Low-Stress Silicon Nitride Platform for Mid-Infrared Broadband and Monolithically Integrated Microphotonics

Pao Tai Lin,* Vivek Singh, Hao-Yu Greg Lin, Tom Tiwald, Lionel C. Kimerling, and Anuradha Murthy Agarwal

Implementation of mid-infrared (mid-IR) chip-scale microphotonic circuits is critical to advancing the science for applications such as (i) ultra-fast telecommunications that require wider bandwidth and (ii) integrated biochemical sensors which can fingerprint using infrared absorption signatures. Though substantial progress has been made in the development of light sources and detectors, a monolithic mid-IR Si-CMOS-compatible platform remains a challenge. Here we experimentally demonstrate a sophisticated mid-IR microphotonics platform adopting engineered Si-rich and low-stress silicon nitride (SiNx) thin films where an extensive infrared transparency up to $\lambda = 8.5 \mu m$ is achieved. Furthermore, because of the designed low-stress property, the SiNx deposition is able to reach a thickness > 2 $\mu m$ that significantly reduces mid-IR waveguide loss to less than 0.2 dB/cm. We show directional couplers functioning over a broad infrared spectrum, thus enabling monolithic mid-IR multiplexing schemes for integrated linear and nonlinear photonics leading to sophisticated label-free sensing.

1. Introduction

Photonic integrated circuits have shown revolutionary growth in past decades and one critical factor that promotes their success is the fast development of near-IR (NIR) materials in the optical communication band. However, there is an urgent demand to move current microphotonic devices beyond $\lambda = 2.5 \mu m$ due to the lack of availability of suitable materials. While conventional Si waveguides have a high optical loss, high optical nonlinearity, mechanical robustness and strong chemical stability. Moreover, SiNx is known to be fully compatible with VLSI processes, as well as capable of being deposited on a variety of substrates; both make it a versatile potential candidate for a universal mid-IR platform. Recently, several studies have evaluated SiNx waveguides for long-wavelength light generation, however spectra do not exist beyond $\lambda = 2.5 \mu m$ due to the lack of availability of suitable optical spectrum analyzers. Also, a big hurdle to fabricating SiNx devices for applications beyond $\lambda = 2.4 \mu m$ comes from the difficulty of depositing thicker crack-free films. In other words, to achieve low propagation loss in the mid-IR ($\lambda > 2.5 \mu m$), waveguides of micron-level thicknesses are essential so that the waveguide is fully confined inside the waveguide. However, typical stoichiometric silicon nitride, SiNx, has a high tensile stress > 1000 MPa. As a result, its thickness cannot easily exceed 250 nm otherwise cracking can ensue.

To overcome this limitation of film thickness, we engineer the chemical composition ratio between Si and N in order to reduce its mechanical stress. In this letter, we demonstrate an optimized SiNx film that has a tensile stress as low as 45 MPa which enables a crack-free deposition thickness of 2.5 $\mu m$. To evaluate the compositional uniformity, energy-dispersive X-ray spectroscopy (EDX) and X-ray photoelectron spectroscopy (XPS) are utilized in both surface mapping and depth scanning modes. The optical properties up to $\lambda = 10 \mu m$ are investigated by very-large-scale integration (VLSI) processes. Current mid-IR ($\lambda > 2.5 \mu m$) platforms that have been evaluated in planar devices include chalcogenide glass thin films, silicon-on-insulator (SOI), pedestal Si waveguides, and Ge on Si. Though chalcogenide glass thin films have a wide mid-IR transparency window, they have yet to become fully compatible with VLSI processes. A Si mid-IR platform requires single crystalline Si to reduce optical scattering losses, and this requirement can be restrictive. Furthermore, preparation of SOI wafers or pedestal Si waveguides require delicate fabrication processes which translate to high costs. Likewise, for a low-loss Ge waveguide, a monocrystalline Ge thin film is required. On the other hand, SiNx films are known for their excellent performance in NIR microphotonics owing to their advantages of extremely low optical loss, high optical nonlinearity, mechanical robustness and strong chemical stability. Moreover, SiNx is proven to be fully compatible with VLSI processes, as well as capable of being deposited on a variety of substrates; both make it a versatile potential candidate for a universal mid-IR platform.
2. Results and Discussion

