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Multi-photon absorption and third-order nonlinearity in silicon at mid-infrared wavelengths

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Abstract: Silicon based nonlinear photonics has been extensively researched at telecom wavelengths in recent years. However, studies of Kerr nonlinearity in silicon at mid-infrared wavelengths still remain limited. Here, we report the wavelength dependency of third-order nonlinearity in the spectral range from 1.6 µm to 6 µm, as well as multi-photon absorption coefficients in the same range. The third-order nonlinear coefficient $n_2$ was measured with a peak value of $1.65 \times 10^{-13}$ cm$^2$/W at a wavelength of 2.1 µm followed by the decay of nonlinear refractive index $n_2$ up to 2.6 µm. Our latest measurements extend the wavelength towards 6 µm, which show a sharp decrement of $n_2$ beyond 2.1 µm and steadily retains above 3 µm. In addition, the analysis of three-photon absorption and four-photon absorption processes are simultaneously performed over the wavelength range from 2.3 µm to 4.4 µm. Furthermore, the effect of multi-photon absorption on nonlinear figure of merit in silicon is discussed in detail.

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1. Introduction

The interest in the development of high Kerr nonlinearity with weak multi-photon absorption (MPA) processes has increased over the past decade, because of their importance for ultra-fast all-optical signal processing. Near-infrared (near-IR) and short wave infrared (SWIR) nonlinear optical properties of silicon, namely, the third-order susceptibility $\chi^{(3)}$, have been widely studied in the past [1,2]. However, in the mid-infrared (mid-IR), its nonlinearity has not been well characterized. The multi-photon absorption places a fundamental limitation on the usefulness of any high $\chi^{(3)}$ material in all-optical switching schemes based on an intensity-dependent refractive index [3], especially strong two-photon absorption (TPA) in silicon. At telecom wavelengths, relatively high peak intensity is normally required to trigger the third-order Kerr nonlinearity, but also leads to a strong TPA [3]. Therefore, the combination of high Kerr nonlinearity and relatively weaker MPA beyond the TPA region makes silicon highly promising for nonlinear applications at mid-IR wavelengths.

Silicon photonics has been identified as a promising material platform for applications in the mid-IR spectral region, especially for building nonlinear optical devices, due to the relatively lower linear and nonlinear absorption within this spectral region [4]. The third-order nonlinear coefficients (n2) and multi-photon absorption processes have been previously reported up to 2.75 µm [5], which shows a decrement of n2 towards longer wavelength from the peak value at 1.9 µm [6,7]. Here, we extend the measurements further to 6 µm in the mid-IR range.

2. Z-scan experiments

We explore the measurements of degenerated MPA coefficients and nonlinear refractive index n2 in silicon, in this broad mid-IR range, using the Z-scan technique [5,7,8]. Z-scan has been widely adopted as a simple single beam technique to obtain MPA coefficients and n2 with the position-dependent transmittance variation by scanning a sample through the focal region of a Gaussian beam [9]. We utilized 150 fs pulses from an optical parametric amplifier source pumped by a 0.8 µm Ti:sapphire regenerative amplifier (Coherent, OperA) at 1 kHz. Combined with a difference frequency generator (DFG), wavelength tunability from 0.24 µm to 0.24 µm can be achieved. The pulses were focused at normal incidence onto the silicon using a CaF2 lens with 7.5 cm focal length, which minimize the optical loss induced by conventional near-IR optics. The sample is a 675 µm thick p-doped single crystal silicon (~1
× 10^{15} \text{ cm}^{-3} \text{ doping concentration}, cut with the normal axis along the [100] crystalline direction, such that the incident light is polarized along the [010] direction. The transmitted optical signal is collected by another CaF$_2$ lens with 7.5 cm focal length, and focused onto PbSe detectors (Thorlabs, PDA20H, 1.5-4.8 µm) in reference and signal arms for lock-in detection. Measurements with wavelengths beyond 4 µm are operated by a pyroelectric power probe (Laser Probe, RkP-575, 0.2-20 µm). There will be a 10-15% error tolerance when using the pyroelectric power probe due to thermal effects.

