Optical nonlinearities in ultra-silicon-rich nitride characterized using z-scan measurements

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The dispersive nonlinear refractive index of ultra-silicon-rich nitride, and its two-photon and three-photon absorption coefficients are measured in the wavelength range between 0.8 μm–1.6 μm, covering the O- to L– telecommunications bands. In the two-photon absorption range, the measured nonlinear coefficients are compared to theoretically calculated values with a simple parabolic band structure. Two-photon absorption is observed to exist only at wavelengths lower than 1.2 μm. The criterion for all-optical switching through the material is investigated and it is shown that ultra-silicon-rich nitride is a good material in the three-photon absorption region, which spans the entire O- to L- telecommunications bands.

Ultra-silicon-rich nitride (USRN), with composition Si7N3, is a promising platform for optical signal processing at the telecommunications wavelengths because of its large nonlinear refractive index and absence of two-photon absorption. The refractive index and band gap of the silicon-rich nitride material can be controlled by the silicon content1. A larger silicon content decreases the energy band gap, towards that of silicon (1.11 eV). In silicon, strong two-photon absorption exists in the entire telecommunications band, which makes it less efficient for non-linear optical processes. The ratio of silicon to nitrogen content can be engineered in order to tailor the bandgap. USRN has been successfully used to demonstrate high gain optical parametric amplification, four-wave mixing and photonic crystal waveguides1–10. A wide band gap reduces not only absorption loss but also the nonlinear refractive index; there exists a tradeoff between high nonlinear refractive index and low absorption loss. This tradeoff arises as a result of Kramers-Krönig (K-K) relations that govern the relationship between nonlinear refractive index and multi-photon absorption. USRN’s larger bandgap allows two-photon absorption to be negligible at the 1.55 μm wavelength. A further advantage of USRN’s larger bandgap pertains to its optical transparency at shorter wavelengths than that in silicon – USRN is optically transparent at wavelengths as low as 0.6 μm compared to 1.1 μm in silicon.

In this paper, we characterize the nonlinear properties of USRN using the z-scan method across a broad range of wavelengths between 0.8 μm to 1.6 μm. We quantify the two- and three-photon absorption coefficients of the USRN platform which possesses a band gap of 2.1 eV4,6 and show that two-photon absorption becomes non-negligible when the wavelength drops below 1.2 μm. Since the USRN films are grown using chemical vapor deposition, the film thicknesses are limited. Consequently, the z-scan measurements will be limited by the measurable intensity dip during the z-scan measurements. Even with these limitations, we were able to characterize the three-photon absorption coefficients up to a wavelength of 1.6 μm.

Results

USRN films with a thickness of 1.1 μm were deposited on a 50 μm thick SiO2 substrate using inductively coupled chemical vapor deposition at relatively low temperature of 250 °C. In the deposition, N2 gas is used in replace of NH3 for chemical reaction in forming Si7N3 to minimize amount of H in the film because Si-H or N-H is a dominant absorption loss bonding at communication wavelength range of 1510–1565 nm11–12. The material composition was previously characterized using FTIR spectroscopy. The FTIR measurements did not pick up the characteristic absorption peak from Si-H bonds close to 1550 nm. Therefore, hydrogen content in the film if present, is below the FTIR detection limit and should be relatively low.

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We employ the z-scan method to characterize the nonlinear coefficient of the USRN; The thin sample thickness places a very high signal to noise ratio requirement to retrieve nonlinear coefficients from very small valley-peak in the measured signal. For this purpose, we formulated a very fast z-scan as shown in Fig. 1 to acquire a large number of averages and in so forth, minimize any contributions to noise. The sample is set at the end of a long arm that is connected to a rotational step motor. The IR ranges of the optical pulses used in the experiment are derived from an optical parametric amplifier (OPA). The OPA is pumped using a Ti:Sapphire laser producing 150 fs pulses at 800 nm at a repetition of 1 kHz. To make many measurements averaged in the same condition, we synchronize the rotational step motor with the laser. Since the sample stage is moving at a specific frequency of 1 Hz and the laser repetition rate is 1 kHz, it is possible to synchronize both frequencies such that all pulses from the laser are incident at the same point on the sample. Without proper synchronization, measurements with each subsequent pulse will have a different z-scan distribution and averaging will not be effective. The laser power has a 10% fluctuation. Consequently, averaging with a large number of measurements (N) is used to reduce the resulting noise. The laser fluctuation scales according to 10%/%N by random walk distribution in which a standard deviation proportional to √N is divided by an average proportional to N. Consequently, the peak-valley difference in the z-scan measurements may be more easily resolved with averaging applied.

