Two-photon absorption and Kerr coefficients of silicon for 850–2200 nm

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The degenerate two-photon absorption coefficient β and Kerr nonlinearity n_2 are measured for bulk Si at 300 K using 200 fs pulses with carrier wavelength of $850<\lambda<2200$ nm for which indirect gap transitions occur. With a broad peak near the indirect gap and maximum value of 2 ± 0.5 cm/GW, the dispersion of β compares favorably with theoretical calculations of Garcia and Kalyanaraman [J. Phys. B **39**, 2737 (2006)]. Within our wavelength range, n_2 varies by a factor of 4 with a peak value of 1.2×10^{-13} cm^2/W at $\lambda=1800$ nm. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2737359]

Within the past few years interest in silicon as a photonic material has been intensely renewed.[1] Silicon lasers,[2] optical amplifiers,[3] modulators,[4,5] all-optical switches,[6–8] waveguides,[9–11] and photonic crystals[6,7,12–14] have been reported. Because many of these systems operate at high optical intensity, accurate knowledge of the nonlinear characteristics of silicon is important for understanding and optimizing their performance. Nonetheless, to date measurements of the two-photon absorption (2PA) coefficient (β) or the nonlinear or Kerr index (n_2) of bulk silicon have only been carried out at a few wavelengths and, in many cases, only inferred from experiments on structured materials such as waveguides. Reintjes and McGroddy[15] were the first to isolate a two-photon absorption process in silicon from cascaded optical processes involving interband and intraband (free carrier) absorption; a value of $\beta=1.5$ cm/GW was obtained at 100 K for picosecond pulses at a fundamental wavelength $\lambda=1060$ nm; a similar value was obtained by Boggess *et al.* at 300 K.[16] Reitze *et al.*[17] measured 2PA across the direct gap (3.4 eV) of Si, reporting $5<\beta<36$ cm/GW for 90 fs pulses in the range $620>\lambda>550$ nm. In recent years, because of technological implications, focus has been on 2PA at telecommunications wavelengths for $\lambda=1300$ or 1500 nm, with values[9,10,18] of $\beta\sim0.8$ cm/GW being reported. However, no systematic experimental spectroscopic study has been carried out for 2PA across the indirect gap ($E_{gi}=1.12$ eV), although theoretical predictions have recently been made.[19,20] Information[18] for n_2 is even more scarce than for β with values ($\sim3\times10^{-14}$ cm^2/W) reported for 1300 and 1550 nm. Here, we present measurements of degenerate β and n_2 for fundamental wavelengths between 850 and 2200 nm and compare our results with theoretical predictions.

Measurements of β and n_2 are performed using the closed and open aperture *z*-scan technique, respectively. *Z*-scan has been widely adopted as a simple single beam technique to obtain β and n_2 with intensity variation achieved by scanning a sample through the focal region of a Gaussian beam.[21] The total time-integrated transmission for the open aperture technique is given by[18]

$$T_{\text{open}}(z) = 1 - \frac{1}{2\sqrt{2}}\frac{\beta I_0 L_{\text{eff}}}{1+(z/z_0)^2}, \qquad (1)$$

where z is the longitudinal scan distance from the focal point of a Gaussian beam with an on-axis peak intensity of I_0 (inside the sample) and z_0 is the confocal beam parameter; $L_{\text{eff}}=\alpha^{-1}(1-e^{-\alpha L})$ is the effective optical length, where α is the linear absorption coefficient for silicon (nonzero[22] for $\lambda<1100$ nm) and L is the sample thickness. For closed aperture z scan a circular aperture with transmissivity $S<1$ is placed behind the sample, and the transmission is recorded as a function of z position. For small absorptive and refractive changes the transmissivity is given by[18]

$$T_{\text{closed}}(z) = 1 - \frac{8\pi}{\lambda\sqrt{2}}\frac{z/z_0(1-S)^{0.25}L_{\text{eff}}n_2I_0}{(1+(z/z_0)^2)(9+(z/z_0)^2)} - \frac{1}{2\sqrt{2}}\frac{L_{\text{eff}}\beta I_0(3-(z/z_0)^2)}{(1+(z/z_0)^2)(9+(z/z_0)^2)}. \qquad (2)$$

