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14. ABSTRACT The goal of this research is to perform an experimental study of the nonlinear properties of semiconductors in the infrared spectral region to develop a fundamental understanding of their optical nonlinearities. This research will characterize the nonlinear response of promising materials, model the nonlinear resonance, design and model nonlinear optical switching systems, and build prototype switches. These materials are useful for a variety of important AF applications including sensor protection and infrared countermeasures.					
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Semiconductor Optical Nonlinearities in the IR Final Report

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1. Abstract:	page 2
2. Final Report Introduction:	page 2
3. Facilities	page 2
4. The major results under this funding:	page 3
5. Our experimental capabilities include:	page 3
6. Three-Photon Absorption (3PA) in semiconductors	page 4
a. Introduction	page 4
b. Results and Discussion	page 5
c. Conclusions	page 6
d. References for three-photon absorption	page 6
e. Appendix A. giving more details of 3PA	page 7
7. Appendix B. Optical limiting in a thick sample of InSb	page 11

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Abstract:

The goal of this research is to develop a fundamental understanding of the nonlinear optical mechanisms leading to nonlinear absorption and refraction occurring in narrow bandgap semiconductors at IR wavelengths. Experiments performed and facilities developed have all been aimed at accomplishing this goal. For facilities, we use a line tunable, single spatial and temporal mode CO₂ TEA laser along with a picosecond Nd:Yag laser which pumps two optical parametric devices (OPG/A) equipped with difference frequency generators (DFG) allowing wavelengths from 2-18 microns. We added a femtosecond Ti:sapphire system also equipped with multiple OPG/A's and DFG tunable out to 11 microns. Using these different systems covering 6 orders of magnitude in time we can unambiguously determine the physical mechanisms and temporal responses of the optical nonlinearities of narrow bandgap semiconductors. We have published work on two-photon absorption and excited state nonlinearities in InSb and are now publishing work on three photon absorption in multiple semiconductors along with a 4-band K.P based theory with excellent agreement giving scaling rules for this high-order nonlinear response. We also began a collaboration with Partha Dutta at Rensselaer Polytechnic Institute who grows single crystal narrow bandgap semiconductors. This bridges well with our interactions with researchers at Wright Labs. In addition we have gotten considerable interest from staff at Lockheed Martin in this research (Mike Dudzik, Clara Rivera).

Final Report Introduction:

Much of the work involved with this research was in developing facilities that were capable of unambiguously determining the nonlinear response of narrow bandgap semiconductors. Previous published work often had to make large extrapolations and assumptions on limited data taken at only one laser pulsewidth and/or one laser frequency. And we began this research program with a CO₂ laser. We now have multiple sources of IR covering the wavelength range out to 18 microns. In addition we can now cover 6 orders of magnitude in source pulsewidth using optical parametric devices (OPG/A's) combined with difference frequency generation (DFG). Finally, using these versatile sources in combination with closed cycle helium cryostats further increases our capabilities for measuring both the linear and the nonlinear optical (NLO) responses. The experimental techniques we have developed, e.g. the Z-scan, allow us to make accurate and absolute determinations of material constants. This versatility allows us to properly tailor experiments to determine the various components of a materials NLO response. It also allows us to develop models of the nonlinear response that we can test over these broad parameter ranges. Experiments of this type are unprecedented in the nonlinear optics literature. The latest success we have had has been to measure the high order nonlinear response of three-photon absorption, 3PA (a $\chi^{(5)}$ process as opposed to the $\chi^{(3)}$ process of two-photon absorption, 2PA). We performed these measurements on a wide variety of materials including wide bandgap semiconductors in addition to the narrow gap materials and this helped us to determine universal scaling rules for 3PA analogous to scaling rules we had earlier developed for 2PA. This 3PA study also included a careful theoretical analysis of semiconductors using a 4 band Kane model of the band-structure ($k \cdot p$ theory). This model fit the experimentally observed spectra extraordinarily well as shown at the end of this report.

