Wavelength dependence of two photon and free carrier absorptions in InP

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Abstract: Nonlinear absorption at 1.064 and 1.535 μm wavelengths by two photon and free carrier absorption processes in undoped and Fe doped InP has been investigated. Using picosecond and nanosecond duration lasers, a self-consistent set of the two photon and free carrier absorption coefficients are experimentally obtained through nonlinear transmission measurements for the first time. Reduced carrier recombination lifetime caused a decrease in nonlinear absorption of nanosecond duration laser pulses in Fe doped samples.

OCIS codes: (190.4400) Nonlinear optics, materials; (190.5970) Semiconductor nonlinear optics including MQW.

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1. Introduction

Two photon absorption (2PA) in InP is a nonlinear optical phenomenon readily observed with lasers having irradiance in the MW/cm^2 level or above if the photon energy of the laser is greater than half the bandgap energy of the semiconductor. The 2PA process generates *e* - *h*

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pairs which in turn absorb one photon by a process known free carrier absorption (FCA). The FCA process is quantified by the free carrier absorption cross section, σ_{abs}. Since the amount of carriers generated is dependent on the pulsewidth and irradiance of the laser source, the total nonlinear absorption (NLA) becomes dependent on the integrated irradiance (fluence) of the incident pulse. For pulse durations in the range of 10's of ps and longer, FCA initiated by 2PA is the dominant nonlinear absorption mechanism at high values of incident irradiance.

Previously reported values of the 2PA coefficient (β) at 1.064 μm vary from 90 to over 200 cm/GW [1,2]. Theoretical values reported vary from 26 to over 300 cm/GW [3]. More recent work at 1.55 μm reports a β between 24 - 33 cm/GW [4]. Although several studies related to 2PA have been carried out, to our knowledge, no nonlinear measurement of σ_{abs} in InP has been reported.

In this work, we report the results of nonlinear absorption measurements conducted on 0.96 and 2.0 mm thick samples of undoped (as grown) InP and a 0.92 mm thick Fe-doped (*n*-type) InP sample using the irradiance scan method [5]. Since FCA in InP predominantly arises from absorption due to holes [14], absorption coefficients of *p*-type InP samples (Zn:InP) with two different thicknesses and concentrations were measured as a function of wavelength. Hall measurements on these two samples provided their carrier concentrations, from which the hole absorption cross section at different wavelengths was determined. The hole σ_{abs} values were consistent with earlier measurements [4,6] but differed from the values obtained through NLA measurements presented here.

2. Theory and Numerical Modeling

Starting from Maxwell's equations with the only assumption being the slowly varying amplitude approximation, where the second order derivatives along the propagation direction and in time are assumed to be zero, Eq. (1) was derived which describes the propagation of light having a wavelength λ and complex electric field amplitude, $A(x,y,z,t)$, through a third order nonlinear medium along the z direction

$$\frac{\partial \mathrm{A}}{\partial \mathrm{z}} = \frac{i}{2k_0}\nabla_{\perp}^2 A + \frac{2i\pi}{\lambda}\left(\sigma_{ref} N + \frac{dn}{dT}\Delta T + \gamma I\right)A - \left(\alpha + \sigma_{abs} N + \beta I\right)\frac{A}{2} \qquad (1)$$

where

$$\nabla_{\perp}^2 = \frac{\partial^2}{\partial \mathrm{x}^2} + \frac{\partial^2}{\partial \mathrm{y}^2} \qquad (2)$$

In Eq. (1), N denotes the density of the photo-generated electrons (and holes). T is the lattice temperature and ΔT is the absorption induced change in temperature. σ_{ref} is the free carrier refraction coefficient (equal to dn/dN, the change in refractive index (n_0) per unit change in carrier density, N), dn/dT is the thermo-optic coefficient and γ [7] denotes the intrinsic nonlinear refraction coefficient. α is the linear absorption coefficient, $k_0 = 2\pi n_0/\lambda$ and the irradiance is given by $I = 2n_0\varepsilon_0 c|A|^2$.