For the experiments below we use $S=0.13$, which yields sufficient contrast for our purposes. The experiments were conducted with an optical parametric amplifier source delivering nominally 200 fs pulses at 1 kHz for $1100<\lambda<2200$ nm. With frequency doubling in a beta-barium borate crystal, we also obtained and used pulses with $850<\lambda<1100$ nm, with the short wavelength limit dictated by the increasing linear absorption[22] ($\alpha=5\times10^2$ cm^{-1} at 0.85 μm). Linearly polarized ~100 nJ pulses were focused onto the silicon using a 10 cm focal length lens, giving $z_0=5$ mm; detection was accomplished using Ge or PbS_2 detectors in reference and signal arms with lock-in detection. The focal spot size of 75 μm full width at half maximum, needed for obtaining peak intensity, was determined using a knife-edge measurement. The sample used was a 125 μm thick wafer of intrinsic, double sided polished crystalline Si, cut with the normal axis along the [001] crystal direction. Fresnel losses were incorporated in determining the peak intensity inside the sample. For the peak intensity (8 GW cm^{-2}) and the linear or 2PA absorption coefficients used in these measurements, the peak temperature increase and carrier density inside the sample are estimated to be <1 K and 6×10^{16} cm^{-3}, respectively. For a 5×10^{-18} cm^2 free carrier absorption cross section[16] at 1060 nm in Si, the estimated change in absorption is $<3\%$ of that obtained from 2PA. Similarly, while three-photon absorption across the direct

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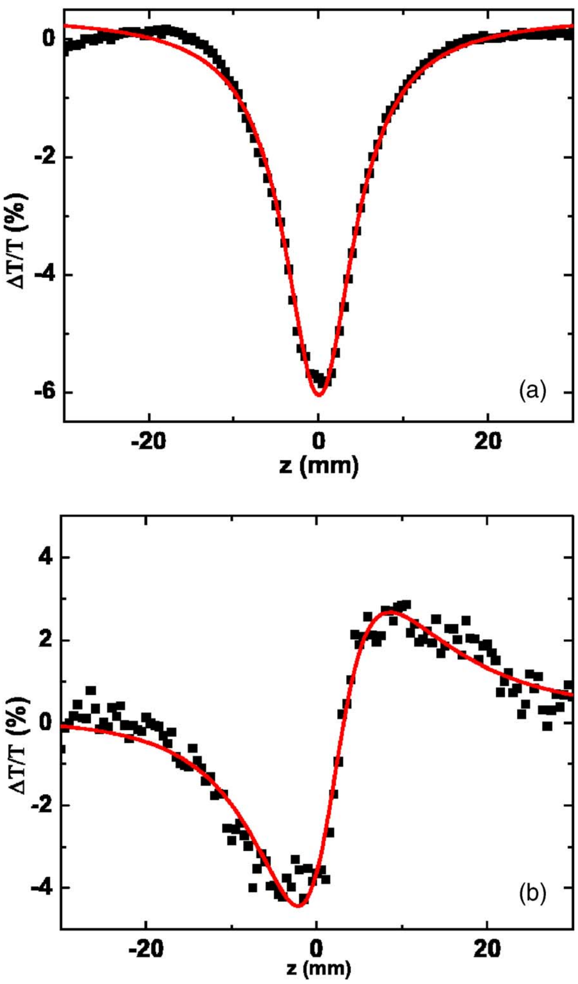

FIG. 1. (Color online) Typical z-scan traces for 125 μm thick Si at 1220 nm with a peak intensity of 7.8 GW/cm^2. (a) Open aperture z trace. (b) Closed aperture z-scan trace with S=0.13. The thin solid lines are best fits based on Eqs. (1) and (2).