The MPA coefficients and $n_2$ were extracted by analyzing open- and closed-aperture measurements, respectively [8]. When the detection is performed with the presence of aperture, the detected signal has a peak-valley trace, depending on the strength of nonlinear refraction at the focal point. In comparison, in the case of open-aperture detection, the detected signal shows a valley-only trace, which indicates the occurrence of nonlinear MPA, depending on the wavelengths. To emphasize, the determination of beam waist (leads to the calculation of peak intensity) could result approximately 15% error during the experiments depending on the wavelengths, which is considered in addition to the 10-15% error introduced from the pyroelectric power probe by adding errors bars in Fig. 1.

The normalized transmittance can be obtained by integrating the intensity $I$. The total time-integrated transmission for the open aperture is theoretically fitted with Eqs. (1), (2) and (3) obtained from the derivations for MPA coefficients by Sutherland et al. [7,10],

\[
T_{OA(\text{op})} = \frac{1}{\{1 + (n-1)\alpha_L[I_{00}/(1 + (z/z_0)^2)]^{n-1}\}^{1/2}} \\
T_{OA(\text{cl})} = \frac{1}{\{1 + 2\alpha_L[I_{00}/(1 + (z/z_0)^2)]^{3/2}\}^{1/2}} \\
T_{OA(\text{cl})} = \frac{1}{\{1 + 3\alpha_L[I_{00}/(1 + (z/z_0)^2)]^{5/2}\}^{1/2}}
\]

Fig. 1. Variation of the MPA coefficients in silicon at mid-IR wavelengths (2.6-4.4 µm).
where $I_{00}$ is the peak intensity, $z$ is the sample position, $z_0 = \pi \omega_0^2 / \lambda$ is the Rayleigh length; $\omega_0$ is the beam waist at the focal point ($z = 0$), $\lambda$ is the laser wavelength; effective path lengths in the sample for three-photon absorption (3PA) and four-photon absorption (4PA) are given as $L'_{\text{eff}} = [1 - \exp(-2\alpha_0 L)] / 2\alpha_0$ and $L''_{\text{eff}} = [1 - \exp(-3\alpha_0 L)] / 3\alpha_0$ respectively. To clarify, the $\alpha_0$ is the linear absorption coefficient with wavelength dispersion, which is negligibly small at mid-IR wavelengths. Therefore, the effective path length at $\alpha_0$ 0° is approximately identical to the L (thickness of the sample). In this case, the beam waist $\omega_0$ ranged from 38 to 123 µm for different wavelengths. As shown, $\alpha_3$ and $\alpha_4$ present nonlinear absorption coefficients for 3PA and 4PA.

The peak intensity $I_{00}$ at the sample is determined as $I_{00} = \text{laser\_power} / \left[ (\pi \omega_0^2) \times \text{pulse\_repetitions} \times \text{pulse\_width} \right] \times 40\%$, due to Fresnel reflection and back scattering at the one-side polished sample surface (40% of the measured incident intensity). Fresnel reflection is normally stated as 30% if both sides are well polished. In our case, single-side polished sample is used. Therefore, in addition to Fresnel reflection, backscattering caused by unpolished side gives rise to additional loss which is ~10% from transmission measurements.

By fitting the experimental data with these curves, MPA coefficients can be estimated. Peak intensity of 57 GW/cm² is applied, the MPA coefficients are extracted from the open aperture Z-scan as shown in Fig. 1. When going from longer to shorter wavelength, we observed a steep increment of MPA coefficients towards the transition wavelength between 3PA and 4PA (the Si bandgap energy $E_g = 1.1$ eV are approximately three times of photon energy at 3.3 µm, four times at 4.4 µm and five times at 5.5 µm, correspondently). In Fig. 1, beyond 2.2 µm where 3PA dominates, the 3PA obeys the two-band theory of MPA [11,12], increasing dramatically with the red-shift of wavelengths and rolling over before reaching the 3PA-4PA transition wavelength. Within the 4PA dominant region, the 4PA follows the similar rule as 3PA, apart from a significant drop in nonlinear absorption coefficients. By further extending the wavelength beyond 4.4 µm, 5PA was not observed at an even higher peak intensity of 100GW/cm², where the first experimental 5PA was only recently reported by Q. Zheng et al [13].