For laser pulses of picosecond and nanosecond duration the following equations apply

$$\frac{\partial \mathrm{N}}{\partial \mathrm{t}} = \frac{\alpha I}{h\nu} + \frac{\beta I^2}{2h\nu} - \frac{N}{\tau} \qquad (3)$$

$$\frac{\partial \Delta \mathrm{T}}{\partial \mathrm{t}} = \frac{1}{\rho c}\left[\left(\alpha + \sigma_{abs} N\right)I + \beta I^2\right] \qquad (4)$$

where ρ denotes the density, c denotes the specific heat of the medium, ν denotes the frequency of light and τ the carrier recombination lifetime. Diffusion of carrier density and temperature was ignored in Eqs. (3) and (4). The accuracy of this assumption was verified by measuring the nonlinear transmission using ns lasers at different spot sizes. The change in

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transmission as a function of irradiance was not spot size dependent, indicating that carrier diffusion is not significant at these ns pulse durations.

The numerical method used to solve Eqs. (1), (3) and (4) is as follows. Equation (1) is numerically solved via the operator splitting method [8–10]. The right side of the equation is separated into linear and nonlinear operations which are solved independently. First the sample length is divided into a number of slices of length Δz. At $z = 0$, experimentally determined field is used and is the incident distribution $A(x,y,0,t)$. Next, with the nonlinearities turned off, Eq. (1) is solved over a distance $\Delta z/2$ for each time slice by use of the two dimensional fast Fourier transform beam propagation method [11,12]. Then diffraction is set to zero and the rate Eqs. (3) and (4) are solved at this z location via finite difference approximations. The values for $N(x,y,t)$ and $\Delta T(x,y,t)$ are used to modify the electric field. Lastly the field is propagated by a distance $\Delta z/2$ with the nonlinearities set to zero in Eq. (1). This process is repeated until the pulse reaches the end of the sample. At the exit of the sample the field is modified by the transmission coefficient and is temporally and spatially integrated to calculate the output energy.

For the ps laser sources, a Gaussian temporal distribution, determined by second harmonic generation autocorrelation, was assumed. For the ns sources the temporal distribution was measured using a fast photodiode and oscilloscope (4 GHz). The pulse shape was normalized and a cubic spline method was used to decrease the number of points while not losing any temporal features. This allowed the actual temporal profile to be used in the model for $A(x,y,z,t)$. For all wavelengths and pulse durations, the actual incident spatial profile was used in the model.

Figure 1 shows the theoretical nonlinear transmission in InP at 1.064 μm as a function of incident irradiance for different laser pulse durations. Here the spatial profile and pulse duration are assumed to be Gaussian and are characterized by r_0 and t_0 which are the Gaussian $HWe^{-1}M$ of the irradiance incident on the sample and temporal profiles respectively.

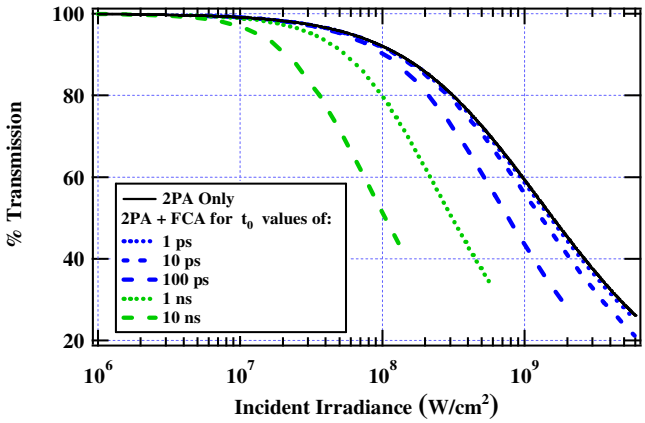

Fig. 1. Theoretical nonlinear absorption in InP at 1.064 μm for different pulse durations. As the pulsewidth increases, contribution from free carrier absorption becomes significant.

