Multiphoton absorption spectra of ZnO

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Motivation

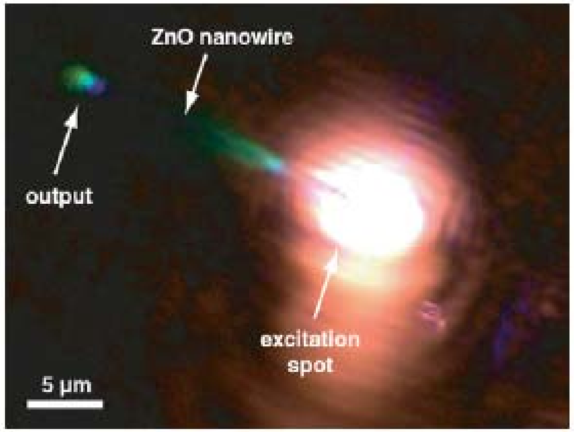
ZnO

- electronic devices
- near-UV optoelectronic devices (light emitters and detectors)
- nanowires

Recently

- nonlinear excitation of ZnO nanowires

study ZnO nonlinear absorption



Tobias Voss, Nanotechnology **20** (2009)

Outline

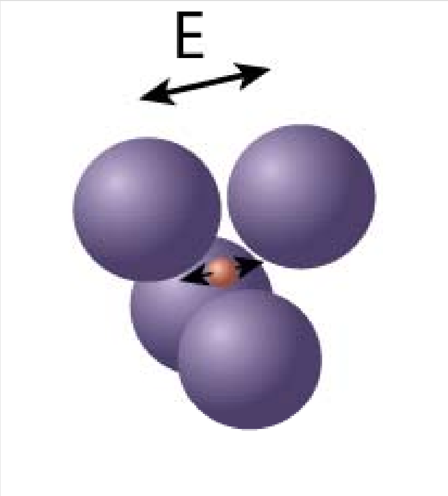
Nonlinear Optics

Experimental Setup

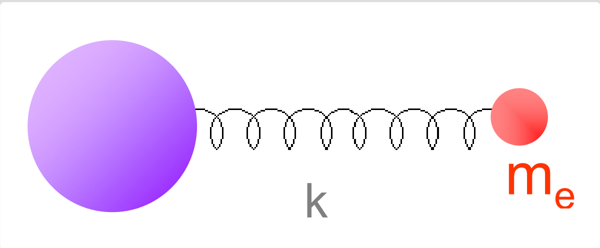
Results and Discussion

Summary

Linear Optics



harmonic oscillator

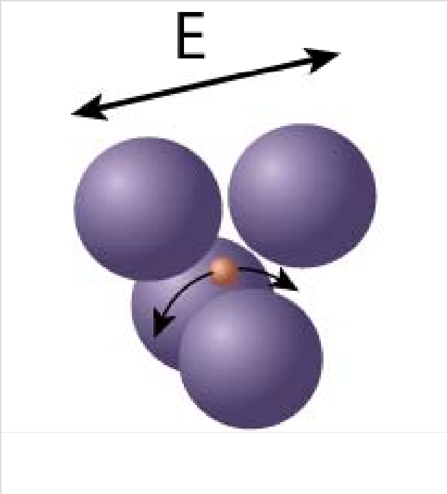


$$E_{rad.} << E_{inter.}$$

linear response

$$P = \chi E$$

Nonlinear Optics



anharmonic oscillator

high light intensity

$E_{rad.} \sim E_{inter.}$

nonlinear response

$$\vec{P} = \chi^{(1)} . \vec{E} + \chi^{(2)} : \vec{E}\vec{E} + \chi^{(3)} \vdots \vec{E}\vec{E}\vec{E} + \ldots$$