Facilities

Our capabilities in the IR now include:

1. CO₂ TEA laser system, that is line tunable and single longitudinal mode using an intracavity low pressure discharge section. This produces ~150 ns gain-switched pulses tunable in the 9-11 μ m range.
2. Nd:YAG laser (Ekspla picosecond system), pumping two 10 ps OPG/A's tunable from 2200 nm (idler) to 400 nm (signal) with integrated difference frequency generation, DFG, systems providing 2.4 - 10 μ m and 8 - 18 μ m tuning ranges respectively. The pulse repetition rate is 10 Hz and the resulting IR pulses are ~5 ps in duration.
3. Ti:Sapphire laser system (Clark MXR 2010 femtosecond system) operating at a 1kHz repetition rate resulting in tunable IR out to 11 μ m with pulsewidths ~200 fs utilizing several nonlinear frequency mixing attachments including DFG.
4. Helium cryostat for controlling the sample temperature down to 10 K

The major results under this funding are listed below:

- 1) CO₂ laser Z-scans and nonlinear transmittance experiments performed
- 2) Measurements of the nonlinear absorption in InSb
- 3) Measurements of the nonlinear refraction in InSb
- 4) Determination of the role of Auger Recombination
- 5) Modeling of beam propagation in narrow bandgap semiconductors
- 6) Picosecond Experimental facilities further developed
- 7) Picosecond Z-scans and pump-probe performed on InSb
- 8) Femtosecond experimental facilities developed
- 9) Femtosecond Z-scans performed on several samples
- 10) Picosecond Pump-Probe data obtained on InSb
- 11) Temperature dependent measurements on InSb
- 12) Thick sample optical limiting experiments (see appendix B)
- 13) Measurements of three-photon absorption (see appendix A)
- 14) Modeling of three photon absorption using Kane 4-band model

In previous reports we have outlined the efforts at developing an understanding of the 2PA, free-carrier absorption and free carrier refraction in InSb along with an explanation of how Auger recombination on the nonlinear responses. This was the topic of an entire dissertation by Vladislav Dubikovskiy and has now been accepted for publication in Physical Review B. We have also given details of pump-probe measurements and Z-scans on narrow gap semiconductors in previous reports. Here we give a more thorough discussion of 3PA in semiconductors which is the topic of a paper submitted to the upcoming CLEO conference.

Our experimental capabilities include:

1. CO₂ TEA laser system, that is line tunable and single longitudinal mode using an intracavity low pressure discharge section. This produces ~150 ns gain-switched pulses.
 2. Nd:YAG laser (Ekspla picosecond system), pumping two 10 ps OPG/A's tunable from 2200 nm (idler) to 400 nm (signal) with integrated difference frequency generation, DFG, systems providing 2.4 - 10 μ m and 8 - 18 μ m tuning ranges. The pulse repetition rate is 10 Hz and the resulting IR pulses are ~5 ps in duration.
 3. Ti:Sapphire laser system (Clark MXR 2010 femtosecond system) operating at a 1kHz repetition rate resulting with several nonlinear frequency mixing attachments including DFG for the IR tunable out to 11 μ m with pulsewidths ~200 fs.
- Results from the CO₂ system:

We have used temperature dependent studies to look at both the linear and nonlinear response of InSb. The bandgap energy of InSb depends on its temperature and can be tuned

significantly with our closed cycle He cryostat. The linear transmittance follows empirical theories well. This temperature tuning also allows us to tune the bandgap energy so that the 2PA edge can be moved right through the 10 μm region where our CO_2 TEA laser operates. This allows us to do NLO spectroscopy without a broadly tunable laser. By using low energy $\sim 150\text{ns}$ pulses and results from femtosecond and picosecond experiments we have obtained reasonable agreement for experiment for the spectrum of 2PA in InSb. For these pulses all nonlinear responses are dominated by the carrier nonlinearities where the carriers are initially created via 2PA (but the 2PA is not directly observable on this time scale and must be inferred and/or measured using shorter pulses).

Results from picosecond DFG experiments:

We find that even going to pulses nearly 2 orders of magnitude longer (several picoseconds) that the nonlinear response is still dominated by the effects of 2PA generated free carriers. This is true both for the nonlinear absorption as well as the nonlinear refraction.

Results from femtosecond DFG experiments:

Using femtosecond IR pulses, which are nearly 6 orders of magnitude shorter than our longest pulses ($\sim 150\text{ns}$), the effects of free carriers are still noticeable and need to be taken in to account in fittings. This is the case even though the number density of carriers scales linearly with the pulsewidth, i.e. longer pulses create more carriers for a fixed irradiance because the pulse continues to build up carriers until it becomes comparable to the carrier decay time which is in the nanosecond regime. Experiments have been performed in the 8-11.5 micron regime to do spectroscopy of both nonlinear absorption and refraction. Also – as outlined at the end of this report we used femtosecond pulses to measure 3PA in several materials including InSb.