Among the various SiNₓ candidates, we select silicon-rich films because they possess superior properties of low mechanical stress, tunable refractive index, and uniform film composition, which meet the requirements for mid-IR waveguides. A sufficiently thick (4 μm) undercladding SiO₂ layer with lower refractive index (n_SiO₂ ∼ 1.45) is obtained by thermal oxidation prior to the growth of SiNₓ thin films to prevent leakage to the higher refractive-index (n_Si∼3.43) silicon substrate. Detailed recipes for the SiNₓ and SiO₂ layers are discussed in the Experimental Section.

Chemical composition and morphology of the SiNₓ film and SiO₂ undercladding layer are characterized by a scanning electron microscope (SEM) equipped with EDX. Figure 1(a) is the cross-sectional SEM image in which sharp and smooth interfaces between (i) SiNₓ film and SiO₂ layer, and (ii) SiO₂ layer and Si substrate, are shown resolved. Smooth core-cladding interfaces are required for low loss optical waveguides; an uneven surface and nano particles can easily cause unwanted scattering loss. In the SEM of Figure 1(a), bright strips are seen in the top layer from dielectric film charging during electron beam scanning. Hence, to better resolve the film structure and analyze the compositional uniformity, EDX is utilized because it is capable of providing a spatial elemental distribution. Figure 1(b),(c),(d) are images from EDX mapping at 1.74 keV, 0.393 keV, and 0.525 keV that correspond to the element emission lines of Si Kα₁, N Kα₂, and O Kα, respectively. Areas revealing high concentrations of N and O correspond to where films are deposited. Thus, the thickness of SiNₓ and SiO₂ can be accurately determined as 4 μm and 2.5 μm, respectively. In addition, from Figure 1(c) the element N has a uniform distribution within the SiNₓ layer showing that the SiNₓ film retains a constant stoichiometry through the growth process.

To quantitatively resolve the stoichiometry and the compositional uniformity of the deposited SiNₓ film, XPS analysis is performed in both vertical (normal-to-plane) and lateral (in-plane) directions. For the vertical analysis, the photoelectron binding energy (B.E.) at each depth in the profile is obtained as the film surface is gradually removed by sputtering. To achieve a high resolution depth profile, a slow sputtering rate is used in the first 5000 s of etching time. As depicted in Figure 2(a), B.E. peaks are found at 105.8 eV and 400 eV, which belong to characteristic signals of silicon 2p and nitrogen 1s respectively. An atomic ratio of Si:N of 55:45 is obtained, which indicates that our low stress (45 MPa) SiNₓ is a silicon-rich silicon nitride film and an increase of N ratio will cause undesired tensile stress. In addition, the chemical composition of the deposited SiNₓ film is highly uniform along the vertical direction because the peak counts from the B.E. scan remain constant during the sputtering period. To investigate the compositional profiles of the SiO₂ undercladding and the substrate, sputtering with higher etching rates is utilized and the results are shown in Figure 2(b). Similar to the results from the upper SiNₓ film, a constant Si:N ratio of 55:45 is observed in the rest of the SiNₓ layer. At what we believe to be the interface between the SiNₓ core and the SiO₂ undercladding (etching time of 12 500 s), a new B.E. peak at 532 eV (oxygen 1s) appears and simultaneously the signal at 400 eV (nitrogen 1s) drops. Another variation of signal is observed at 24 000 s, where the signal at 532 eV (oxygen) disappears and only a peak at 105.8 eV (Si) lasts; indicating that the depth profiling has reached the Si substrate. We should also note that a consistent in-plane Si:N ratio is critical to realizing low loss mid-IR devices since any abrupt change of the composition ratio will cause deviations in the SiNₓ refractive index and cause severe optical scattering. Thus, high resolution element morphology is investigated using an XPS surface scan where the B.E. is chosen at 92.58 eV for Si 2p bonding and 390.08 eV for N 1s bonding. From the results shown in Figure 2(c), the elemental concentration of Si and N is highly uniform within the scanned area (4 mm × 4 mm). The average variation of Si:N atomic ratio is less than 2%, which is already close to the XPS detection limit, and so this will not affect the refractive indices which are determined by the Si:N ratio. Overall, by means of the layer-to-layer B.E. scanning and surface mapping, we confirm a low-stress thick SiNₓ film with exceptional homogeneity of Si:N ratio. Implementation of low stress deposition method can prevent undesired cracking or stress-induced compositional or optical variation.

