Three photon absorption in silicon for 2300–3300 nm

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We measure the spectral dependence of the degenerate three photon absorption coefficient, γ, for a Si [100] wafer using 200 fs pulses in the range 2300–3300 nm, i.e., photon energy between half and one-third the indirect band gap. For pulses linearly polarized along the [001] crystal axis γ increases from a value of near 0 cm^3/GW^2 at 3300 nm to a peak value of 0.035 cm^3/GW^2 at 2700 nm before decreasing with shorter wavelength; this is consistent with the dispersion expected from allowed-allowed-allowed transitions. At 2600 nm the γ value is ~30% larger for light polarized along [011] than along [001]. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2991446]

Interest in silicon as a photonic material has increased dramatically in recent years.[1,2] Many of the applications, such as silicon lasers,[3] optical amplifiers,[4] modulators,[5,6] and all-optical switches,[7,8] make use of silicon's lack of linear absorption at wavelengths longer than 1100 nm, corresponding to its indirect gap, $E_{gi}=1.1$ eV at 300 K. Since some of these devices operate at high intensities, however, nonlinear processes such as multiphoton absorptions and the Kerr effect become significant, and knowledge of the related optical constants is crucial. Silicon's two photon absorption (2PA) coefficient, β, and Kerr coefficient, n_2, have now been measured in the region from[9–11] 850 to 2200 nm, i.e., for a photon energy, $\hbar\omega$, as small as $E_{gi}/2$. As researchers explore applications beyond 2200 nm, e.g., a silicon Raman laser and amplifier,[12–14] higher order nonlinear absorption processes such as three photon absorption (3PA) with coefficient, γ should appear. Nonlinear absorption not only attenuates beams but can also inject electron-hole pairs, leading to free carrier absorption and dispersion. While knowledge of γ has been obtained in a number of direct and indirect gap III-V and II-VI semiconductors,[15–17] to date there has been no direct measurement of γ in silicon, apart from an estimate[18] ($\gamma \sim 7\times10^{-2}$ cm^3/GW^2) based on the value in GaP, also an indirect gap semiconductor. In this letter we report the spectral dependence of γ for silicon in the region 2200–3300 nm, i.e., for $E_{gi}/3<\hbar\omega<E_{gi}/2$.

Measurements of γ were performed in the open aperture z-scan geometry, a single beam technique more commonly used to obtain β.[19] For z-scan measurements of β using temporal/spatial Gaussian beams, analytical formulas exist for extracting the nonlinear coefficient. For higher order nonlinearities, a numerical approach must be taken. When 3PA is the only source of sample absorption the attenuation of a pulse with local intensity I is governed by

$$\frac{dI(z',r,t)}{dz'} = -\gamma[I(z',r,t)]^3, \qquad (1)$$

where r and z' are beam radial and longitudinal coordinates within the sample and t is time. This equation is numerically integrated over r, z, and t together with Fresnel boundary conditions to obtain the output (fluence) transmittance, T, in terms of the low-intensity transmittance T_0 and hence the differential transmittance, $\Delta T(z)/T_0=(T(z)-T_0)/T_0$, as a function of the pulse's focal point distance, z, from the middle of the sample.

The experiments were conducted with an optical parametric amplifier source delivering 200 fs [full width at half maximum (FWHM)] pulses at 1 kHz in the range $1100<\lambda<3600$ nm. Spectral measurements of the pulses give a typical pulse bandwidth of ~200 nm, slightly larger than that for transform-limited pulses. Linearly polarized 300 nJ pulses were focused at normal incidence onto the silicon using a 4 cm focal length CaF_2 lens; lock-in detection was employed with PbSe detectors in reference and signal arms. The focal spot size of 50 μm (FWHM) was determined using a knife-edge measurement; the value is consistent with the expected Gaussian confocal parameter of 1.5 mm. The sample used is a 500 μm thick wafer of intrinsic double sided polished single crystal Si, cut with the normal axis along the [100] crystal direction. For a peak incident intensity of 25 GW cm^{-2} and the γ obtained from our measurements, the peak temperature increase inside the sample is estimated to be <1 K. The estimated change in absorption[20] due to free carriers (density $<10^{17}$ cm^{-3}) induced by 3PA is <1% of that obtained from 3PA.