Results from temperature dependent studies:

The nonlinear absorption in undoped InSb is measured by temperature dependent transmittance studies using a CO_2 laser. A numerical model considering two-photon and free-carrier absorption along with recombination of carriers is used to fit the data. Measurements of sample from Partha Dutta have also been performed.

The results determined from the combination of the experiments covering 6 orders of magnitude in pulsewidth form a consistent picture of the nonlinear response of InSb which greatly facilitates future experiments on new materials.

Three-Photon Absorption in semiconductors (summary)

1. Introduction

Multiphoton processes in zinc-blende semiconductors have been extensively studied in the past. The theoretical magnitude and spectrum of the 2PA coefficient have been confirmed experimentally. The theoretical models employed ranged in complexity from simple 2-parabolic band models [1,2] to complex multi-band models which were used to predict the anisotropy of nonlinear coefficients [3]. Very good agreement with the expected bandgap and wavelength scaling has been shown, allowing prediction of 2PA coefficients in new materials using only a few parameters such as bandgap energy and index of refraction [4].

In contrast, data on three-photon absorption (3PA) is scarce and previous work shows measurements over narrow spectral ranges. Simple models have been used to predict the behavior of 3PA but there has been disagreement in the results. Moreover, the trends in the

newly available data [5] were not properly explained by the theories previously available, since the value of the 3PA coefficient was observed to increase as the wavelength gets closer to the 2PA band edge where a decrease was predicted theoretically.

2. Results and Discussion (summary)

It was previously predicted that the magnitude of the 3PA coefficient scales with E_g^{-7} [1,2] and has been observed experimentally by performing a series of measurements on semiconductors with bandgaps spanning from 0.18 eV to 3.9 eV [6]. Due to disagreements in the predicted spectra by the available two-band theories we decided to use a more realistic approach for the specific zinc-blende structure of the semiconductors studied. The model used for our theoretical calculations was developed by Kane [7] and is based on $\mathbf{k} \cdot \mathbf{p}$ theory with spin-orbit interaction. The resulting band structure consists of a conduction band, light-hole and heavy-hole valence bands degenerate at $k = 0$, and a split-off valence band. The model gives a full description of the electronic wave-functions for the four bands which allows for calculation of momentum matrix elements for transitions between any of the bands at each point in k space. In order to calculate the 3PA coefficient we made use of second-order perturbation theory and did a summation over all contributions arising from transitions from all the valence bands to the conduction band including all possible intermediate states.

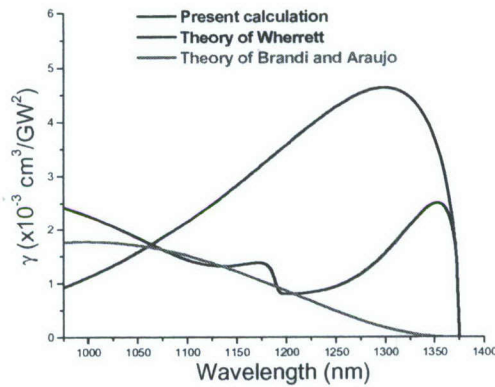


Figure 1. Comparison of present calculations to available theories in absolute theoretical values.

The results obtained for ZnSe are shown in Figure 1. The most noteworthy features of the 3PA spectrum are the peak at 1350 nm close to the band edge and the contribution due to split-off band transitions observed around 1200 nm. The dip in the spectrum appears because of quantum interference since there are multiple possible paths for the same pair of initial and final states. Figure 1 also shows the prediction of two previous theories of Wherrett and Brandi and Araujo [1,2].

We used a femtosecond tunable laser to perform Z-scans on a sample of ZnSe with a zinc-blende structure. The nonlinear measurements were performed in the wavelength range from 1000 nm to 1400 nm, covering the entire range between the 2PA and 3PA band edges. The values obtained are estimated to have absolute error of approximately 40% with relative errors shown in Figure 2(b). At each wavelength, several Z-scans were performed at different input energies and the analysis showed that we were indeed observing a pure 3PA process, with no effects from free carriers. Also, at each wavelength, the setup was used to measure the known 2PA coefficient in a sample of CdTe in order to check for calibration accuracy.