Nonlinear Optics

Multiphoton absorption

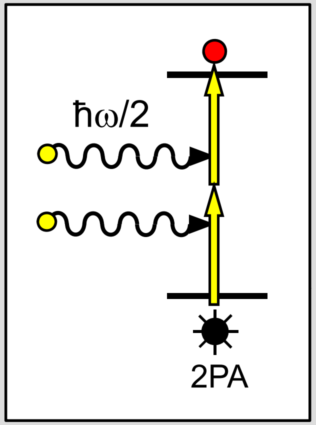
$$\vec{P} = \chi^{(1)}\vec{E} + \chi^{(2)}:\vec{E}\vec{E} + \chi^{(3)}\vdots\vec{E}\vec{E}\vec{E} + \chi^{(4)}\vdots\vec{E}\vec{E}\vec{E}\vec{E} + \chi^{(5)}\vdots\vec{E}\vec{E}\vec{E}\vec{E}\vec{E} + \ldots$$

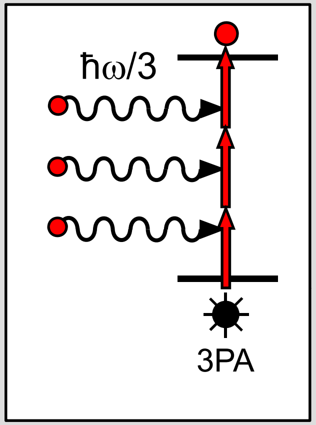
$$\mathrm{Im}\left[\chi^{(3)}\right] \qquad \mathrm{Im}\left[\chi^{(5)}\right]$$