The structures of the fabricated mid-IR devices are then inspected by optical microscope and SEM. Figure 3(a) shows
the optical images of our “paperclip-shaped” waveguides used for optical loss measurements. The top cladding is air and is the bottom cladding is oxide. The waveguide bending radius is 200 μm and the variable portion of the straight part of the waveguide, \( D \), changes from 0 mm to 1 mm, 2 mm, 3 mm, 4 mm, and 5 mm. To characterize the height, \( h \), and width, \( w \), an SEM image of the waveguide is captured at a tilt angle of 54° as shown in Figure 3(b). A height \( h = 4 \) μm and a width \( w = 2.5 \) μm are measured, and the waveguide reveals a smooth surface and sharp edges without bumps or indentations.

The mid-IR directional coupler is displayed in Figure 3(c). Two separate waveguides are merged into a coupling section with a coupling length of \( L \). From the SEM image shown in Figure 3(d), the gap \( g \) between the adjacent waveguides is 700 nm where no fusion is observed over the entire coupler. The sharp gap edge and the constant gap width explain the well-resolved mid-IR light coupling observed.

The optical properties, including both the index of refraction \( n \) and extinction coefficient (imaginary refractive index) \( k \), of our SiN\(_x\) film are characterized by IR-VASE, a technique that measures and analyzes the polarization change from the reflected mid-IR light.\(^{[26]}\) Since the optical constants of SiN\(_x\) films depend on the deposition technique and condition, knowing the optical constants is critical in order to optimize the performance of our mid-IR devices.\(^{[27–29]}\) The \( n \) and \( k \) plots of the SiN\(_x\) film are shown in Figure 4, where a comprehensive characterization is accomplished from the NIR to the mid-IR, showing that \( n \) decreases slowly from 2.1 at \( \lambda = 1 \) μm to 2.0 at \( \lambda = 4 \) μm before a strong dispersion is found after \( \lambda = 7 \) μm. The almost constant \( n \) over a broad mid-IR spectral range offers the advantage of low optical dispersion desired for many planar microphotonic devices, including nonlinear light generation and chip-scale biochemical sensors. Another merit is that only a trivial extinction coefficient \( k < 10^{-4} \) is observed before \( \lambda = 8 \) μm; the rise of \( k \) here is due to the Si-N stretching absorption. Furthermore, the absorptions from N-H stretch at \( \lambda = 3 \) μm and Si-H stretch at \( \lambda = 4.5 \) μm are barely found because our LPCVD has a high precursor ratio between dichlorosilane and ammonia, with ammonia being the hydrogen source.\(^{[30]}\) As a result, the hydrogen concentration is minimized in our Si-rich SiN\(_x\) and eventually prevents the absorption...
attributed to H bonds. Thus, we are able to demonstrate a SiNx film revealing high transparency all the way from NIR to the mid-IR region (λ = 1–8 μm). With the benefit of low-dispersion and optical loss, our low-stress Si-rich SiNx provides a unique platform for broad mid-IR integrated circuits.