The parameters used in Fig. 1 are β = 25.5 cm/GW and σ_{abs} = 1.54 x 10^{-17} cm^2. Recombination lifetimes (τ, $HWe^{-1}M$) are assumed to be long compared to t_0. As seen from the graph, for t_0 below 1 ps free carrier effects can be ignored allowing β to be found directly. However once t_0 begins to exceed ~10 ps, free carrier effects can no longer be ignored. Since the lasers used in this work have pulse durations on the order of 10 ps and longer, both β and σ_{abs} are included in the analysis for all pulse durations.

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3. Linear Transmission and Free Carrier Absorption

Transmission spectra of the samples used in the NLA measurements are shown in Fig. 2. All three samples had anti-reflection (AR) coatings, the Fe doped sample was coated with a single layer AR film and the undoped samples had broadband AR coatings.

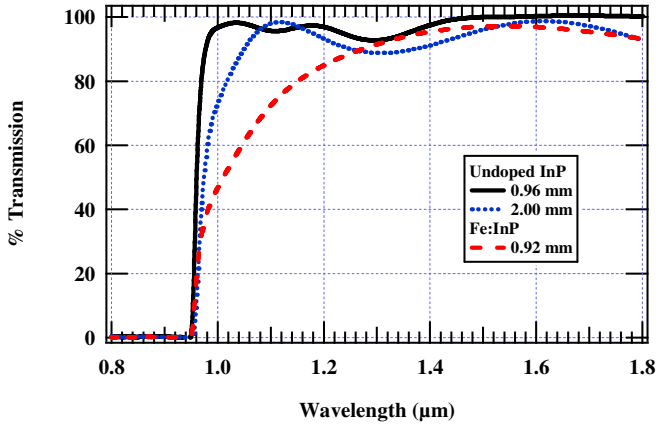

Fig. 2. Transmission spectra of undoped and Fe doped InP samples used for NLA measurements. Undoped samples have multi-layer AR coatings while the Fe sample has a single layer coating.

In addition to nonlinear measurements on undoped and Fe doped (*n*-type) samples, linear measurements on two uncoated, Zn doped (*p*-type) samples of different carrier concentrations were performed. They were 0.39 and 0.35 mm thick with concentrations (*N*) of 3.1 x 10^{-17} and 1.9 x 10^{-18} cm^2 respectively as determined by Hall measurements. The refractive index as a function of wavelength was found using [13]. Linear absorption (α) was determined from the standard etalon transmission expression. Since the samples are *p* type, the free carrier absorption cross section due to holes was determined by $\sigma_h = \alpha / N$. Figure 3 shows the transmission and average σ_h from both Zn doped samples as a function of wavelength. The σ_{abs} values used in the NLA analysis are from contributions of both σ_e and σ_h, although NLA due to holes is the dominant FCA mechanism. The values of σ_h at 1.064 and 1.535 μm were found to be 9.1 x 10^{-18} and 2.2 x 10^{-17} cm^2 respectively.

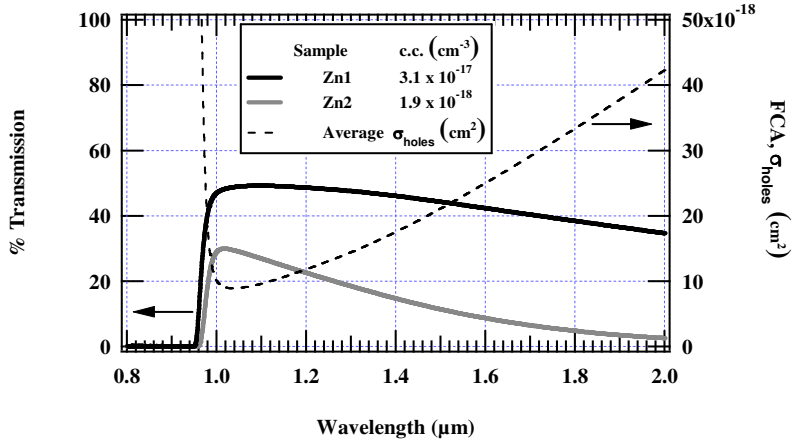

Fig. 3. Transmission spectra of Zn doped InP samples and average hole free carrier absorption cross section.