$$\alpha = \alpha_0 + \beta I \qquad \alpha = \alpha_0 + \gamma I^2$$

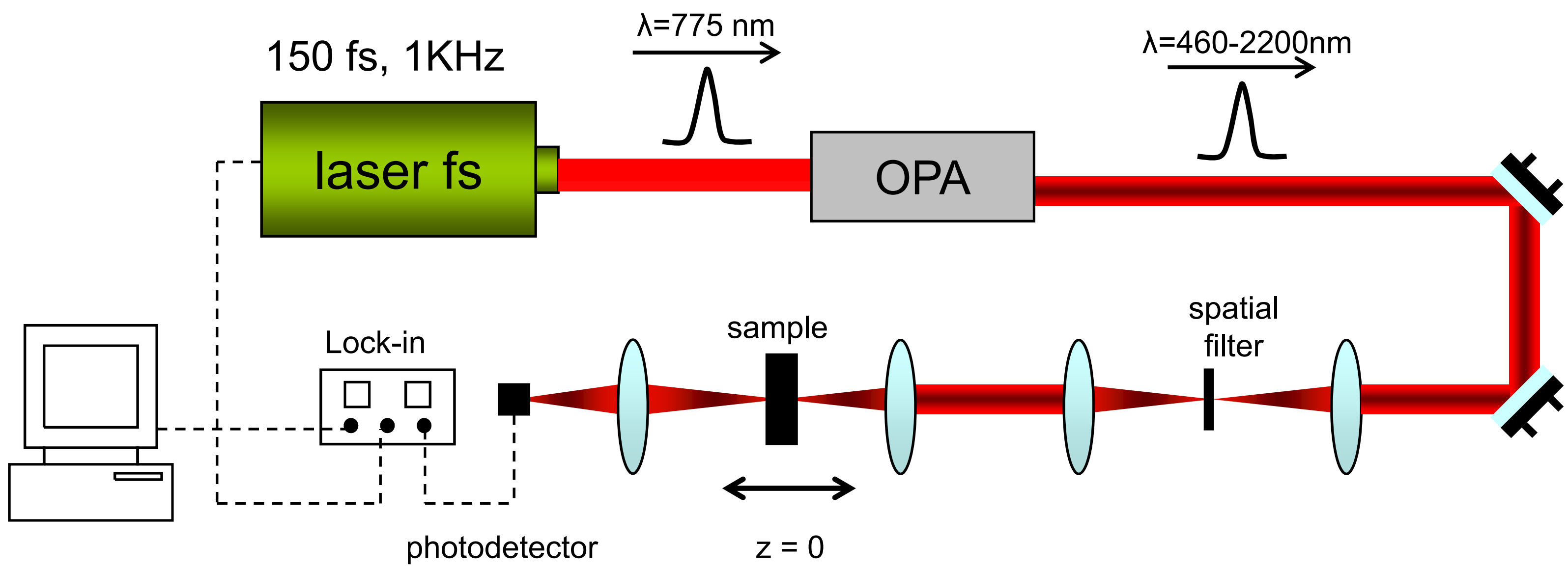
2PA coefficient

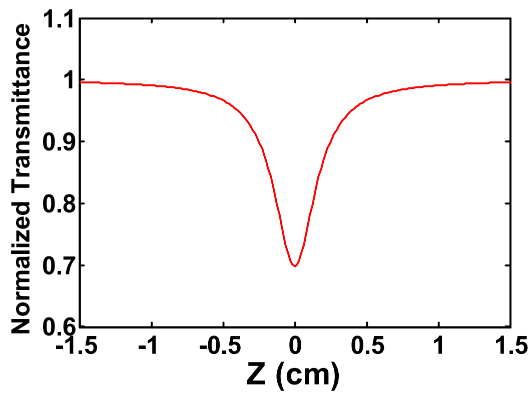
3PA coefficient





Z-scan setup



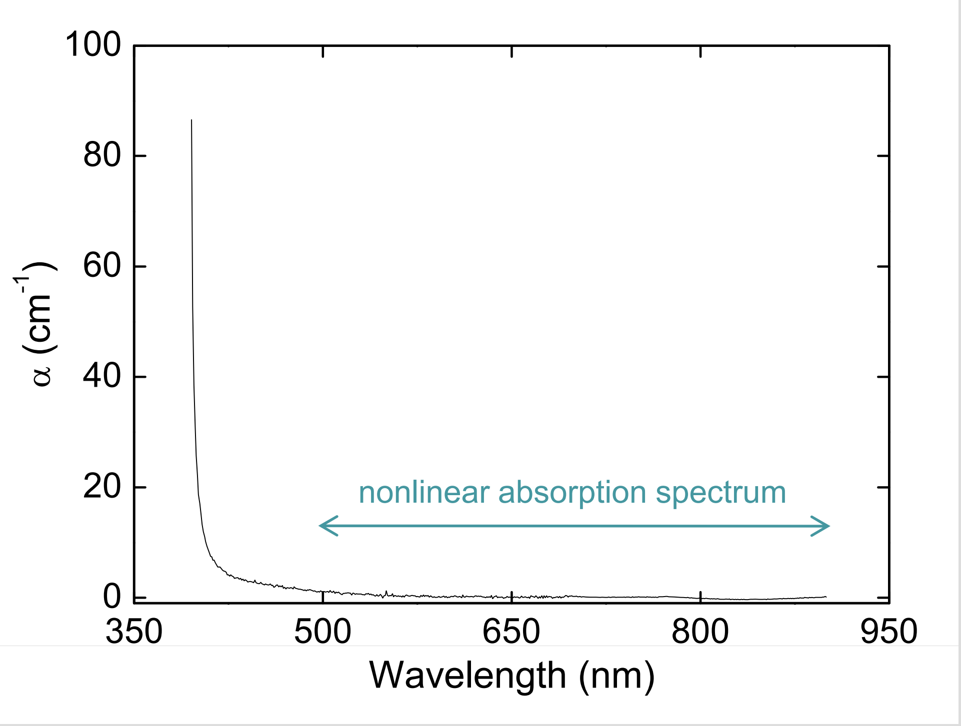


$$T(z) = \frac{1}{\sqrt{\pi}\, q_o(z,0)} \int_{-\infty}^{\infty} \ln\left[1 + q_o(z,0) e^{-\tau^2}\right] d\tau$$

