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Slot waveguide ring resonators coated by an atomic layer deposited organic/inorganic nanolaminate

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Abstract: In this study, slot waveguide ring resonators patterned on a silicon-on-insulator (SOI) wafer and coated with an atomic layer deposited nanolaminate consisting of alternating layers of tantalum pentoxide and polyimide were fabricated and characterized. To the best of our knowledge, this is the first demonstration of atomic layer deposition (ALD) of organic materials in waveguiding applications. In our nanolaminate ring resonators, the optical power is not only confined in the narrow central air slot but also in several parallel sub-10 nm wide vertical polyimide slots. This indicates that the mode profiles in the silicon slot waveguide can be accurately tuned by the ALD method. Our results show that ALD of organic and inorganic materials can be combined with conventional silicon waveguide fabrication techniques to create slot waveguide ring resonators with varying mode profiles. This can potentially open new possibilities for various photonic applications, such as optical sensing and all-optical signal processing.

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1. Introduction

Over the last two decades, tremendous interest has been given to silicon photonics [1], which greatly benefits from the extensive manufacturing infrastructure and mature processing technology of silicon, providing potential for cost-efficient fabrication [2]. Despite all the eminent advantages of silicon, other materials typically need to be integrated to fulfill the functions that are difficult to realize with silicon alone. This is because silicon has a low light emission efficiency and a low electro-optic coefficient, limiting the use of silicon as a material for light sources and modulators [3]. A strong two-photon absorption (TPA) at high intensities also limits the use of silicon in applications exploiting nonlinear effects [4]. For example, optically active materials can be applied as a cladding material to provide electro-optic modulation [5]. However, propagating mode is strongly confined in the silicon core due to the high refractive-index contrast between the silicon core and a low-index cladding material (e.g., SiO$_2$ in SOI waveguides), leading to weak interaction between these active materials and guided light. To address this issue, slot waveguides have been proposed to maximize the overlap between the low refractive-index cladding material and the propagating mode [6]. In this case, light is confined in a low refractive index material, and the operation is still based on total internal reflection (TIR). At the interface, where index contrast is high, the electric field must undergo a large discontinuity with a much higher amplitude in the low-index material; therefore, slot waveguides offer stronger light confinement for light-matter interactions outside the silicon core for various applications (e.g., sensing and all-optical signal processing) [7, 8].

Currently, organic materials have been widely used for various photonic and optoelectronic devices, because they possess properties that are desirable for optical communications. These properties are, for example, large and ultrafast nonlinear responses, high optical cross-sections and a broad spectral tunability [9]. Recently, a silicon-organic hybrid (SOH) approach has been used to demonstrate high-speed signal processing with a combination of slot waveguides and organic materials [4]. Spin-coating is a widely used for fabrication of traditional organic photonic devices [10]. However, it is not an ideal method for completely filling narrow slots, which is important for achieving low propagation losses. Also, the film thickness cannot be controlled accurately with the spin-coating approach. Atomic layer deposition (ALD) can be used for depositing thin-film organic materials on various substrates, offering huge advantages [11, 12]. In ALD, thin films are deposited with alternating pulses of precursor gases [13]. The film growth mechanism is self-limiting which enables the control of film thickness and composition with

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atomic-level accuracy. ALD provides conformal films with large-area uniformity [14], even on patterned structures, which makes ALD a promising technique for improving performance of photonic devices. ALD can be used for depositing materials on top of flexible and non-standard substrates, which is a valuable advantage especially for depositing organic materials. In addition, ALD is compatible with standard CMOS technology. The main limitation of ALD is its slow growth rate. However, ALD is inherently suitable for batch processing. Simultaneous coating of large number of substrates compensates for the relatively long deposition time. This is particularly true in the case of silicon nanophotonic waveguides, where large wafer batches are used, but layer thickness in the order of 100 nm is typically sufficient. Indeed, ALD has recently been proven to be an excellent method to improve the quality of slot waveguides (e.g. reduce propagation loss in slot and strip waveguides [15, 16]), due to the high-precision of ALD and a wide variety of available ALD materials. Inorganic ALD materials, such as aluminum oxide (Al2O3), titanium dioxide (TiO2) and hafnium oxide (HfO2) have been used in several different waveguiding applications [17–21].