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4. Experiment and analysis

A mode locked Nd:YAG laser at 1.064 μm (Ekspla PL2134) provided the ps duration pulses. This laser is also frequency doubled to pump a tunable optical parametric generator providing pulses in the 0.7 to 2.4 μm spectrum (Ekspla PG501). Nanosecond duration pulses were obtained from flashlamp pumped, electro-optically Q-switched lasers. A commercial laser was used at 1.064 μm (Quantel Brilliant). Nanosecond measurements at 1.535 μm were done with a laser built in-house with an Er doped phosphate glass laser rod (Kigre, Inc.). Based on the available energy, the beams were loosely focused and the spatial profiles at the beam waists were measured using either a camera or through an *xy* pinhole scan.

The irradiance scan method [5] was used where the sample was placed at the beam waist and the incident energy was varied through the use of a half waveplate and polarizer. A beam splitter was placed prior to the sample and the incident energy was monitored using a pyroelectric energy detector. A second energy detector was placed immediately after the sample in order to capture the total transmitted energy. Significant nonlinear refraction was observed and transmission measurements were verified to ensure that all the transmitted energy was captured. Energy measurements were performed using a ratiometer (LaserProbe, Inc. RM6600 and RJ7620).

Prior to any nonlinear measurements, linearity of the system was verified by ensuring that the ratio of the detector readings remained constant over the entire range of attenuation. Care was also taken to ensure that the detectors were not damaged by using neutral density filters as necessary.

The β and σ_{abs} values were obtained by fitting the experimentally determined energy transmission values to those obtained by simultaneously solving Eqs. (1), (3), and (4) numerically using different values of the parameters. At each wavelength the upper limit of β (β_{max}) was found by fitting the data obtained using the ps duration laser with σ_{abs} set equal to zero. The lower limit of β (β_{min}) was then determined by choosing the minimum value of β for which the ps duration data fitted with the theoretical values obtained with various values of σ_{abs}. For example, at 1.064 μm, if β was chosen to be 20 cm/GW and below, there was no value of sigma for which agreement would be obtained between the theoretical and experimental values. Thus, from picosecond measurements at 1.064 μm it was concluded that β was between 20 and 30 cm/GW. At 1.535 μm the range was found to be between 11 and 23 cm/GW.

Analysis of the ns measurements of total transmission showed that the fitting parameter was the product of β and σ_{abs} that provided the best fit to the experimental data. For the 1.064 μm ns data, the β σ_{abs} product that provided the best fit to the experimental data was 38 ± 15%. At 1.535 μm the product giving the best fit was found to be 105 ± 15%. Keeping the product constant, the β and σ_{abs} values could be varied by factors of 2 or more yet still gave good agreement to the data. This was not the case for the ps data. Although FCA is present at these ps pulse durations, the dominant absorption mechanism is 2PA with only a slight, but nonzero, FCA contribution.

Using the theoretical values in [14], the σ_{abs} values from the transmission spectra as starting values, and with bounds on β, both the ns and ps results were iteratively modeled by varying the NLA coefficients until consistent values were found which fit the data for all the samples at a given wavelength. Recombination lifetimes were assumed to be much longer than the pulse duration for the undoped samples. Good agreement was found for both the 0.96 and 2.0 mm undoped InP samples at both pulse durations and wavelengths. Table 1 summarizes the best fit β and σ_{abs} values.

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Table 1. Values of β and σ_{abs} at 1.064 and 1.535 μm.