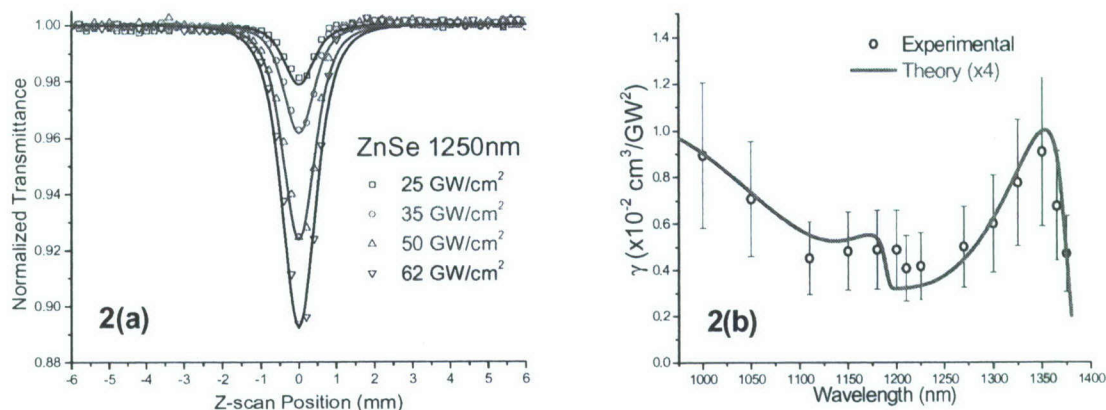


Figure 2. (a) Z-scans taken at 1250 nm, (b) Experimental values vs. scaled theory.

Figures 2(a) and 2(b) respectively show typical experimental data and the obtained spectrum together with the theoretically predicted one. As can be seen from the plots we obtained good agreement between the observed spectral shape and theory; however, there is a disagreement in the absolute values of a factor of 4. The theoretical values are lower than experimental so that in Fig. 2(b) the theory is multiplied by this factor. Note in Figure 1, all three theories show similar magnitudes for 3PA. Nevertheless, we can clearly identify in the experimental spectrum the features predicted theoretically. To the best of our knowledge, this is the first time that such a detailed structure has been obtained experimentally.

3. Conclusions

We used perturbation theory and Kane's 4-band model for zinc-blende structures to predict the 3PA spectrum in ZnSe. Our experimental data shows good agreement for the theoretically predicted spectral shape and with experimentally obtained literature values. We have measured 3PA in several other materials and are currently working on measuring their spectra.

More details on the 3PA experiments are given in Appendix A

References for three-photon absorption

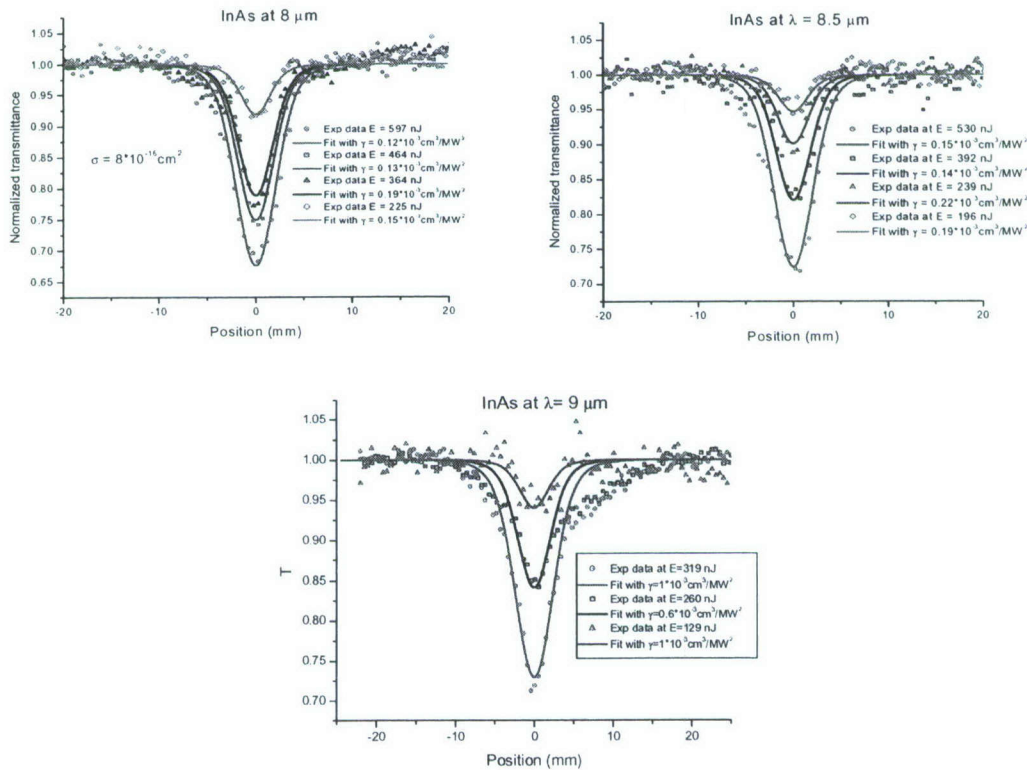
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Appendix A – 3PA