To verify that the measured $\Delta T/T_0$ is consistent with 3PA we took z-scan traces at $\lambda=2900$ nm for different peak laser intensities, I_0, with pulses linearly polarized along the crystal [001] direction. Figure 1 shows the maximum differential transmission $(\Delta T/T_0)_{max}$ [i.e., $\Delta T(z=0)/T_0$] for the different peak intensities along with the best theoretical fits for 2PA, 3PA, and four photon absorption (4PA). It is evident, especially when considering data at the highest and lowest intensities, that the data are consistent with 3PA. A typical z-scan trace is given in the inset to Fig. 1 along with a fit calculated with the approach outlined above, resulting in $\gamma=0.04\pm0.01$ cm^3/GW^2.

For pulses polarized along [001], values of γ as a function of λ are illustrated in Fig. 2. Overall, one sees that γ tends to zero near $\lambda=3300$ nm as expected for 3PA. γ increases with decreasing wavelength from 3300 nm before exhibiting a broad peak near 2700 nm with a maximum value of 0.035 ± 0.01 cm^3/GW^2. A theoretical expression for the dispersion of the 3PA coefficient can be obtained using the procedure outlined by Garcia and Kalyanaraman.[21] As pointed out by these authors, and also earlier by Wherrett,[22]

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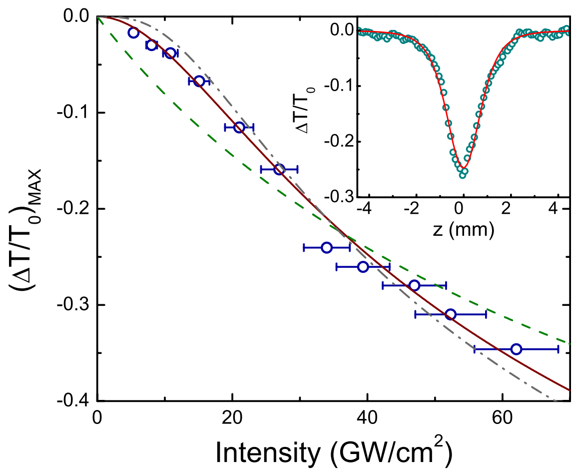

FIG. 1. (Color online) $(\Delta T/T_0)_{\max}$ as a function of peak intensity at 2900 nm. The circles represent experimental data while the lines are best theoretical fits based on 2PA (- - -), 3PA (—), and 4PA (– – –). The inset shows a typical z-scan trace with corresponding 3PA fit.

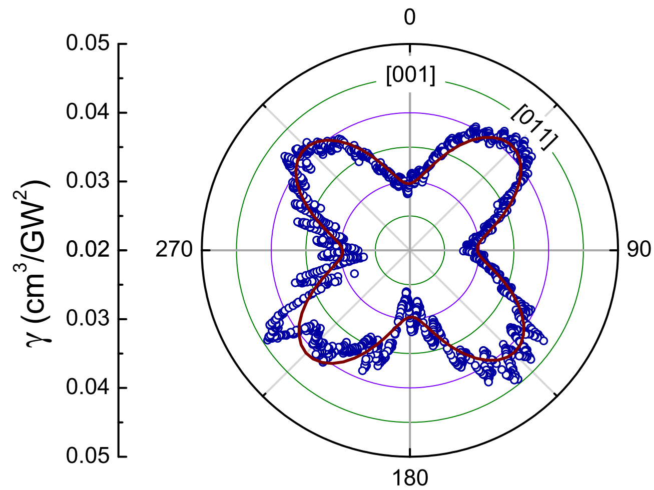

FIG. 3. (Color online) Value of γ at 2600 nm as a function of crystal rotation angle, φ, about the [001] axis. The solid curve is the best fit based on $\gamma = \gamma_0(1-\sigma\cos 4\varphi)$.