Using the aforementioned approach, power fluctuations in the laser (~10%) were successfully reduced to less than 0.3% over a thousand averages. The arm length was designed to be almost 30 cm long to ensure parallel movement and fast scanning with the rotational motor.

The USRN films are grown on a thin SiO₂ substrate (50 μm) to minimize unnecessary effects from the substrate. The n₂ value for SiO₂ is well known to be low (2.7 × 10⁻²⁰ m²/W at 1.55 μm). The effect of the substrate is accounted for in the z-scan measurements. We considered field distortion by multi-films especially two-photon absorption for the simplicity of mathematics. It can be generalized to a higher order of nonlinear absorption. The effect of material on the field going through the sample is described by two coupled equations. \( dΔφ/dz' = 2πn₂I/λ \) and \( dl/dz' = -β₂d² \), where \( z' \) is the sample coordinate, \( Δφ \) is nonlinear phase distortion by the nonlinear sample, \( n₂ \) is the nonlinear refractive index and \( β₂ \) is the two photon absorption coefficient. The field distribution at the sample output can be obtained analytically, \( E_s = E_{in}(z, r, t)(1 + β₂L/L_{0})^{α/β₂—1/2} \). For small multi-photon absorption coefficient approximation, \( β₂L/L_{0} ≪ 1 \), \( E_s = E_{in}(z, r, t)e^{β₂z}L_{0}^{1/2}e^{−β₂L^{1/2}} \). In this approximation, the multi-film effect is additive because the nonlinear coefficients are located the argument of the exponential function. The measured nonlinear coefficients have some contribution of the substrate as follows, \( n₂ + n₂ \left( 1 - \frac{n - n₂}{n + n₂} \right) L'/L + \beta₂ + \beta₂ \left( 1 - \frac{n - n₂}{n + n₂} \right)^2 L'/L \). For n-photon absorption, the expression can be generalized to \( \beta₂ + \beta₂ \left( 1 - \frac{n - n₂}{n + n₂} \right)^2 L'/L \), where the term \( \left( \frac{n - n₂}{n + n₂} \right)^2 \) denotes the Fresnel reflection loss in the substrate.

The measured closed and open aperture z-scan of the thin USRN sample at three representative wavelengths are shown in Fig. 2. At 1.55 μm, the normalized intensity dips by 0.1 – a small value which can still be clearly resolved with the z-scan setup. The nonlinear absorption coefficients are retrieved by fitting the measured open aperture z-scan data with the expression, \( T_{PA} = 1/(1 + (N - 1)α_{0}L_{0}^{(N)}I_{0}/(1 + (z/z_0)^{2}))^{N-1}, \) where \( α_{0} \) is the absorption coefficient, \( N \) is the contributed number of photons, \( I_{0} \) is the peak intensity of the beam at the focal point, \( L_{0}^{(N)} \) is the effective thickness of sample and \( z_0 \) is Rayleigh length. Closed aperture z-scan data is normalized by open aperture z-scan data to remove nonlinear absorption effects before fitting with the
The nonlinear refractive index is expressed as $n_2 = \frac{3\Delta\phi \lambda}{2\pi I_0 L_{sep} n_2 \lambda}$, where $\Delta\phi$ is the change in phase, $I_0$ is the laser intensity, and $L_{sep}$ is a separation length.

For the USRN film, we measured the z-scan for sapphire glass, as shown in Fig. 2(d). The retrieved value by the system was $4.2 \times 10^{-17} \text{m}^2/\text{W}$, which indicates small nonlinear losses in this region.

Some research groups have calculated the nonlinear refractive index and two-photon absorption coefficients based on K-K relations, which describe the nonlinear refractive index to possess its highest value near the two-photon wavelength, as described by the expression $\alpha_2(\omega; \Omega) = \frac{c}{\pi} \int_0^{\infty} \frac{d\omega'}{\omega^2 - \omega''^2}$, where $\alpha_2(\omega; \Omega; \Omega')$ and $\alpha_2(\omega; \Omega; \Omega')$ denote respectively, a non-degenerate nonlinear index and an absorption at the existence of photons of $\omega$, $\Omega$, and $\Omega'$ frequency. The peak of three-photon absorption appears near $1.45 \mu m$ as shown in Fig. 3(b). At wavelengths larger than $1.2 \mu m$, two-photon absorption vanishes and the dominant nonlinear loss mechanism is three-photon absorption. The three-photon absorption coefficient is on the order of $10^{-25} \text{m}^2/\text{W}$ within the measurement range between $1.2 \mu m$ to $1.6 \mu m$. Nonlinear losses in this region are therefore characterized to be very small in the USRN film – a highly advantageous feature for nonlinear optics applications.