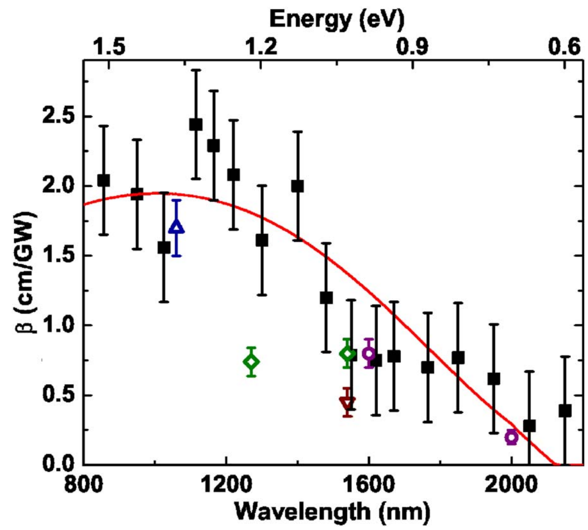

FIG. 2. (Color online) Measured value of β (squares) as a function of wavelength for a 125 μm thick sample silicon wafer. Data from other sources are also given: circles (Ref. 7), uptriangles (Ref. 15), down triangles (Ref. 9), and diamonds (Ref. 18). The solid curve represents the best fit based on calculations of Garcia and Kalyanaraman (Ref. 20).

gap can occur for $\lambda < 1070$ nm, its influence is estimated to be negligible based on the scaling rules of Wherret.[23] The maximum change in refractive index from free carrier (Drude) contribution is $\sim -10^{-4}$, more than an order of magnitude smaller and of opposite sign to that measured by us. Similarly, band filling and band gap shrinkage effects are calculated as negligible.[24] At several wavelengths it was also verified that the induced transmission change varied with I_0^2, indicating that cascaded processes such as 2PA followed by free carrier absorption were negligible. Measurements were attempted to observe anisotropy in the nonlinear absorption; however, for $\lambda > 1400$ nm the variation of β with linear polarization direction was $<5\%$ and within experimental error.

Typical closed and open aperture z-scan traces are shown in Fig. 1 for $\lambda = 1220$ nm. They display the shapes expected from Eqs. (1) and (2). Fits to the data yield $\beta = 2.1 \pm 0.4$ cm/GW and $n_2 = (4.7 \pm 2.0) \times 10^{-5}$ cm^2/GW. Values of β as a function of λ are illustrated in Fig. 2. The data are displayed along with values measured by others, as indicated. The experimental results are consistent within experimental error, which, in our case, is mainly related to obtaining values of peak laser intensity and small differences in beam shape with tuning. Not shown is the result $\beta \sim 5$ cm/GW obtained by Sabbah and Riffe[25] since this is only an estimate. Overall, one sees that β tends to zero for $\lambda > 2200$ nm, corresponding to a photon energy of $E_{gi}/2$. The β increases with decreasing wavelength before exhibiting a broad peak or plateau for $\lambda < 1200$ nm. Also illustrated in Fig. 2 is the theoretical dispersion curve for the degenerate β based on the calculations of Garcia and Kalyanaraman[20] (GK). While Dinu[19] earlier had only considered[26] "forbidden-forbidden" (f-f) indirect gap transitions, GK also incorporated "allowed-forbidden" (a-f) and "allowed-allowed" (a-a) optical transitions and showed that these latter processes dominate β for photon energies below E_{gi}. Overall, $\beta = \Sigma_{n=0}^{2} \beta^{(n)}$, with $n = 0, 1, 2$ for a-a, a-f, and f-f transitions.[26] The contribution of each process depends on fundamental photon energy ($\hbar\omega$) as $\beta^{(n)} = 2CF_2^{(n)}(\hbar\omega/E_{gi})$ for a material-dependent constant C, with the factor of 2 accounting for phonon emission and absorption processes in indirect gap optical absorption. For parabolic electron and hole bands and $x = \hbar\omega/E_{gi}$, $F_2^{(n)}(x) = [\pi(2n+1)!!/2^{n+2}(n+2)!](2x)^{-5}(2x-1)^{n+2}$, with the assumption that phonon energies are $\ll E_{gi}$. The f-f process, much weaker than the two other processes, peaks at $\hbar\omega \approx 5E_{gi}/2$ and only begins to dominate when linear absorption becomes strong. The other two processes peak for photon energies near E_{gi}. Given the difficulty in accounting for all the phonon-assisted processes, we fit our data with the functional form $\beta = \Sigma_n \beta^{(n)}$ to obtain $C = 43$ cm/GW. As seen in the figure, apart from the need to incorporate the scaling parameter, the dispersion characteristics of β are consistent with the GK calculation.[20]