There are several conclusions that can be drawn from the femtosecond experiments: fsec pulses nearly eliminate free-carrier effects from the nonlinear absorption experiments; the free-carrier component to the nonlinear refraction is still important and cannot be neglected; experiments to measure nonlinear refraction may prove difficult to interpret in the presence of large absorptive nonlinearities.

Recent measurements on InSb at very low temperatures (15K) with a nsec CO₂ laser suggested 3PA as a mechanism for nonlinear absorption. Since the predicted magnitude for 3PA is increasing with wavelength in the range studied we wanted to check whether 3PA is a factor in the nonlinear experiments carried out at room temperature. Since the wavelengths required were long and not easily accessible we turned our attention to InAs. We attempted measurements (as shown below) between 8 and 9 microns. The experiments themselves pose more difficulties since we're dealing with a higher nonlinear process and the quality of the n-doped polycrystalline sample wasn't great. Even though we were able to fit the data and extract values for the 3PA coefficient at those wavelengths, it became clear that getting a spectrum with easily identifiable features may be difficult because of the error bars associated with our experiments.

We then decided to do a more comprehensive study of 3PA by doing measurements in a large number of semiconductors with wavelengths in a much broader spectrum. The measurements carried out supported available scaling theories for 3PA, which predicted a dependence on the 7th power on the bandgap energy.



Measurements of higher order nonlinearities in InSb, e.g. three-photon absorption (3PA) plus free-carrier absorption at 10K have also been performed. In order to confirm available 3PA theory we also measured 3PA in ZnS, ZnO, ZnSe, CdS, ZnTe, CdSe, CdTe, GaAs using femtosecond pulses.

Material	E_g [eV]	λ [μ m]	N	E_p [eV]	Data α_3 [cm^3/GW^2]	Theory $K_3 = 25.1 \alpha_3$ [cm^3/GW^2]
ZnS	3.66	0.8	2.3	20.4	0.0014	0.0012
ZnS	3.66	0.85	2.3	20.4	0.0018	0.0017
ZnO	3.2	1.05	2.05	21	0.0047	0.0106
ZnO	3.2	1.06	2.05	21	0.0220	0.0112
ZnSe	2.67	1.05	2.48	24.2	0.0078	0.0090
ZnSe	2.67	1.06	2.48	24.2	0.015	0.0099
ZnSe	2.67	1.2	2.48	24.2	0.0074	0.0211
ZnSe	2.67	1.3	2.48	24.2	0.0075	0.0290
CdS	2.42	1.06	2.34	21	0.015	0.009
CdS	2.42	1.2	2.34	21	0.011	0.022
ZnTe	2.26	1.3	2.8	19.1	0.02	0.028
CdSe	1.74	1.5	2.5	21	0.24	0.085
CdTe	1.44	1.75	2.7	20.7	1.2	0.19
GaAs	1.42	1.75	3.4	25.7	3	0.13
InAs	0.36	8	3.42	21	200	4200
InSb	0.235	10.6	3.95	21	120000	18000

Table 1. Values of material constants, 3PA coefficients from our data and theoretical prediction.

Values obtained for 3PA show general agreement with E_g^{-7} as predicted by both Wherrett and Brandi & de Araujo. The value obtained from modeling nanosecond data is in agreement with extrapolations using data for other semiconductors and theoretical bandgap scaling. We also measured 3PA spectra for ZnS and ZnSe which showed a need for a more detailed theoretical approach. We decided to develop a 3PA theory using four bands instead of two and include all possible transitions instead of assuming that certain transitions were dominant. We have obtained excellent agreement between 3PA spectral data and our four band theory for ZnSe. Figure 7 shows the bandgap scaling of the 3PA for the materials studied as compared to the theory of Wherrett.