$$q_o = \beta I_o L \left(1 + \left(z^2 / z_o^2\right)\right)^{-1}$$

Results – linear absorption

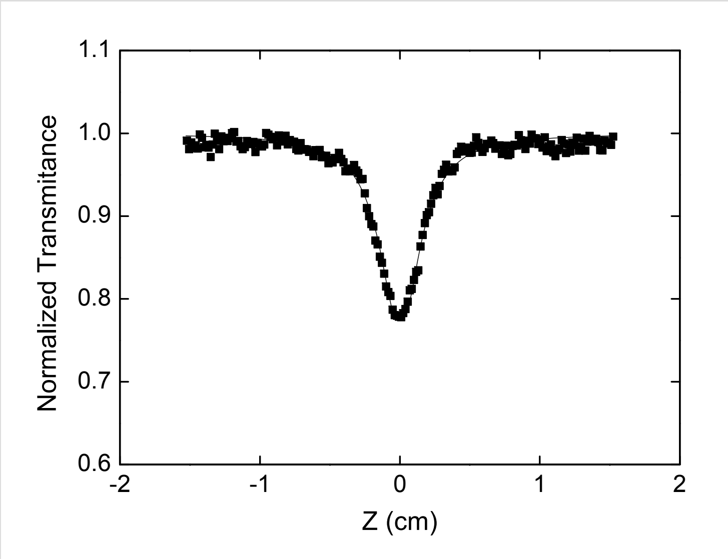
linear absorption of ZnO



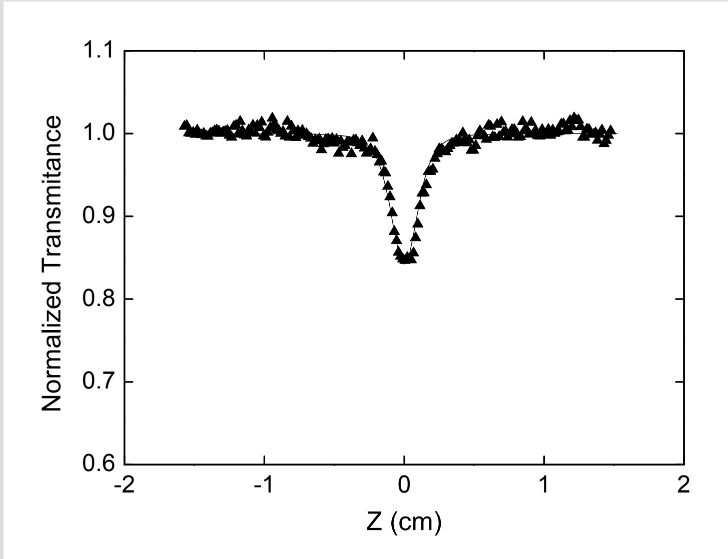
Results – Z-scan

Z-scan curves

730 nm – 2PA

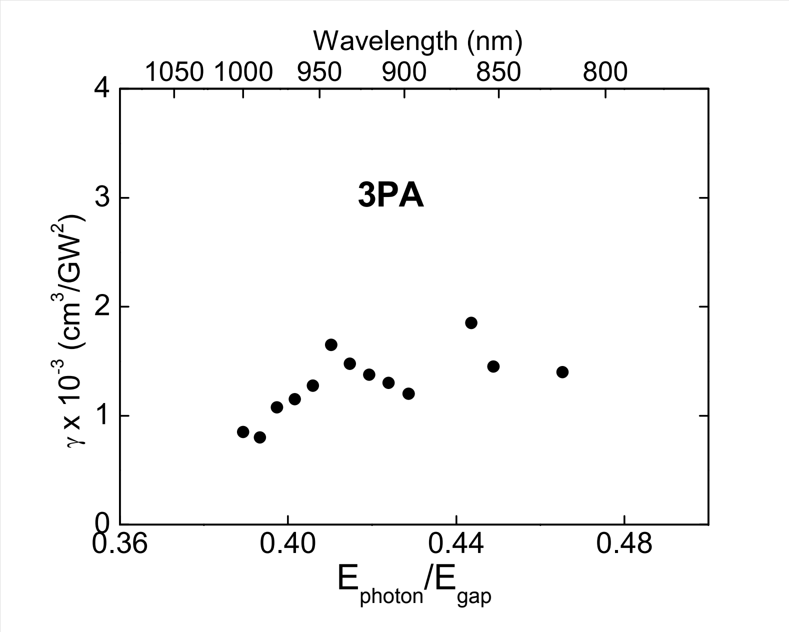


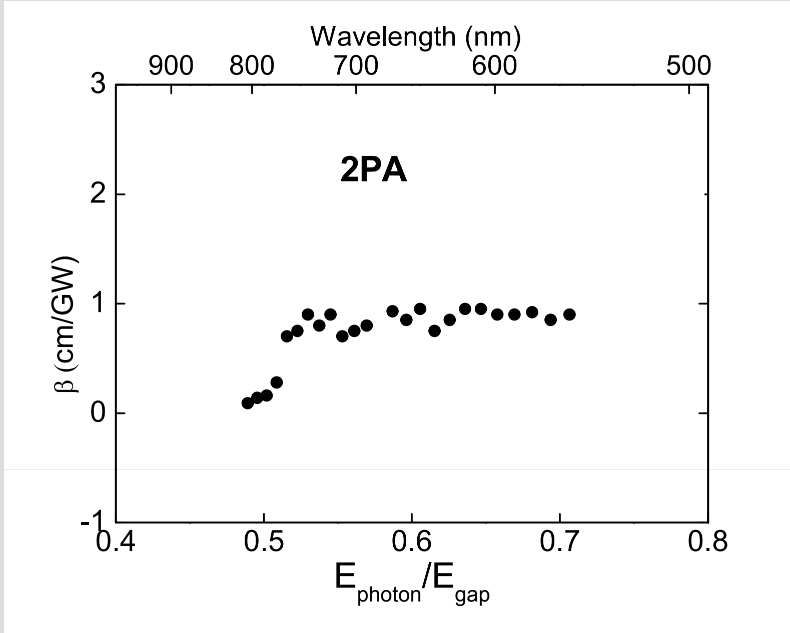
850 nm – 3PA



solid line: fit of the Z-scan curve to obtain β and γ