In this work, we demonstrate that ALD can be employed to create high-performance slot waveguide devices with hybrid organic/inorganic materials. The nanolaminate coating acts as a metamaterial having a refractive index depending on the thickness of each layer. Thus the effective index of the propagating mode and the field confinement can be accurately tuned by varying the layer thicknesses without changing the slot material. This is valuable for optimizing slot waveguides for applications in e.g. all-optical signal processing. Strong nonlinearities over broad wavelength range are desired in many applications, including all-optical signal processing [8]. For these applications it is important to be able to control the group velocity dispersion (GVD) of the waveguides [22]. The dispersion properties in slot waveguides have been tailored by changing the waveguide structure [23,24]. Also, ALD has been used for tailoring the dispersion properties in strip waveguides [25] and ring resonators [20]. The nanolaminate structure demonstrated here offers potential for dispersion engineering by optimizing the layer thicknesses and the number of individual layers. Also the layer materials can be selected from wide variety of available ALD materials for tailoring the dispersion properties.

Ring resonators are selected to be studied in this work, as they are one of the basic building blocks for numerous different applications in silicon photonics (e.g., modulators [26–29], optical delay lines [30], filters [31] and sensors [7,32,33]). Tantalum pentoxide (Ta2O5) and polyimide (PI) are selected as ALD materials since the ALD process for depositing Ta2O5/PI nanolaminate films is already available [11,12,34] and both materials exhibit high third order nonlinearities [35–37]. In principle other inorganic materials, such as Al2O3 or TiO2, can also be used instead of Ta2O5. Our results show that ALD of organic and inorganic materials can be combined with conventional silicon waveguide fabrication techniques to create slot waveguide ring resonators with varying mode profiles. This can potentially open new possibilities for various photonic applications, such as optical sensing and all-optical signal processing.

2. Device design and fabrication

We used the same dimensions of the slot waveguides and strip-to-slot waveguide couplers as in [17]. The ring radius was 50 μm and the gap between the bus waveguide and the ring was varied from 200 nm to 500 nm. Ring resonators with a drop port were fabricated for demonstrating the feasibility of add-drop filters with the nanolaminate slot waveguides. Similar ring resonators without a drop port were also fabricated for estimation of the propagation losses in the nanolaminate waveguides.
2.1. Waveguide fabrication

The samples were fabricated onto an 8-inch SOI wafer with a 220 nm thick silicon device layer on top of a 2 μm thick buried oxide. The patterning of the silicon was done with 248 nm deep ultraviolet (DUV) lithography. Inductively coupled plasma (ICP) reactive ion etching (RIE) with Cl₂/HBr chemistry was used to etch the silicon. Inverse tapers were patterned to the waveguide ends for fiber coupling. In addition, a few nanometers thick, wet etch stopping layer of amorphous silicon was deposited on top of the device. Finally the whole structure was covered with a 1 μm thick layer of PECVD SiO₂ for upper cladding of the input and output strip waveguides. The areas where the ring resonators are located were exposed to ambient by wet etching the PECVD SiO₂, before the ALD layer was deposited. A deep trench etching process was used to produce smooth vertical facets and ensure enough space for lateral fiber access at the coupling interface [38]. The experimental dimensions of slot waveguides was measured from the scanning electron microscope (SEM) images. Figure 1 illustrates typical SEM images of one of the fabricated ring resonators with measured dimensions. We examined slot waveguides with rail widths (w_r) of 195 nm and 175 nm, and slot widths (w_s) of 220 and 240 nm.

![Fig. 1. SEM images of a fabricated device. a)-c) before the ALD and d) after ALD of Ta₂O₅/PI nanolaminate. e) A schematic illustration of the nanolaminate structure. Inset in d): enhanced image of the nanolaminate film. a)-c) are taken in a tilted angle.](image-url)
2.2. Atomic layer deposition of nanolaminates

The samples were covered with an ALD grown nanolaminate consisting of alternating layers of Ta$_2$O$_5$ and PI. The total number of layers in the nanolaminate structure is 12 (6 × Ta$_2$O$_5$ and 6 × PI). Two of the topmost Ta$_2$O$_5$ layers are doubled, which can be seen from a schematic illustration of the nanolaminate structure shown in Fig. 1(e). An SEM image of a slot waveguide cross-section after ALD is illustrated in Fig. 1(d). Only 5 PI layers are visible in the SEM image because the PI layers appear dark in the image, thus the first layer cannot be seen. The total nanolaminate thickness measured from the SEM image is 105 nm, which would make one individual layer 7.5 nm thick (15 nm for the double Ta$_2$O$_5$ layers). However, as noted in the earlier work on ALD of Ta$_2$O$_5$/PI nanolaminate, the growth rates can be slightly reduced in the nanolaminate process, especially for the polyimide layers [12]. We estimate that the actual layer thicknesses for Ta$_2$O$_5$ and polyimide are 8 nm and 7 nm, respectively. This would make the total nanolaminate layer 106 nm thick ($4 \times 8 \text{ nm} + 2 \times 16 \text{ nm} + 6 \times 7 \text{ nm} = 106 \text{ nm}$), which is supported by the SEM images.