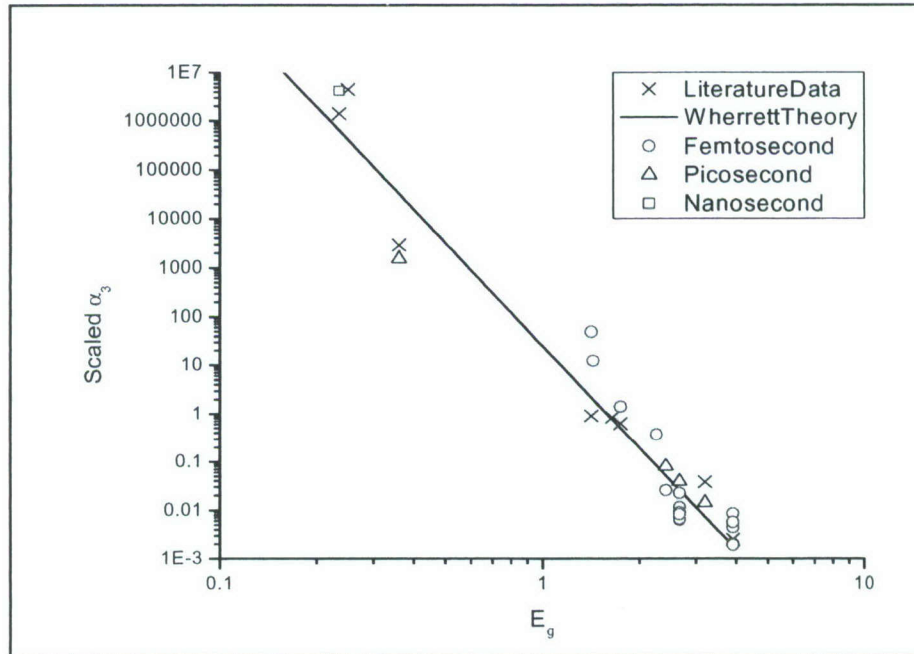


Fig 7. Log-log plot of the scaled 3PA coefficient, α_3 , versus E_g . For some materials we measured α_3 at several wavelengths. The fact that they do not all sit at the same point shows that the spectral function does not completely explain the data.

For ZnSe in particular, we were able to take data in a range of wavelengths spanning over the entire part of the 3PA spectrum not overlapped with the 2PA spectrum. At this point it became obvious that the spectral dependences predicted by either of the available theories aren't able to satisfactorily explain the experimental data.

The theories advanced by Wherrett and Brandi&Araujo started by assuming 2 parabolic bands. The former used a perturbative approach and considered the allowed-allowed-allowed transitions to dominate, while the latter used tunneling theory to predict the 3PA spectrum. This is practically equivalent to considering only the allowed-forbidden-forbidden type of transitions from the point of view of perturbation theory. Needless to say, this leads to very different spectral functions even though the predicted values are within the same order of magnitude.

In order to obtain a more accurate theoretical description we used a more realistic 4-band model as proposed by Kane for zinc-blende structures. One conduction band and 3 valence bands (heavy-hole, light-hole and split-off) are used and a complete description of the band's shapes and wavefunctions for the electronic states are given. We write the interaction Hamiltonian as

$$H_{opt} = \frac{e}{m_0 c} \mathbf{A} \cdot \mathbf{p}$$

and using perturbation theory, we calculate the 3PA rate as

$$W_3 = \frac{2\pi}{\hbar} \sum_{v,c} \left| \sum_{i,j} \frac{\langle \Psi_c | H_{opt} | \Psi_j \rangle \langle \Psi_j | H_{opt} | \Psi_i \rangle \langle \Psi_i | H_{opt} | \Psi_v \rangle}{(E_{jv}(k) - 2\hbar\omega)(E_{iv}(k) - \hbar\omega)} \right|^2 \delta(E_{cv}(k) - 3\hbar\omega)$$

Making use of the identity $\gamma(\omega) = 3\hbar\omega W_3 I^{-3}$ we can then easily calculate the 3PA coefficient at any frequency. The summation is made over all possible initial states (from any of the valence bands), all possible final states (in conduction band) and taking into consideration all possible intermediate states denoted by i and j (any state for the matching k value in any of the bands). This approach takes into account the most realistic description of the bands including non-parabolicity effects and using the non-zone center wavefunctions one can accurately determine the transition rates for any transition involving any pair of bands.