λ (μm)	β (cm/GW)	σ_{abs} (cm^2)
1.064	25.5	1.5 x 10^{-17}
1.535	14.6	7.2 x 10^{-17}

Experimental and numerical modeling results of NLA at 1.064 μm are given in Fig. 4 and at 1.535 μm in Fig. 5. For clarity, measurements on the Fe:InP sample are not shown in Figs. 4 or 5. At 1.535 μm, data from the Fe:InP sample are identical to the results from the undoped, 0.96 mm sample. At 1.064 μm the linear transmissions are different between the samples, but accounting for transmission losses, the same set of β and σ_{abs} coefficients gives good agreement to the data.

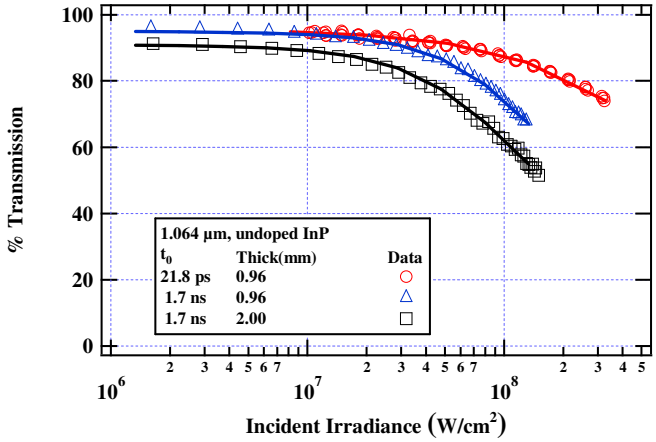

Fig. 4. Nonlinear absorption results (open symbols) at 1.064 μm on undoped InP samples. Solid lines are theoretical results using values of β = 25.5 cm/GW and σ_{abs} = 1.5 x 10^{-17} cm^2 for all data.

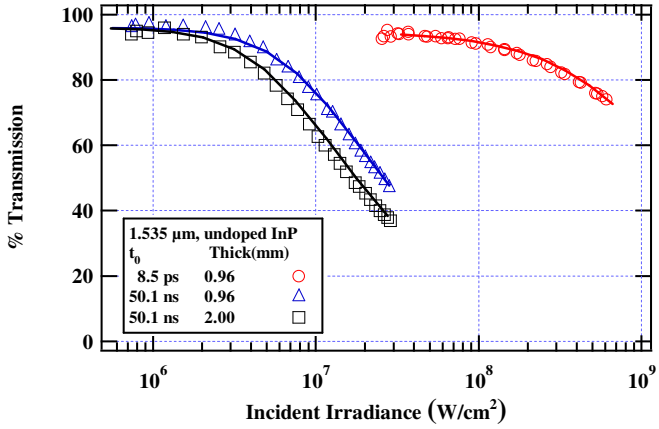

Fig. 5. Nonlinear absorption results (open symbols) at 1.535 μm on undoped InP samples. Solid lines are theoretical results using values of β = 14.6 cm/GW and σ_{abs} = 7.2 x 10^{-17} cm^2 for all data.

The σ_{abs} value from analysis of the 1.064 μm NLA data is in reasonable (1.6 times higher) agreement with the value determined from linear measurements of the Zn doped samples. At 1.535 μm the value determined from the NLA data is a factor of 3.3 times higher. Note that if the value of σ_h obtained from the linear transmission measurement is assumed, the β value that will fit the ns measurements will not be in agreement with the ps results. The inverse is also

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true, if the ps data is analyzed first using σ_{abs} from the linear measurements and the best fit β obtained, this β σ_{abs} pair will not agree with the ns results. The values given here are consistent between the undoped InP samples at both pulse durations as shown in Fig. 5. The apparent difference between the cross sections determined from linear and nonlinear transmission measurements is hypothesized [14] to arise from two sources—(a) additionally allowed absorption by holes that are created away from the zone center where energy conservation can be satisfied, and (b) additional *direct* absorption of photons by the electrons thermally excited into X valley of the conduction band. With increased temperature in NLA experiments, thermal excitation is more probable.