Results – nonlinear spectra





Results: Multiphoton spectra

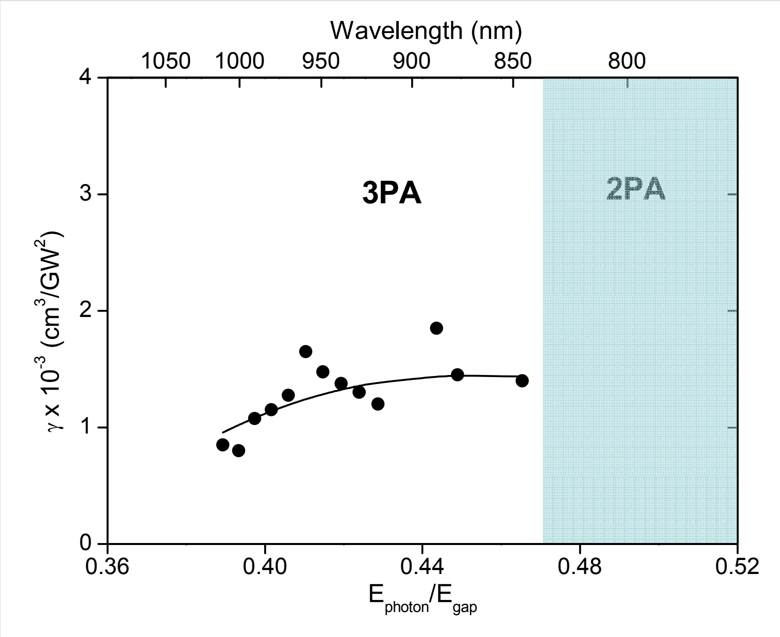
2PA and 3PA spectra model

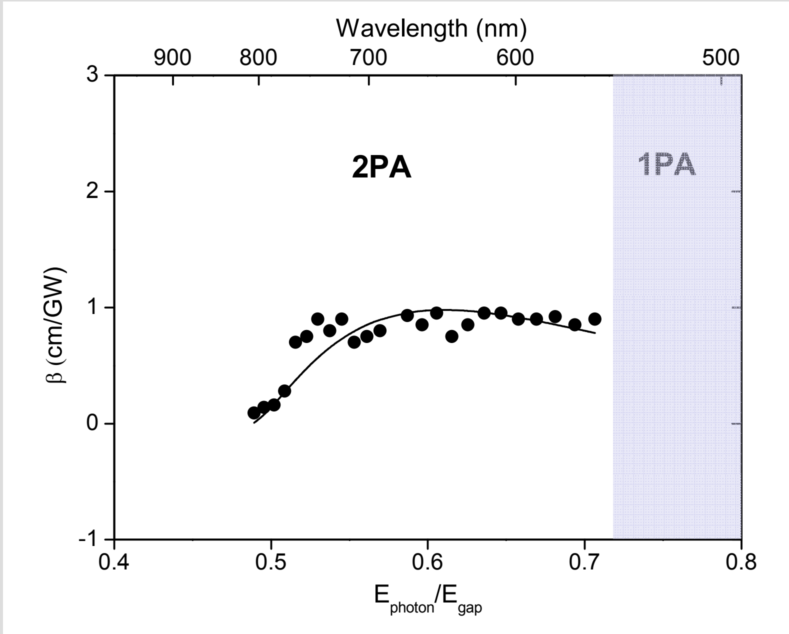
$$\beta = \frac{2^{5/2}(2\pi)}{4}\left(\frac{e^2}{\hbar c}\right)^2 \hbar^2 \left(\frac{m^{3/2}}{m_1^2 n_0^2 E_g^{5/2}}\right)\left(\frac{\left(2E_g/E_{photon}\right)-1}{\left(2E_g/E_{photon}\right)^5}\right)^{3/2}$$

$$\gamma = \frac{2^{9/2}3^{11}\pi^2}{20}\left(\frac{e^2}{\hbar c}\right)^3 \hbar^5 \left(\frac{m^{5/2}}{m_1^4 n_0^3 E_g^{11/2}}\right)\frac{\left(3E_g/E_{photon}\right)^{5/2}-1}{\left(3E_g/E_{photon}\right)^9}$$

Multiphoton absorption coefficients in solids: a universal curve; Brandi and Araujo - J.Phys. C: Solid State Phys. **16,** 5929-5936 (1983).