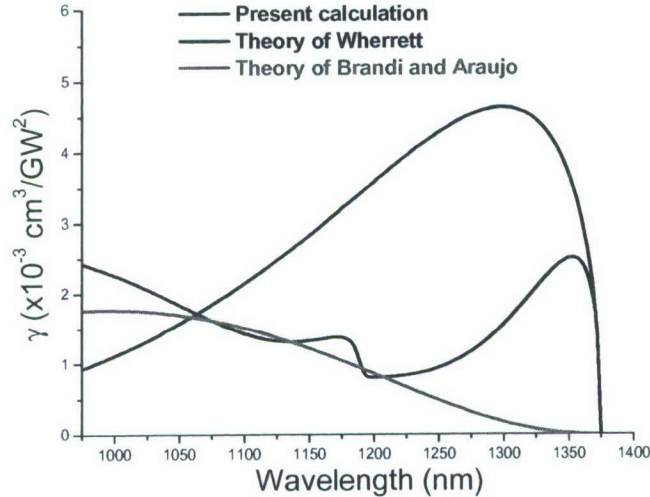


Figure A1. Predictions of various theories for 3PA. Our 4-band model is in black with the turn on of the split-off band showing details in the spectrum.

The comparison of our calculations to the available theories is presented in the plot of Fig. A1. Features of importance here are the dip in the spectrum and the turn on of the contribution of the transitions from the split-off band, particularly obvious because it's around the dip in the spectrum, wavelength wise.

In order to verify the predicted spectrum experimentally we used a femtosecond OPG and made a series of measurements between 1 micron and 1.4 microns with step sizes as small as 20 nm. For each wavelength Z-scans were taken at a few energies and the curves were fitted individually to make sure that the 3PA was the only nonlinear process present. Below are shown typical data at 1.25 microns and a plot of the obtained experimental spectrum together with our theoretical predictions. Overall, there's a discrepancy of about a factor of 4 in the absolute values between experiment and theory. Therefore, for easier comparison we scaled our predicted theoretical values by this factor.

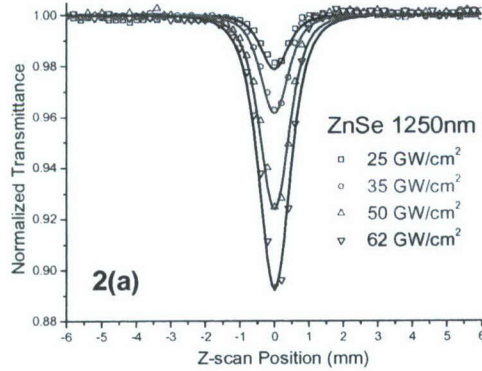


Figure A2. Examples of Z-scan data at a single wavelength on ZnSe at several input energies using femtosecond pulses.

As it can be seen in Fig. A3, there is excellent agreement for the shape of the spectrum. Also a couple of other publications seem to suggest a similar shape. Similar measurements on other semiconductors are underway to produce full spectra. This leads to the possibility of predicting the spectral features on the 3PA in semiconductors using only few parameters of bandgap, split-off energy, Kane momentum parameter and refraction index.

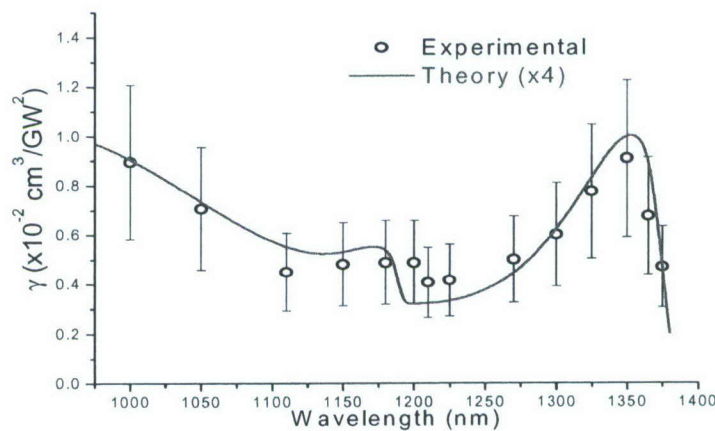


Figure A3. Spectral dependence of the 3PA in ZnSe, experiment (circles) and theory (solid line). The theory has been scaled vertically by a factor of 4.0.

Appendix B. Use of thick optical materials in optical limiting geometries in the IR

One of the goals of this work has been to test the performance of an InSb thick limiter. The 1cm thick InSb sample has been mounted with an offset sample holder in the Helium cryostat to allow focusing on the back surface using an F3 focusing geometry. The alignment process for this experiment is extremely difficult and time consuming. Some preliminary results have been obtained using a F9 focusing geometry and the setup is shown in Fig. B1.