Graphs are shown for the measured z-scans of the USRN at different wavelengths: 0.9 μm, 1.2 μm, and 1.55 μm, demonstrating the nonlinear refractive index and absorption properties.
calculated from the K-K relation by assuming $2\omega \rightarrow \omega + \Omega$ and $\Omega = \omega$ is set after the integral calculation. This assumption is valid for the case of degenerate nonlinear refractive indices. Consequently, the nonlinear refractive index is calculated as

$$n_2 = \frac{1}{c} \frac{\partial^2 \alpha}{\partial \omega^2} \left| \text{Re} \int_0^\infty \frac{d\nu}{\nu} \frac{\gamma}{(\nu + \gamma)^{3/2}} \right|^2$$

where we inserted a small quantity, $\gamma \ll 1$, which represents a phenomenological damping loss to avoid singularities in the integral calculation. The calculated theoretical values are drawn in Fig. 3(c,d). The red solid line represents the theoretical calculation in a direct band gap Sheik-Bahae (S.-B.) model; the blue lines are the fitted result by changing the $K$ value in the Sheik-Bahae model, and magenta lines are the fitted results with the model for indirect band materials.

Figure 3. (a) The nonlinear refractive index measured using z-scan (black squares) and USRN waveguide experiments (red star). (b) Multi-photon absorption coefficients of USRN in the IR range characterized using z-scan measurements. Measured and theoretical values of (c) two-photon absorption coefficients and (d) the Kerr nonlinear refractive index are shown. The red solid line represents the theoretical calculation in a direct band gap Sheik-Bahae (S.-B.) model; the blue lines are the fitted result by changing the $K$ value in the Sheik-Bahae model, and magenta lines are the fitted results with the model for indirect band materials.
and nonlinear absorption loss is calculated using, where corresponding to the $H$. The nonlinear figure of merit (FOM) defined as $n^2/\mu_m$. The variation can be calculated according to length of 100

The nonlinearity value fitted with the indirect band gap model. The allowed-forbidden transitions dominant near the two-photon edge is used for the fit. The peak position of the refractive index is well matched to the experimental results for the transition calculation in the indirect bandgap. However, with the fitted value for nonlinear refractive value, the two-photon absorption graph was not satisfied as shown in Fig. 3(c). Consequently, for theory and experiment of dispersive nonlinear optical properties in the USRN to have perfect agreement, detailed information pertaining to the band structure and existing localized band states is needed. In the indirect band gap model, the nonlinear parameters are related to the electron-phonon matrix. Therefore, it follows that the characterized nonlinear coefficients could be used for indirect measurements of the electron-photon matrix, which is important to understand the interaction between electrons and phonons.

It is noted that the measured nonlinear refractive index of the USRN film is higher than that in crystalline silicon. Some groups have reported that deposited amorphous Si:H has a higher nonlinear refractive than crystalline Si$^{23,24}$. The higher nonlinear refractive index arises from a free carrier effect excited by two step absorption in the existence of defect states in amorphous-Si$^{23}$. The nonlinear refractive index has the same sign as the Kerr nonlinear index$^{24}$. We postulate that the same phenomenon could explain the high nonlinear refractive index in USRN films. Although more systematic explanation of the origin of the high nonlinearity is needed, it is possible that the increased nonlinear refractive index observed in USRN relative to crystalline silicon could arise from a single photon resonance from band states located in the half-band gap. The 3rd order nonlinear susceptibility tensor in the randomly homogeneous amorphous systems has two independent components, the two-photon process and one-photon process. The one-photon process is usually negligible and ignored for wide band gaps. However, the one-photon process could be important in band gap states generated by defects in the amorphous USRN material. For the nonlinear refractive index, the susceptibility $x_{1111}$($\omega; \omega + \omega + \omega$) corresponding to one photon process is proportional to $\alpha_{1111}^2/\omega_0^2$, where $\alpha_{1111}$ refer to the energy of band states, the valence band, and the photon respectively. The vanishing denominator term contributes to the resonant increase of nonlinear refractive index in the existence of band states. These defect states potentially give rise to an increase in the nonlinear refractive index.