Overall, the nonlinear absorption still shows a dramatic drop in magnitude with wavelength dispersion. Additionally, the MPA induced free-carrier absorption (FCA) can
induce an error for calculation of MPA coefficients. Especially at longer wavelengths, the FCA coefficient is proportional to $\lambda^2$, thus, the error will increase gradually once approaching the 4PA region, but the total contribution to the MPA is believed to be comparably small due to low doping concentration [14].

The $n_2$ can be obtained by analyzing closed-aperture Z-scan measurements [14], where the transmitted light through silicon sample is filtered by an aperture, with a transmittance of 40%. Figure 2 shows selected Z-scan results for Si and SiO$_2$ reference samples: Fig. 2(a) and (b) show the normalized closed- (40%) and open-aperture transmittance for 675µm-thick Si at 2.6 µm. Figure 2(c) shows normalized transmitted transmittance versus z-coordinate with the presence of a 40% transmitted aperture for a 1mm-thick SiO$_2$ at 1.55 µm. For the proof of accurately analytic fit, the $n_2$ of SiO$_2$ is measured to be $2.98 \times 10^{-16}$ cm$^2$/W, which is almost identical to two reference values of $2.2 \times 10^{-16}$ cm$^2$/W [15] and $2.96 \times 10^{-16}$ cm$^2$/W [16] found in the literature. A closed-aperture Z-scan example at a longer wavelength of 3.8 µm is also fitted in Fig. 2(d), with a $n_2$ value of $5.8 \times 10^{-14}$ cm$^2$/W. The closed-aperture data, $T_{CA}$, are fitted to Eq. (4) [17],

$$T_{CA} = 1 + \frac{4 \Delta \phi_0 (z/z_0)}{[1 + (z/z_0)^2][9 + (z/z_0)^2]}$$

(4)

where $\Delta \phi_0$ is the phase change of the laser beam due to nonlinear refraction. Subsequently, the $n_2$ value can be calculated through Eq. (5):

$$n_2 = \frac{4 \Delta \phi_0 \lambda}{2 \pi I_{0\mu} L_{eff}}$$

(5)

In Fig. 2(a), at a peak intensity of 57 GW/cm$^2$, the $n_2$ value at 2.6 µm is calculated to be $2.5 \times 10^{-14}$ cm$^2$/W with the presence of 3PA. The 3PA coefficient at this wavelength is determined as $2 \times 10^{-21}$ cm$^3$/W$^2$ as shown in Fig. 2(b). Furthermore, the dispersion of $n_2$ from 1.6 to 6 µm is measured and shown in Fig. 3. In these measurements, any errors induced from

![Fig. 3. Variation of $n_2$ in silicon at mid-IR wavelengths from 1.6 to 6 µm: crosses (data from ref. 7) and diamonds (data measured by SUTD-NUS).](image)
the determination of intensity could translate to errors in the accuracy of the $n_2$ calculation, including the pulse energy fluctuation, background noise, thermal effects in power detection and beam waist determination. The displayed error bars in Fig. 3 indicate the considerations of all distributing uncertainty factors listed above, which is estimated by taking a few independent measurements under the identical conditions.

A sharp increment of $n_2$ from telecom wavelength towards mid-IR range is observed with a peak at 2.1 µm, following by a dip at longer wavelength, which matches with the reference data in the literature (red crosses in Fig. 3) [7]. In order to further explore the dispersion of $n_2$ at mid-IR wavelengths, the Z-scan measurements are extended to longer wavelength beyond 2PA region up to 6 µm. According to Kramers-Kronig (KK) relations [18], two-photon resonance leads to the peak of $n_2$ at the edge of transition wavelength (2.2 µm), which is in line with measured results in Fig. 3. The value of $n_2$ appears to decay rapidly after entering 3PA region, and saturates at a relatively constant value of $5.2 \times 10^{-14}$ cm²/GW. The variation of $n_2$ appears to be consistent with KK transform [19,20] as shown in Fig. 3. These results are a strong indication of consistent strength of Kerr nonlinearity at mid-IR wavelength.