To evaluate the performance of our devices, a mid-IR optical bench is set up and the details are addressed in the method section. Using this system we are able to image the waveguide mode and record the output powers at different mid-IR wavelengths. Figure 5(a) illustrates the captured waveguide intensity profile, which shows a sharp round spot defining the fundamental mode as the dominant waveguide mode. The optical losses of the waveguides are then characterized by fitting the variation of the optical output powers from different propagation lengths. The results from the loss measurements are shown in Figure 5(b) where a loss of 0.16 dB/cm at λ = 2.65 μm is obtained. As a comparison, previous studies report SiNx waveguides prepared by thermal cycling processes have a propagation loss of 0.5 dB/cm, whereas other waveguides composed of low-hydrogen SiNx films from low temperature (350 °C) PECVD method have an optical loss of 2.1 dB/cm.[31,32] Clearly, our low stress SiNx waveguides exhibit the lowest mid-IR propagation loss among a variety of SiNx films.

To demonstrate that our low-stress SiNx waveguide can be used as a multiport component for mid-IR microphotonics, directional couplers are fabricated and their spectral responses are recorded and shown in Figure 6(a). When the input light wavelength is λ = 2.45 μm, a strong guided mode appears in the left output (channel 1) and only a dim signal is observed at the right output (channel 2). As the wavelength increases, the intensity of the left guided mode gradually decreases whereas the light from the right output increases progressively. Once the wavelength reaches λ = 2.55 μm the power transmitted through both channels is equal. Eventually, the guided wave in the

Figure 3. (a) Optical images of the “paperclip” waveguides. D is the length of the variable portion of the straight section of the paperclip waveguide. (b) SEM image of the waveguide captured at a tilt angle of 54°. h and w are the waveguide height (4 μm) and width (2.5 μm), respectively. (c) Optical microscope and SEM images at one end of a mid-IR directional coupler. The yellow box highlights the coupling section that is shown in (d). (d) Magnified SEM image of the mid-IR directional coupler. The coupling gap, g = 700 nm, is highlighted by the red bar.

Figure 4. The n and k plots of SiNx sample from IR-VASE measurement. The refractive index n has low dispersion up to λ = 6 μm, and negligible absorption is found before λ = 8 μm. The increase of attenuation coefficient (imaginary refractive index) k after λ = 9 μm is due to the Si-N stretching.

Figure 5. (a) Captured mid-IR image of SiNx, waveguide output at λ = 2.65 μm. A fundamental mode emerging from the waveguide is resolved. (b) The relative optical power from different waveguide lengths. Fitting the variation (red curve) gives a propagation loss of 0.16 dB/cm.
right arm becomes the dominant mode at $\lambda = 2.65 \, \mu m$. These images show a visible demonstration of our mid-IR directional coupler. To quantitatively analyze the spectral dependence of coupling, intensity profiles as a function of wavelength are shown in Figure 6(b). At each wavelength two peaks appear that represent the light output from channels 1 and 2. As the input lightwave moves to longer wavelengths, the peak intensities of channel 1 fall whereas the output from channel 2 increases and an extinction ratio of 7 dB is obtained. We also note that the guided modes from both channels retain sharp Gaussian profiles over the entire spectrum, confirming that the fundamental modes are well preserved during coupling. This preservation of the fundamental mode is critical for many applications, including mid-IR sensing and nonlinear light generation, because a higher order mode is associated with a different refractive index leading to undesired dispersion which can (a) lead to a false positive sensor signal during mid-IR detection and (b) lower the efficiency of light generation. In addition, we found the total power from two channels remains constant during the lightwave coupling proving that the fabricated mid-IR directional coupler achieves a high switching efficiency with minimal optical loss.

