope efficiency is given by the slope of the line.

After fabrication, we first measure the effective refractive index (n_{eff}) of the devices to verify the presence of alumina in the silica films. The effective refractive index is related to, but distinct from, the refractive index of the film, and can affect the threshold power of the laser as seen in Eq. (4). The effective refractive index is a composite refractive index, which can be described by: n_{eff} = \alpha n_{\text{silica}} + \beta n_{\text{air}}, where n_{\text{silica}} and n_{\text{air}} are the indices of the silica toroid and the air, respectively, and \alpha and \beta describe the percent of the optical field which is located in each media. Because the free spectral range (FSR), or the spacing between sequential resonant wavelengths, is directly related to the n_{eff}, one effective method for experimentally measuring the n_{eff} is to measure the free spectral range (FSR) of the device. Specifically, the n_{eff} = \lambda^2/(\pi D \cdot \text{FSR}) where D is the diameter of the device and \lambda is the wavelength. In the present work, the FSR values are experimentally determined by measuring the distance (in nanometers) between adjacent lasing peaks. Based on the measured FSR values, the effective refractive index increases with increasing alumina concentration (Fig. 2(a)). Given that alumina’s refractive index (~1.77) is greater than silica’s (~1.44), this refractive index increase indicates that alumina has been successfully added to the films [23]. Since the sol gel silica is slightly porous, the effective refractive index of the alumina-free toroids is only 1.42. As more alumina is added, the refractive index increases to over 1.45.
4. Effects of alumina concentration on quality factor and microlaser performance

Typically, the quality factor of these devices depends primarily on material absorption losses and is given by the equation $Q = \frac{2\pi n_{\text{eff}}}{\lambda \alpha_{\text{eff}}}$ where $\lambda$ is the wavelength of light and $\alpha_{\text{eff}}$ is the effective material absorption coefficient. An initial concern about adding metal ions such as aluminum to silica is that material absorption losses ($\alpha_{\text{eff}}$) would increase and therefore degrade the quality factor and lasing performance. However, as seen in Fig. 2(b), adding alumina actually increases the quality factor. This behavior can be attributed to two effects. First, as observed in the previous section, the refractive index increases with increasing alumina concentration causing the quality factor to also increase. Second, when alumina is added to Nd$^{3+}$-doped silica, the optical absorption coefficient has been observed to decrease due to more favorable coordination of Nd$^{3+}$ with alumina in the silica [18, 24]. Therefore, high quality factors of over 1 million can be achieved in the alumina-containing devices.

When pumped with the tunable laser near ~780nm, all of the neodymium-doped toroid lasers showed emission peaks in the 900-940nm range and 1050-1150nm range (Fig. 3(a)). These emitted wavelengths correspond to stimulated emission from the $^4F_{5/2}$ energy level to the $^4I_{9/2}$ and $^4I_{11/2}$ levels, respectively [4, 5]. It is important to note that the number of observed lasing peaks (hence, the number of lasing modes) depends on many factors, including the coupling, input power, dopant concentration, device materials, and geometry. This balance is also evident in Eq. (5). Therefore, by optimizing these parameters, either single mode or multimode lasing can be achieved.

As the alumina concentration increases, the lasing wavelength range shifts from ~940nm and ~1080-1160nm in alumina-free microlasers to ~900nm and ~1050-1130nm in silica containing 2 mol% alumina (Fig. 3(b)). At greater alumina concentrations, more neodymium ions are coordinated with Al-O bonds instead of Si-O bonds. When coordinated with alumina, the Nd$^{3+}$ ions have reduced clustering, quenching and nonradiative decay compared to plain silica [18, 21, 24]. In addition, the alumina has a lower phonon energy than the sol gel silica, causing the Nd$^{3+}$ emission to blue shift slightly [18, 21, 24]. Therefore, the observed lasing at slightly higher energy wavelengths further indicates that the neodymium ions become surrounded and enhanced by alumina.
As a result of the improved coordination between alumina and Nd$^{3+}$, the lasing threshold is significantly reduced. Samples with 2 mol% alumina achieved ultra-low thresholds as low as 530 nanoWatts. A representative lasing spectrum and the threshold curve are shown in Fig. 4. As mentioned, previous work using Nd$^{3+}$-doped toroid cavities achieved 69$\mu$W thresholds, so the present work represents a 130-fold improvement in performance [15].

Due to the lasing threshold’s dependence on Q, directly plotting threshold versus the alumina concentration can lead to a misinterpretation of the results. One method for disambiguating this behavior is to normalize the threshold by the quality factor of the device (Eq. (5)). While the dependence due to the enhancement in excitation efficiency (E) can be easily isolated in Eq. (5) and normalized from the data, the dependence on cavity loss is more challenging to extricate from the other terms, particularly when high dopant concentrations are used. In these situations, normalizing by Q, and not Q$^2$, is appropriate. When this analysis is performed, a clear trend emerges. Specifically, the lasing threshold/Q decreases linearly with increasing alumina concentration (Fig. 5(a)). Additionally, the lasing threshold/Q ratio decreases over 5-fold as the alumina concentration increases, significantly enhancing the device performance.
In addition to decreasing the lasing threshold, increasing the alumina concentration greatly improves the slope efficiency of the toroid microlasers. As the alumina concentration is increased from 0 to 2 mol%, the slope efficiency of the toroid laser increases up to 29-fold. This efficiency increase is linear with alumina concentration over this range (Fig. 5(b)). The simultaneous improvement of lasing threshold and slope efficiency is notable, as typically improving one performance metric comes at the cost of worsening the other.

It is also important to mention the slope efficiency values in Fig. 5(b) could be further improved by optimizing the coupling of the laser’s emitted light back into the tapered fiber or employing an alternative method for extracting the emission. As shown previously in Fig. 1(d), the tapered fiber in the present work simultaneously coupled the ~780nm pump light into the toroid and coupled the ~900-1150nm Nd\textsuperscript{3+} lasing out of the toroid. As a result, the coupling efficiency over this entire wavelength range was not ideal, and the slope efficiency values appear lower.

5. Conclusion

In conclusion, we have successfully fabricated toroid microlasers from neodymium-doped silica. The addition of alumina to the Nd\textsuperscript{3+}-doped silica reduces clustering of Nd\textsuperscript{3+} dopant, causing emission at slightly shorter wavelengths, up to a 29-fold increase in slope efficiency, and enabling sub-microwatt lasing thresholds to be achieved in a lasing device which is integrated on silicon. Therefore, these efficient and ultra-low threshold alumina-enhanced lasers will benefit applications in integrated optics and communications. Since the absorption of water is especially low at the ~800nm pump and ~1064nm emission wavelengths of neodymium [25], these devices will find useful applications in biosensing as well [26–29].

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