### 3. Calculation of nonlinear figure of merit and discussion

The manipulations and usefulness of Kerr nonlinearity are always limited by MPA at high peak intensities and subsequently generated free carrier absorption (FCA) [2]. Especially in the 2PA region, the strong nonlinear absorption induced at high optical intensities strongly presents. For instance, as the application of optical switching, a low nonlinear absorption and higher $n_2$ are ideal. Therefore, here, we introduce the nonlinear figure of merit (FOM) to be $n_2 / \lambda I \alpha_{3pA}$ and $n_2 / \lambda^2 I^2 \alpha_{3pA}$ [5], corresponding to 3PA and 4PA dominant regions, with peak pump intensities ranging from 35 to 57GW/cm².

![Fig. 4. Nonlinear figure of merit (FOM) with presence of MPA: crosses (data from ref. 7) and stars (data measured by SUTD-NUS).](image)

According to Boggess et al. [21], the FCA coefficient $\sigma_{\text{FCA}}$ at 1064nm is $5 \times 10^{-18}$ cm², due to $\sigma_{\text{FCA}}$ is proportional to $\lambda^2$, thus, the corresponding $\sigma_{\text{FCA}}$ at 2600nm can be estimated to be $5 \times 10^{-17}$. By our calculation, the absorption change due to high-peak-intensity induced FCA ($\Delta \alpha_{\text{FCA}}$) is determined as 0.07 cm⁻¹, where the absorption change due to 3PA $\Delta \alpha_{3pA}$ is estimated as 0.84 cm⁻¹. Therefore, we can conclude that the high-peak-intensity induced FCA
counts less than 10% error in our experiments. The doping induced FCA is expressed as $N_0 \sigma_{FCA}$ ($N_0 = \text{doping concentration}$), which has been calculated with a value of 0.03 cm$^{-1}$. As discussed, the FCA absorption caused by high peak intensity is calculated to be 0.07 cm$^{-1}$, which gives a total FCA value of 0.1 cm$^{-1}$ by adding up peak-intensity-induced FCA and doping inducing FCA. To compare, the absorption change due to 3PA is 0.84 cm$^{-1}$. The total FCA induced (0.1 cm$^{-1}$) remains approximately 10% of total nonlinear absorption. Therefore, the FCA originated from doping has limited impact on determining the MPA coefficients.

In Fig. 4, we use normalized FOM data at the 2PA region from F. Gholami et al. [5] to bridge up with our FOM results beyond 2.2 µm. It can be clearly observed in Fig. 4 that the FOM is lower at short wave-IR region, and increases sharply with increasing wavelengths, due to decaying MPA in the mid-IR region as shown in Fig. 1. Especially when it moves from near-IR towards the transition wavelength for 2PA to 3PA at 2.2 µm, the FOM rises as the effect of 2PA diminishes. In the region of 3PA, in contrast to that for 2PA, 3PA coefficient exhibits an increment for the red-shift of the wavelengths, resulting in a drop in FOM.

As discussed above, TPA is forbidden beyond 2.2 µm and hence, three-photon absorption determines FOM by $n_2 / \lambda I \alpha_{3PA}$, that indicates that FOM decreases if $n_2$ decreases, $\alpha_3$ increases or wavelength increases. The data in Fig. 3 shows that $n_2$ decreases at $\lambda > 2.2$ µm. The theory used in ref. 10 shows that $\alpha_3$ should increase with wavelength in this spectral range, which is consistent with our data in Fig. 1. All these factors lead to the decreasing trend at $\lambda > 2.2$ µm.

Furthermore, the value of $n_2$ is observed to decay beyond 2PA region, thus, the calculated FOM rolls over until it reaches the 3PA-4PA transition wavelength at 3.3 µm. Due to the dramatic decrement of MPA in 4PA dominant region, the nonlinear FOM exhibits a steady rise beyond 3PA region, which is almost identical to the magnitudes in the 2PA dominant short wave-IR region [19].

4. Conclusion

In summary, we report the measurements of degenerated MPA and Kerr nonlinear coefficients covering the near-IR and mid-IR spectral region up to 6 µm. The results indicate a good strength of Kerr nonlinearity and significant enhancement in nonlinear FOM at longer mid-IR wavelengths (3-6 µm), which appears to be a promising candidate for on-chip all-optical signal processing.

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