To design a low-loss and broadband mid-IR $\text{SiN}_x$ waveguide, simulations of waveguide modes are performed using two dimensional finite difference method (FDM) calculations. Figure 7(a) is the 3D structure used in the calculations, where the layout is built based on the structure obtained from our SEM characterization shown in Figure 3. Here, the waveguide has a height $h = 4 \, \mu m$ and a width $w = 2.5 \, \mu m$. Underneath the $\text{SiN}_x$, is a 4 $\mu m$ thick buffer oxide serving as an optical undercladding that prevents substrate leakage. The simulated refractive index profile along the waveguide cross-section plane (x-z plane) is shown in Figure 7(b). Index of $\text{SiN}_x$, $n_{\text{SiN}_x}$ is 2.05 obtained from ellipsometry measurements at $\lambda = 2.55 \, \mu m$. The calculated 2D mode images at $\lambda = 2.75 \, \mu m$ is depicted in Figure 7(c) where the calculated mode index is 1.96. Clearly a fundamental mode is obtained at both mid-IR wavelengths and it is well confined to the $\text{SiN}_x$ waveguide in both lateral (x) and vertical (y) directions. The intensity of the evanescent wave at the undercladding layer ($z < 0 \, \mu m$) slightly increases as wavelength increases. To better visualize the mode shape, the associated calculated 1D intensity profile is plotted in Figure 7(d). Our simulations predict that only a single Gaussian peak should be found, and so it agrees well with our experimental results showing that the fundamental mode is the dominant waveguide mode.

Performance of the directional couplers is calculated by finite-difference time-domain (FDTD) methods. The layout of the simulation is shown in Figure 8(a) which is based on the captured SEM images shown in Figure 3. Here, channel 1 and channel 2 represent the two individual waveguides within one coupler. These two waveguides are identical, where the structure parameters including width $w$, height $h$, and length $l$, are set to 4 $\mu m$, 2.5 $\mu m$ and 8 mm, respectively. The main factor that determines the coupling efficiency is the separation between channels, $d$, which is 0.7 $\mu m$ in this case. Figure 8(b) is the intensity distribution between channel 1 and channel 2 calculated from $\lambda = 2.4 \, \mu m$ to $\lambda = 2.8 \, \mu m$. Clearly the light output alternates as...
we increase the simulated wavelength to $\lambda = 2.65 \, \mu m$ we find the mid-IR light switches almost completely to channel 2. Furthermore, we find that the guided mid-IR light retains its “fundamental mode” nature (single sharp spot), even after several rounds of coupling and this agrees well with our experimental results shown in Figure 6. Both simulation and experiment shows that the maximum intensity of channel 1 (input) appears at $\lambda = 2.45 \, \mu m$ and its minimum occurs at $\lambda = 2.65 \, \mu m$. Thus, we confirm that our design is optimized for mid-IR directional couplers and can potentially be used in mid-IR wavelength-division multiplexing (WDM).

3. Conclusion

We demonstrate Si-CMOS-compatible mid-IR microphotonics platform using specially engineered SiN$_x$ thin films. The prepared SiN$_x$ film has a wide infrared transparency covering $\lambda = 1 \, \mu m$ up to $\lambda = 8.5 \, \mu m$, along with a low optical dispersion as seen from IR-VASE characterization. Because of its low mechanical stress (<45 MPa), the SiN$_x$ film can reach microns in thickness optimized for mid-IR devices while still remaining crack-free with a homogeneous chemical composition over the entire 4 mm x 4 mm chip. Using the developed SiN$_x$ we are able to demonstrate (a) a waveguide that achieves a remarkably low optical-loss (<0.2 dB/cm) as well as (b) a working broadband mid-IR directional coupler. Thus, we provide a new monolithic mid-IR platform that will accelerate the development of high capacity telecommunication as well as on-chip infrared sensors.