electric dipole allowed and forbidden (self-scattering) optical transitions can typically contribute to nonlinear absorption. In the case of an indirect gap material, phonon scattering can assist with overall momentum conservation. However, for odd-order nonlinear absorption, Wherrett[22] showed that allowed transitions should dominate any forbidden transitions; in the case of 3PA such processes would contain only allowed-allowed-allowed sequences. Assuming parabolic valence one can easily show that

$$\gamma = C\left[\frac{E_{gi}}{\hbar\omega}\right]^9 (\hbar\omega/E_{gi} - 1/3)^2, \tag{2}$$

where C is a constant. Given the difficulty in calculating the magnitude of all the phonon-assisted optical absorption processes, we make no attempt to provide a complete theoretical expression for γ Rather we fit our data to Eq. (2) to obtain $C = 1.54 \times 10^{-3}$ cm^3/GW^2. As seen in Fig. 2, the dispersion characteristics of the data are consistent with this theoretical expression although, at the longer wavelengths, there is an apparent wavelength offset between experimental and theoretical values. While part of this may reflect simplifying assumptions in the theory, it is expected that the nonzero bandwidth of the pulses would increase the apparent value of γ in a region where γ increases with decreasing λ leading to the apparent wavelength offset. This would also account for the nonzero value of γ at λ ~3300 nm, where theoretically it should be zero.

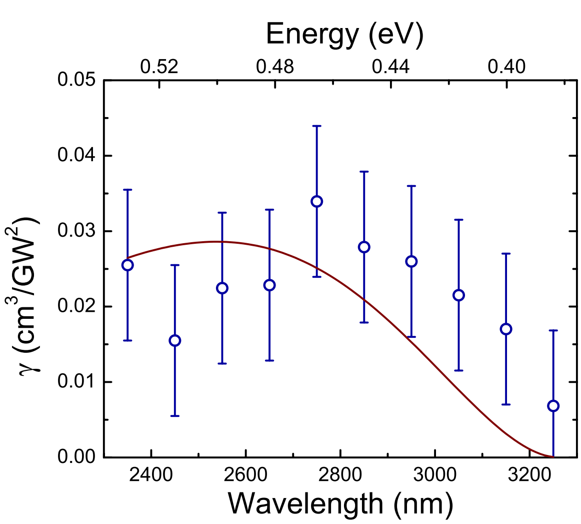

FIG. 2. (Color online) Measured values of γ as a function of wavelength for pulses polarized along the [001] axis. The solid curve represents the best fit based on a simple dispersion model.

For $\lambda < 2200$ nm 3PA contributions to the total absorption are normally neglected since 2PA processes should dominate for low intensity. While the magnitude of γ is expected to decrease with decreasing wavelengths, 2PA and 3PA absorption rates would become comparable when $\beta = \gamma I$. For example, at $\lambda = 1900$ nm, from Ref. 9 $\beta = 0.62$ cm/GW. For the same λ we obtain $\gamma = 0.015$ cm^3/GW^2 from the theoretical dispersion relation for γ. Hence one could expect comparable absorption rates at a local intensity of $I \sim 40$ GW/cm^2.

Finally, we have measured the anisotropy of γ by performing z-scan measurements while rotating the sample about its [100] axis. Results are shown in Fig. 3 for pulses with $\lambda = 2600$ nm. The data show the expected fourfold symmetry and, indeed, can be fitted to an expression of the form $\gamma = \gamma_0(1-\sigma\cos 4\varphi)$, where φ is the angle of the pulse linear polarization relative to the [001] axis. A best fit of this expression to the data gives $\gamma_0 = 0.035$ cm^3/GW^2 and $\sigma = 0.16$. The anisotropy coefficient, σ is somewhat larger than the corresponding value of ~0.06 observed[23] for 2PA. For a material with cubic symmetry, the fourth rank susceptibility tensor governing degenerate 2PA has only two independent nonzero elements,[23] and the observed anisotropy for a given crystal face can be used to deduce β for any crystal orientation and beam polarization. However, degenerate 3PA is governed by a sixth rank tensor, and we determine that there are four independent nonzero elements for a cubic medium. Additional measurements of γ would therefore be necessary to fully characterize the 3PA anisotropy for arbitrary orientations of crystalline Si.

In summary, for a [100] oriented wafer of crystalline silicon we have measured the 3PA coefficient in the 2300–330 nm range. We have also measured the anisotropy of this coefficient by rotating the wafer about the [100] axis and found that the 3PA coefficient varies by ~30% with a maximum value for light polarized along the [011] direction.

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