Although NLA data between the Fe doped and undoped InP samples were identical for ps duration pulses, a difference was observed at pulse durations in the ns, for which the Fe:InP showed less NLA compared to the undoped sample. The temporal shape of the laser pulse exiting the sample at high irradiances was monitored. The undoped samples showed asymmetry in the output temporal profile due to free carrier generation and absorption during the pulse. For the Fe doped sample, the output pulse closely followed the incident pulse, indicating a recombination lifetime shorter than the ns laser t_0. Figure 6 shows the input and output temporal profiles for the 0.96 mm undoped and the Fe doped InP samples at 1.535 μm, t_0 = 50 ns.

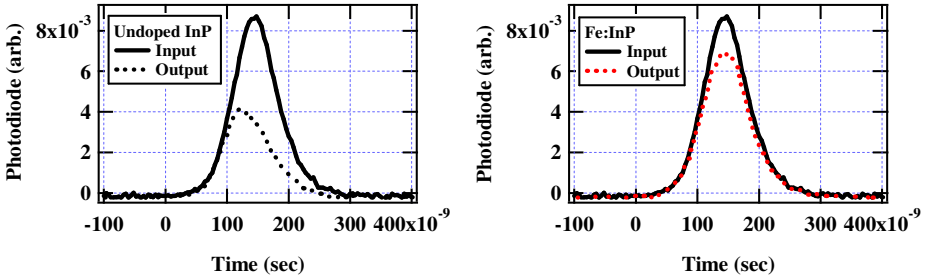

Fig. 6. Output traces for undoped (left) and Fe doped (right) InP samples at similar incident irradiances. Input wavelength is 1.535 μm, t_0 = 50 ns.

Doping of the samples introduces defect states into the material which allow the generated *e-h* pairs to recombine more efficiently, resulting in a decreased recombination lifetime. Keeping the same β and σ_{abs} values as in the undoped samples, the value of the decay time τ that best fit the Fe:InP NLA data was ~3 ns. Figure 7 shows a comparison of the NLA data at 1.535 μm, t_0 = 50 ns for undoped and Fe doped InP of similar thicknesses. Nanosecond results at 1.064 μm are similar.

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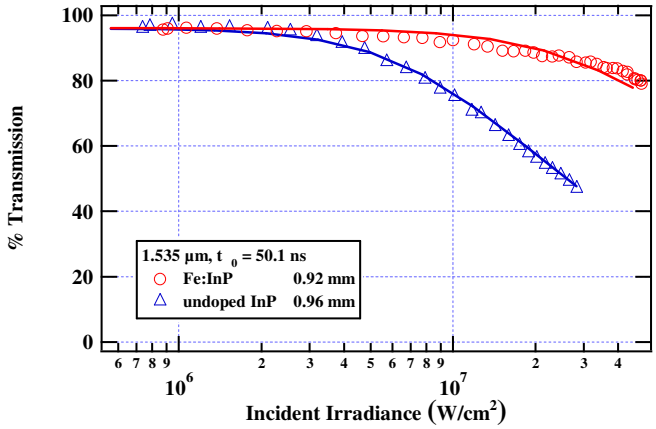

Fig. 7. Nonlinear absorption in undoped (triangles) and Fe (circles) doped InP at 1.535 µm, t_0 = 50 ns. Values of β and σ_{abs} used for both samples are the same. Lifetime in Fe:InP is set to 3 ns and greater than t_0 for the undoped sample.

5. Conclusion

Nonlinear absorption due to two photon and free carrier absorption effects has been investigated in InP. Values for the free carrier absorption coefficient are measured for the first time using the nonlinear transmission method. A self consistent set of β and σ_{abs} have been determined at 1.064 and 1.535 µm using undoped samples. We find a decreased recombination lifetime for Fe doped samples which manifests as a decrease in nonlinear absorption for ns duration laser pulses. The σ_{abs} values obtained through the fitting of modeling result to nonlinear experimental data are about 1.6 and 3.3 times higher than those determined from linear absorption measurements at 1.064 and 1.535 µm respectively, possibly indicating additional carrier absorption at higher irradiances.

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