4. Experimental Section

Preparation of SiO$_2$ Buffer Layers and Low Stress SiN$_x$ Thin Films: The buffer SiO$_2$ layer is prepared by wet oxidation, in which the gases used are H$_2$/O$_2$ and a pyrogenic torch with a flow rate of 3000 sccm. The oxide film is grown under atmospheric pressure and the temperature is set at 1100 °C. A deposition rate of 9 nm/min is obtained. The low stress SiN$_x$ films are prepared by LPCVD. Before loading the SiO$_2$ on the 4 inch Si substrates in the LPCVD furnace, they are cleaned using a standard Piranha solution, a mixture of sulfuric acid (H$_2$SO$_4$) and 30% hydrogen peroxide (H$_2$O$_2$) with volume ratio of 3:1, in order to remove any organic residues. For SiN$_x$ film growth, the silicon source is dichlorosilane (DCS) and the nitrogen source is ammonia NH$_3$. A high DCS:NH$_3$ gas ratio of 3:1 is chosen in order to produce Si-rich SiN$_x$ films. The deposition pressure is set at 200 mTorr and the reactor temperature is 825 °C. After deposition, the first cool down time from 835 °C to 550 °C is 100 minutes and the second cool down time from 550 °C to room temperature is 20 min. A deposition rate of 10 nm/ min is determined from film thickness characterization.
Device Fabrication Process: To fabricate the mid-IR planar devices on the Si-rich SiN\textsubscript{x} film, conventional photolithography and inductively coupled plasma reactive ion etching (ICP-RIE) are utilized. Hexamethyldisilazane (HMDS) and a micron thick photoresist (Shipley 8131) are initially coated on the SiN\textsubscript{x} film with a speed of 4000 revolutions per minute (rpm), and the coated wafer is baked at 115 °C for 1 minute. Desired layouts including waveguides, splitters and couplers are defined by the photo-mask through UV patterning and then developed using MF-319 solution. These structures are transferred into the SiN\textsubscript{x} layer through an optimized ICP-RIE etching process of 15 minutes in Ar/H\textsubscript{2}/CHF\textsubscript{3}/CF\textsubscript{4} with a flow rates of 6/30/50/2 sccm respectively.

XPS Characterization: XPS is performed by K-Alpha XPS system from Thermo Scientific. The probe for measurement is aluminum K-alpha X-ray line with energy at 1.4866 keV and the X-ray spot size is 400 μm. A flood gun, which supplies low energy electrons and ions is used throughout the entire experiment for sample surface charge compensation. Depth profiling is used to examine the silicon nitride film homogeneity with the assistance of a built-in argon sputtering gun that has 3000 eV acceleration energy with raster size at 2 mm. Both survey spectra and high resolution scan data are collected for each depth monitored. For survey spectra, each scan is an average of 5 snapshots with pass energy at 200 eV, from –10 eV to 1350 eV B.E. For high resolution scan, snapshot scan mode was done by taking 5 snapshots with pass energy at 200 eV, from –10 eV to 1350 eV B.E. For high resolution scan, snapshot scan mode was done by taking 5 snapshots with pass energy at 200 eV, from –10 eV to 1350 eV B.E.

IR-VASE Characterization: Two instruments were used to acquire the spectroscopic ellipsometry data. Approximately 1050 wavelengths from 0.21 to 1.7 μm were acquired with a dual rotating compensator ellipsometer with a multichannel CCD spectrometer. An FTIR-based single rotating compensator ellipsometer acquired 1260 wavelengths from 1.7 to 30 μm at 8 cm\textsuperscript{-1} resolution. Each instrument acquired data at several incidence angles between 55° to 75°. All data sets and incidence angles were simultaneously fit to a SiN\textsubscript{x}/SiO\textsubscript{2}/Si model, resulting in a single SiN\textsubscript{x}/n+ik function for all wavelengths. This multiple-sample analysis method allows for variations between the wavelength ranges and incidence angles, as well as depolarization effects caused by variations in thickness and spectrometer bandwidth.\textsuperscript{[33]}

Mid-Infrared Measurement System: Figure 9 illustrates the experimental set-up used to evaluate the mid-IR SiN\textsubscript{x} microphotonic platform. The light source is a pulsed laser tunable between λ = 2.4 μm and λ = 3.8 μm and its average power is 150 mW. The probe light is first collimated into a single mode fluoride fiber and then butt-coupled into the mid-IR waveguide. The alignment between the optical fiber and the waveguide is monitored by an overhead microscope. The Mid-IR output signals and waveguide mode are recorded by an InSb camera.