For the nanolaminates studied in this work, the growth rate was 0.21 nm/min. It must be noted, however, that in this work no attempts were made to optimize the growth rate. Therefore, faster growth rates can probably be achieved with shorter cycle times. The nanolaminates were deposited in a hot wall, flow type F120 ALD reactor (ASM Microchemistry Ltd., Finland) at 170 °C under a nitrogen pressure of about 10 mbar. Pyromellitic dianhydride (PMDA) and dianinohexane (DAH) were used as precursors for polyimide, and tantalum ethoxide and water for Ta$_2$O$_5$. PMDA, DAH and tantalum ethoxide were evaporated in open boats inside the reactor at 150 °C, 35 °C and 95 °C, respectively, and water at room temperature in an external source. High-purity nitrogen was used as the carrier and purging gas.

2.3. Mode properties

The electric field distributions in the slot waveguide and mode effective indices were calculated with the Finite Element Method (FEM) using commercial software COMSOL multiphysics. Figure 2 illustrates the intensity distributions of the quasi-TE and quasi-TM modes for waveguides coated by an ALD-Ta$_2$O$_5$/PI nanolaminate. The refractive indices used were $n_\text{Si} = 3.48$, $n_\text{SiO}_2 = 1.45$, [17], $n_\text{Ta}_2\text{O}_5 = 1.884$ and $n_\text{PI} = 1.615$ at $\lambda = 1550 \text{ nm}$. The refractive indices of Ta$_2$O$_5$ and PI were determined with ellipsometer measurements. Film thicknesses for Ta$_2$O$_5$ and PI used in the simulations are 8 nm and 7 nm, respectively.

In addition, the electric field distributions were also simulated for slot waveguides completely filled with PI, Ta$_2$O$_5$ and with a material having the refractive index of mean value of $n_\text{Ta}_2\text{O}_5$ and $n_\text{PI}$ (Fig. 3 and Table 1). Figure 3 shows that in the nanolaminate structure the electric field is enhanced in the PI layers across the whole slot, due to multiple discontinuities at the PI/Ta$_2$O$_5$ interfaces. As can be seen from the Table 1, the effective indices of quasi-TE modes in the nanolaminate and the average material are almost the same but clearly different compared to modes in PI or Ta$_2$O$_5$. This shows that the electric field can be enhanced in the low refractive index material using the nanolaminate coating, while controlling the effective index of the mode.

| Slot material | $n_{\text{eff}}$ |
|---------------|-----------------|
| PI            | 1.56            |
| Ta$_2$O$_5$   | 1.73            |
| Average       | 1.64            |
| Nanolaminate  | 1.65            |
Fig. 2. Simulated mode profiles of the TE (a) and TM (b) modes at 1550 nm. c) and d) normalized x and y- components of the electric field along dashed lines in a) and b). Slot mode characteristics can also be seen in the electric field distribution of the TM mode. Waveguide dimension used in the simulation are \(w_r = 195\) nm and \(w_s = 240\) nm.

Fig. 3. Electric field distribution with different slot materials. The graphs are normalized so that the maximum electric field intensity in the slot with polyimide film equals one.
3. Experiments and results

The waveguide characterization was done using a superluminescent LED source with a center wavelength of 1560 nm, a 3-dB bandwidth of 100 nm and a maximum output power of 26 mW. The input light was coupled to the chip with a tapered polarization maintaining (PM) fiber with a spot size of 2 μm. Output light was coupled to a similar tapered PM fiber and the transmission spectrum was collected with an Anritsu MS9740A optical spectrum analyzer (OSA). Measured transmission spectra of a device with dimensions $w_r = 195$ nm, $w_s = 220$ nm and $g = 500$ nm are shown in Figs. 4(a) and 4(c) for TE- and TM-polarization, respectively. Free Spectral Ranges (FSR) were determined from the transmission measurements to solve the group indices for TE and TM modes. The group index $n_g$ is related to the FSR of the ring resonator with equation