Nonlinearity gives rise to unwanted attenuation in signal processing applications$^{25-28}$. For all-optical optical switching devices leveraging the nonlinear refractive index, a tradeoff exists between high nonlinear refractive index and low nonlinear absorption. The nonlinear phase variation is achieved according to $n_k I L$, where $k$ is the wave number, $I$ is peak intensity and $L$ is a propagation length of a beam through a nonlinear material. The nonlinear absorption loss is governed by $\alpha_2 I L$. The nonlinear figure of merit (FOM) defined as $n_k^2/\lambda \alpha_2$, is a commonly used quantity in the two-photon absorption region for assessing the suitability of a material for all-optical switching applications. For the directional coupler possessing the most stringent requirements, FOM > 2 should be satisfied$^{29}$. The calculated FOM for USRN in the two-photon absorption region ($\lambda < 1.2 \mu m$) is less than 0.2, not satisfying the criterion for switching application as shown in Fig. 4(a). The achievable phase variation by the nonlinear effect and signal loss by the nonlinear absorption at 1.05 $\mu m$ is drawn in Fig. 4(b), where the propagation length is assumed to be 100 $\mu m$. The figure implies that the energy of the input beam is largely absorbed before the required phase variation for a switching application is achieved. However, for longer wavelengths larger than 1.2 $\mu m$ within the three-photon absorption region, switching applications may be efficiently implemented as shown in Fig. 4(c). The telecommunications wavelength of 1.55 $\mu m$, and a propagation length of 100 $\mu m$ is used. Using a technique similar to that used for the two-photon absorption region, the phase variation can be calculated according to $n_k k I L$ and nonlinear absorption loss is calculated using $\alpha_2 I L$. The phase variation is 1.75 at the 10 $GW/cm^2$ peak intensity although the three-photon nonlinear absorption loss is just 7%. Greater phase variation can be obtained if the requirement on the absolute amount of nonlinear loss is

Figure 4. (a) FOM for the USRN material within the two-photon absorption region. (b) The achievable phase variation and two-photon absorption loss vs. peak intensity at 1.05 $\mu m$ based on the measured nonlinear coefficients. (c) The achievable phase variation and three-photon absorption loss vs. peak intensity at 1.55 $\mu m$. 
relaxed. Consequently, the USRN material is a good material for all-optical switching in the complete O- to L-telecommunications bands because of the high nonlinear refractive index and the small three-photon absorption coefficient.

Conclusions

We have experimentally characterized the dispersive nonlinear refractive index, two- and three-photon absorption coefficients at wavelengths between 0.8µm–1.6µm. Between 0.8µm–1.2µm, two-photon absorption is observed and the trends for the nonlinear refractive index and two-photon absorption are observed to satisfy K-K relations. The measured coefficients are comparable to the theoretically calculated values based on K-K relations. The measured spectral form is different from the calculation based on a simple band structure assuming a parabolic form. This implies that the difference observed in the measured spectral form could be used to retrieve a complex band structure. The resonant peak having the highest value, 8.1 × 10⁻¹⁵ m²/W, exists at 1µm corresponding to the two-absorption edge. The FOM in the two-photon absorption region is too small to satisfy the criterion for all-optical switching. Fortunately, the USRN material is demonstrated to have a vanishing two-photon absorption at wavelengths beyond 1.2µm, where telecommunications applications operate. In the three-photon absorption region, 1.2µm–1.6µm, covering the complete O- to L- telecommunications bands, the nonlinear refractive index is large and the three-photon absorption is small, making USRN highly advantageous for nonlinear optics in this region. The criterion for all-optical switching in this region is easily satisfied. Based on the measured dispersive nonlinear coefficients of the USRN, a proper propagation lengths and intensity can be selected to design low power, all-optical switching devices or modulators at optical communication wavelengths. The results further show USRN to be an advantageous material for nonlinear optics applications at the telecommunications wavelengths.

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Author Contributions
B.-U.S., J.W.C. and D.K.T.N. performed the optical characterization experiments, performed material growth and characterization. B.-U.S. analyzed the nonlinear experimental data. B.-U.S., J.W.C., D.K.T.N. and D.T.H.T. wrote, read and contributed to the manuscript. D.T.H.T. supervised the project.

Additional Information

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