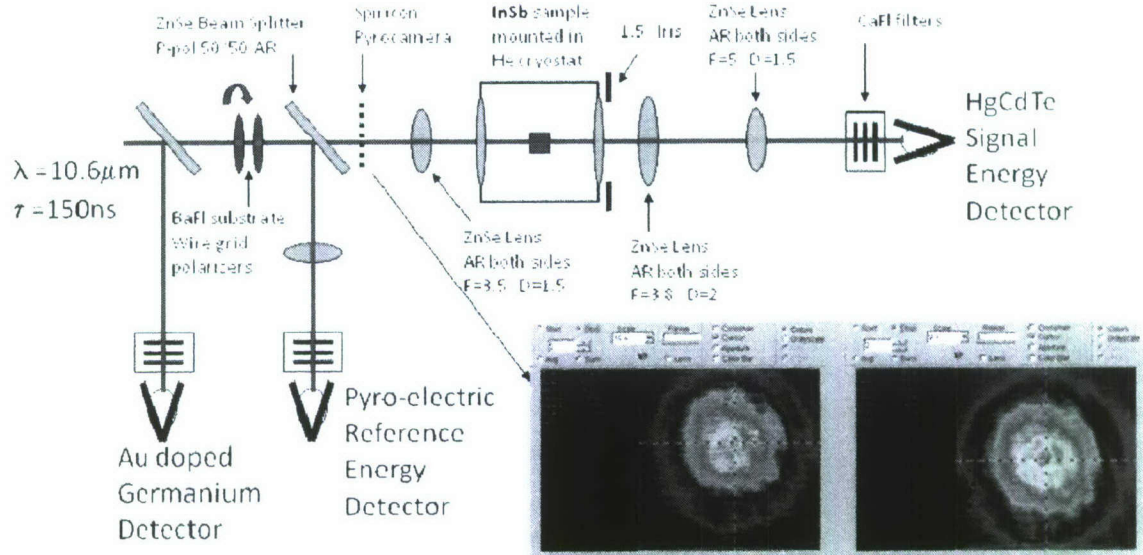


Figure B1. Experimental schematic for thick sample limiting experiment using F=3.5”
D≈10mm F# ≈ 8.89.

Using the F9 system the normalized transmittance dropped to 50% at 225nJ at 80K and the damage threshold was observed to be approximately 500μJ therefore demonstrating a dynamic range of greater than 2000. According to the scaling relations developed previously by our group using a thick ZnSe sample the dynamic range for an equivalent F3 experiment is expected to be 160,000. Experiments using a F3 setup are underway, future work will focus on modeling.

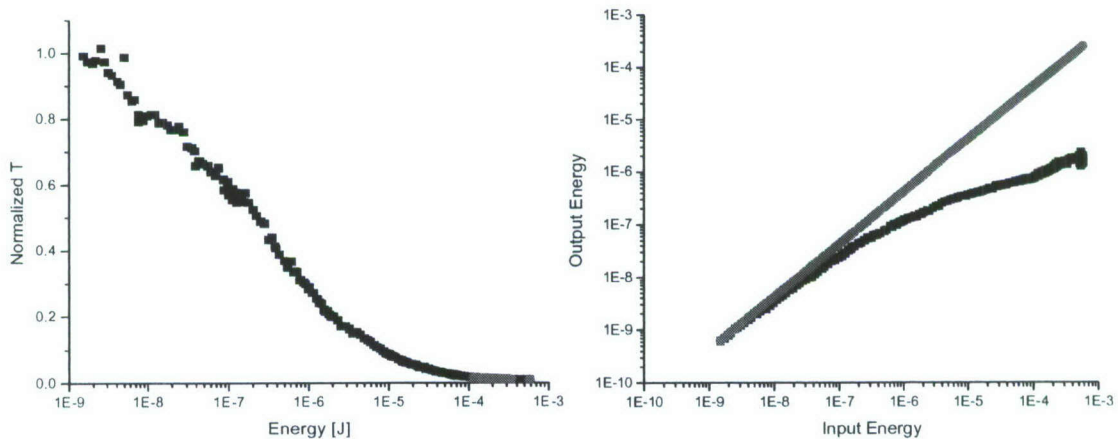


Figure B2. Experimental data for thick sample limiting experiment using F=3.5” D≈10mm
F# ≈ 8.89.

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