$$\text{FSR} = \frac{\lambda^2}{n_g 2\pi R}, \quad (1)$$

where $\lambda$ is the resonance wavelength and $R$ is the radius of the ring [39]. Measured and simulated group index values for both TE and TM modes are presented in Fig. 5. Simulated group indices are obtained by solving the effective indices ($n_{\text{eff}}$) of the modes at different wavelengths and applying the equation

$$n_g = n_{\text{eff}} - \lambda_0 \frac{dn_{\text{eff}}}{d\lambda}. \quad (2)$$

![Fig. 4. Measured transmission spectra of an ALD-nanolaminate ring resonator with waveguide dimensions $w_r = 195$ nm, $w_s = 220$ nm and $g = 500$ nm. a) TE polarization. b) Enlarged view of the resonance at 1528.7 nm yielding a Q-factor of 6709. c) TM polarization d) Enlarged view of the resonance close to 1544 nm yielding a Q-factor of 4082. Blue: through port transmission, red: drop port transmission.](image)

Summary of the measured and simulated FSR values for all ring resonators investigated in this work is shown in Table 2. The simulated values were obtained from the group index with Eq. (1) and the measured values were calculated from the peaks close to 1550 nm.
Fig. 5. Measured and simulated values of the group index of a ring resonator with dimensions \( w_r = 195 \) nm, \( w_s = 220 \) nm and \( g = 500 \) nm.

As can be seen from Fig. 5, the TE and TM modes are clearly distinguishable. The quasi-TM mode has a higher group index than the quasi-TE mode, which is mostly localized in the slot. Moreover, the very good agreement between the simulations and the measurements proves that the nanolaminate coating operates as expected. This result supports also our estimation of the thickness of the Ta_2O_5 and PI layers from the SEM images. The resolution of our OSA limits the minimum measurable change in the FSR to 0.05 nm, which causes the sawtooth-like variation of \( n_g \) values in the case of quasi-TM mode.

Table 2. Comparison of measured and simulated values of FSR (at 1550 nm) and typical measured quality factors (Q) and extinction ratios (ER).

| \( w_r \) (nm) | \( w_s \) (nm) | FSR\_meas. [nm] | FSR\_sim. [nm] | Q | ER [dB] |
|----------------|----------------|----------------|----------------|---|--------|
| **TE**         |                |                |                |   |        |
| 195            | 240            | 3.1            | 3.05           | 2 400 | 15.8   |
| 175            | 220            | 2.9            | 2.97           | 6 000 | 4.7    |
| **TM**         |                |                |                |   |        |
| 195            | 240            | 2.9            | 2.95           | 1 900 | 17.8   |
| 175            | 220            | 2.7            | 2.84           | 3 800 | 8.7    |

The measured resonance peaks were fitted with a Lorentzian in order to determine more precisely the quality factors of the fabricated ring resonators, by using an equation

\[
Q = \frac{\lambda_{\text{res}}}{\text{FWHM}},
\]

where \( \lambda_{\text{res}} \) is the wavelength of the resonance and FWHM is the full width at half maximum of the peaks [39]. Measured Q-factors and extinction ratios for ring resonators with different slot and rail dimensions are shown in Table 2.

3.1. Estimation of propagation losses

Propagation losses in the nanolaminate coated slot waveguides were estimated by following the fitting method presented in [40]. The amplitude transmission from a ring resonator without a
Drop port can be expressed with Eq. (4),

\[ T = \frac{a^2 - 2ar\cos\phi + r^2}{1 - 2ar\cos\phi + (ra)^2}, \]  

where \( \phi = 4\pi^2n_{\text{eff}}R/\lambda \) is the phase shift per round trip, \( a \) is the amplitude transmission coefficient \((a^2 = \exp(-\alpha^22\pi R))\) and \( r \) is the self-coupling coefficient [39]. Coefficients \( a \) and \( r \) were determined by fitting the Eq. (4) into the measured transmission spectra of a ring resonator without a drop port. The effective indices used in the fit were obtained from FEM simulations at wavelengths corresponding to each resonance. The propagation losses can be calculated with the Eq. (5) below. Because the Eq. (4) is commutative with \( a \) and \( r \), these coefficients can not be distinguished directly from the fit [39]. However, they can be distinguished by examining how they behave as a function of the coupling distance between the bus waveguide and the ring [41]. The attenuation in the ring is not expected to depend on the coupling distance and \( r \) is expected to increase as the coupling distance is increased. This is because smaller portion of the propagating mode is coupled to the ring when the gap is larger. We performed the same analysis for similar rings with coupling distances of 200, 300 and 400 nm and could clearly distinguish which one of the coefficients increases with the gap. This is illustrated in the Fig. 6(c).