Figure 3 shows our measurements of n_2 along with the measurements at 1300 and 1550 μm by Dinu *et al.*[18] The value of n_2 increases by a factor of 4 with increasing wavelength until 1800 nm, after which it declines slightly in value. Also shown is a theoretical estimate based on the Kramers-Krönig transform $[n_2(\omega) = c/\pi \int \beta(\omega,\omega')/\omega^2 - \omega'^2 d\omega']$ which makes use of the nondegenerate β with the approximation[27] $\beta(\omega,\omega') = \beta([\omega+\omega']/2)$. While the experimental and theoretical estimates peak at approximately the same wavelength, the estimate gives values well below the measured results. Including 2PA associated with direct gap transitions[17] in the calculation shifts the estimate upward by

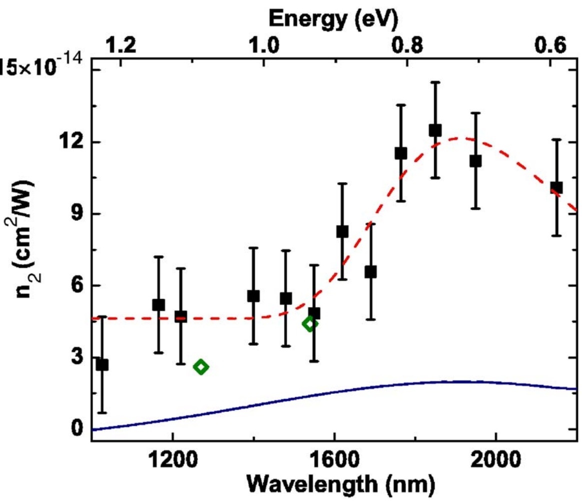

FIG. 3. (Color online) Measured value of n_2 (squares) as a function of wavelength for a 125 μm thick sample of intrinsic silicon. Data from Ref. 18 are also given (diamonds). The dashed curve is a guide to the eye, while the solid curve is based on a Kramers-Krönig transformation of the solid curve for β shown in Fig. 2.

only 1×10^{-14} cm^2/W. The remaining discrepancy is likely due to the neglect of Raman and quadratic stark contributions[27] in the Kramers-Krönig transform as well as the simple approximation for the nondegenerate β.

In summary we have measured the degenerate two-photon absorption and Kerr coefficient of bulk Si for $850<\lambda<2200$ nm associated with absorption across the indirect gap. The dispersion in the two-photon absorption coefficient is in good agreement with the functional form of recent theoretical predictions by Garcia and Kalyanaraman, considering the various approximations incorporated in the theory and the experimental error in the data. The dispersion of the Kerr contribution to the refractive index shows a factor of 4 variation in magnitude with the peak value located at a wavelength consistent with that estimated from a simple Kramers-Krönig transform of the theoretical two-photon absorption coefficient.

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