![Fig. 6.](image)

Fig. 6. a) Measured transmission spectrum around the resonance at 1550 nm and corresponding dip obtained by fitting the measured data to Eq. (4). The parameters obtained from the fit are \( a = 0.9251 \) and \( r = 0.6813 \). Calculated propagation loss and intrinsic quality factor for the resonance in a) are \( \alpha = 21.5 \) dB/cm, \( Q_i = 13 \) 509, respectively. b) Measured transmission spectrum (quasi-TE mode) of all-pass ring resonator with dimensions: \( w_r = 195 \) nm and \( w_t = 240 \). c) Coefficients \( a \) and \( r \) for ring resonators with gaps of 200, 300 and 400 nm. d) Histogram of calculated propagation losses from all resonances in the spectrum displayed in b).
The propagation loss is calculated from the equation
\[
\alpha = -\frac{2 \times \ln a}{2\pi R} \times 10 \times \log e = -\frac{2 \times 10 \times \log_{10} a}{2\pi R} \quad \text{[dB/cm].}
\]  
(5)

To obtain propagation loss in dB/cm, the radius needs to be inserted in cm into Eq. (5). The intrinsic quality factor can be calculated from \( a \) using the equation [42],
\[
Q_i = \frac{4\pi^2 R_{\text{eff}}}{|2\ln a|\lambda_0}.
\]  
(6)

The highest quality factors were obtained for \( w_s = 240 \) nm and \( w_r = 195 \) nm. Measured transmission spectrum and fit to a resonance close 1550 nm are illustrated in Figs. 6(b) and 6(a), respectively. Figure 6(d) shows a histogram of calculated propagation loss values from all the resonances from the transmission spectrum in Fig. 6(b). The highest obtained intrinsic Q-factor was 14 000 and the lowest propagation loss was 19.3 dB/cm. The average calculated propagation loss was 29.5 ± 5.1 dB/cm, which is higher than previously reported values of propagation loss for ALD coated slot waveguides. In [17] propagation losses of 5.2 ± 0.3 dB/cm were measured by a cut-back method for ALD-Al\(_2\)O\(_3\) coated slot waveguides fabricated with the same technique as the slot waveguides examined in this work. For comparison we also fabricated similar slot waveguide ring resonators with 160 nm of ALD-Al\(_2\)O\(_3\) and calculated the propagation losses with the same fitting method. We obtained a propagation loss of 20 dB/cm, which is also higher than 5.2 ± 0.3 dB/cm reported in [17]. The only difference between the waveguides examined here and in [17] is the amorphous silicon wet etch stopping layer, which can be a reason for the higher losses. These results suggest that the real propagation losses of Ta\(_2\)O\(_5\)/PI nanolaminate slot waveguides can actually be lower than the calculated values. Furthermore the fitting method that was used for obtaining the propagation loss is quite sensitive for small features in the measured spectra. Also the bends in the ring can cause bending loss, even though we used relatively large ring radius of 50 \( \mu \)m.

4. Discussion and conclusions

In this work, we have designed and experimentally demonstrated organic/inorganic nanolaminate coated silicon slot waveguide ring resonators fabricated with CMOS compatible processes; 248 nm DUV lithography and ALD. To our best knowledge, this is the first time when ALD of organic materials is demonstrated in waveguiding applications. In 248 nm DUV lithography used in this work, the minimum achievable slot width is around 180 nm, which is too wide for air slots. However, with ALD nanolaminates the mode can be squeezed into several sub-10 nm vertical slots in addition to the central air slot. Furthermore, the effective index is determined by the layer thicknesses, which means that after selecting the materials for the nanolaminate, the effective index can be tuned between the refractive indices of the two materials. We achieved a good fit between experimental values and simulated group indices, confirming that the light is indeed localized in the slot for TE-polarization. We obtained an intrinsic Q-factor of 14 000 and the lowest propagation loss was 19.3 dB/cm, which shows that our devices are fully functional. Our earlier results indicate that there is potential for even lower propagation loss and thus higher quality factors. The nanolaminate structure demonstrated here gives extra degrees of freedom for engineering the mode profile of slot waveguides for different applications. Different organic and inorganic materials and layer thicknesses can be combined to create multitude of different nanolaminate structures.
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