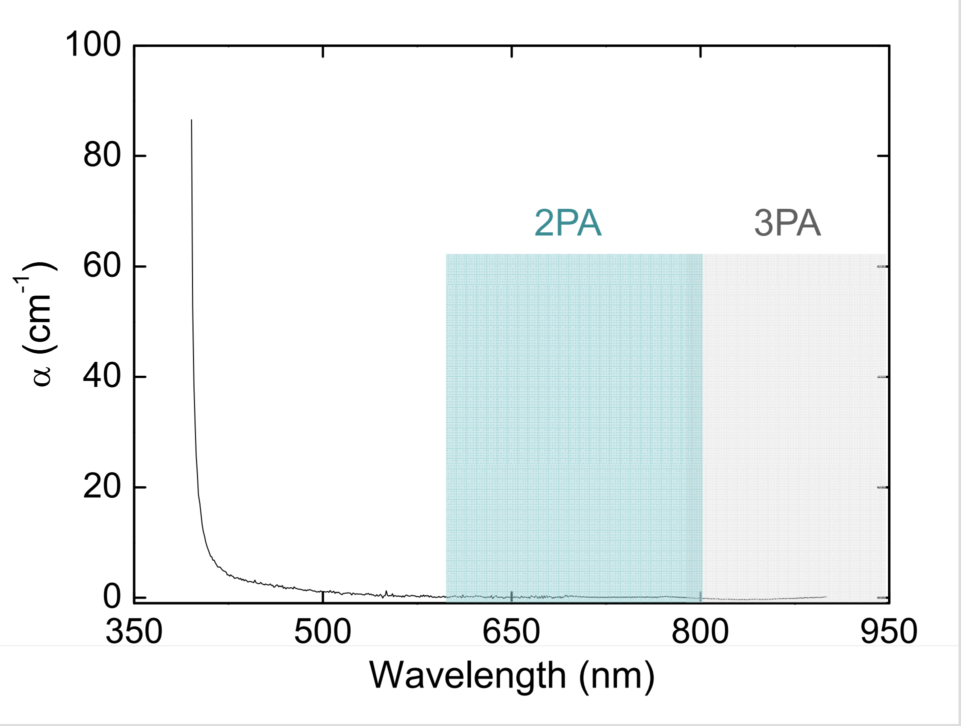
Results – nonlinear spectra





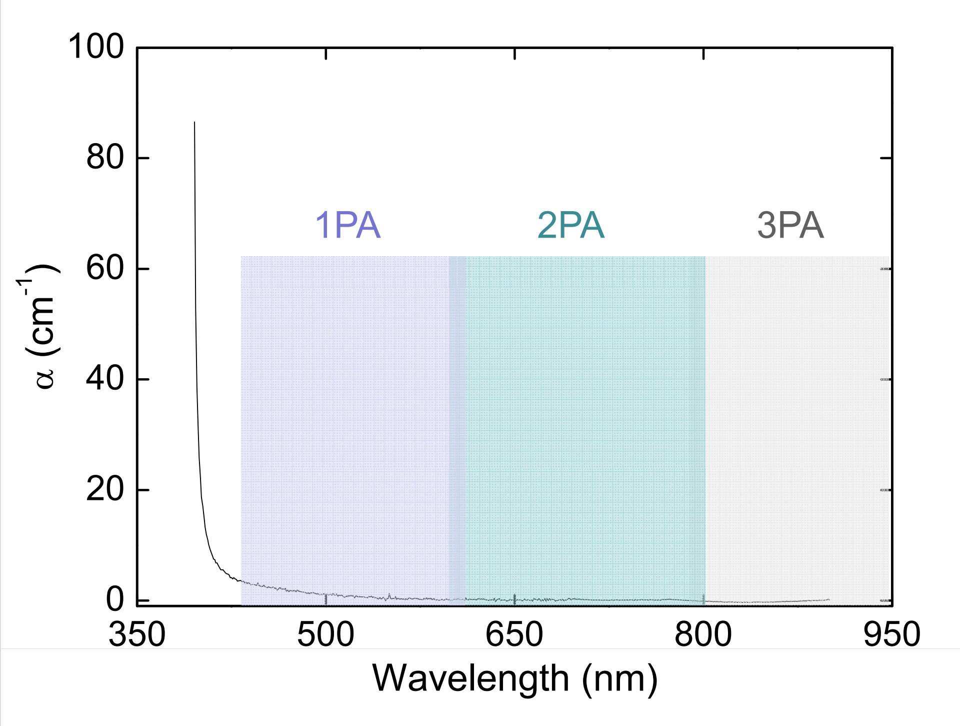
Results

nonlinear absorption regions



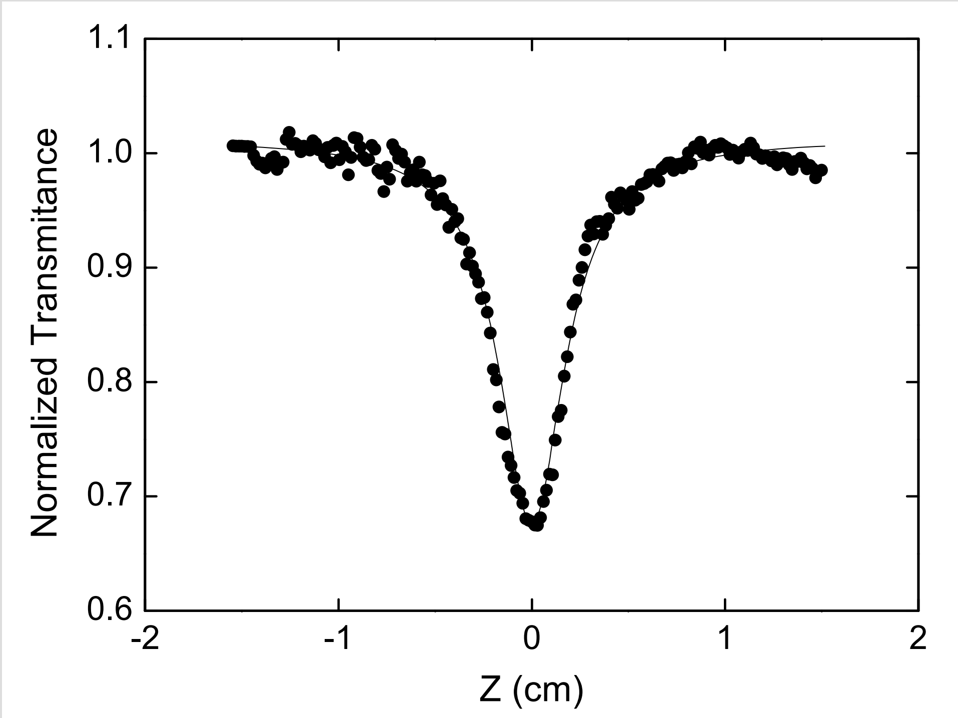
Results

one photon absorption region



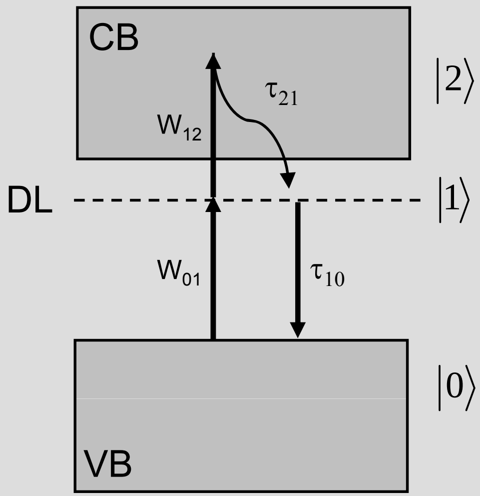
Results – Z-scan for 1PA

500 nm



solid line: fit of the Z-scan curve for a 1PA process

Results - Reverse Saturable Absorption



Rate equations:

$$\frac{dn_0}{dt} = -w_{01}n_0 + \frac{n_1}{\tau_{10}}$$

$$\frac{dn_1}{dt} = +w_{01}n_0 - w_{12}n_1 - \frac{n_1}{\tau_{10}} + \frac{n_2}{\tau_{21}}$$

$$\frac{dn_2}{dt} = +w_{12}n_1 - \frac{n_2}{\tau_{21}}$$

$$\alpha(t) = N\left\{n_0(t)\sigma_{01} + n_1(t)\sigma_{12}\right\}$$

reverse saturable absorption
RSA

excited state absorption cross-section higher than the ground state one

excited state effective cross-section:

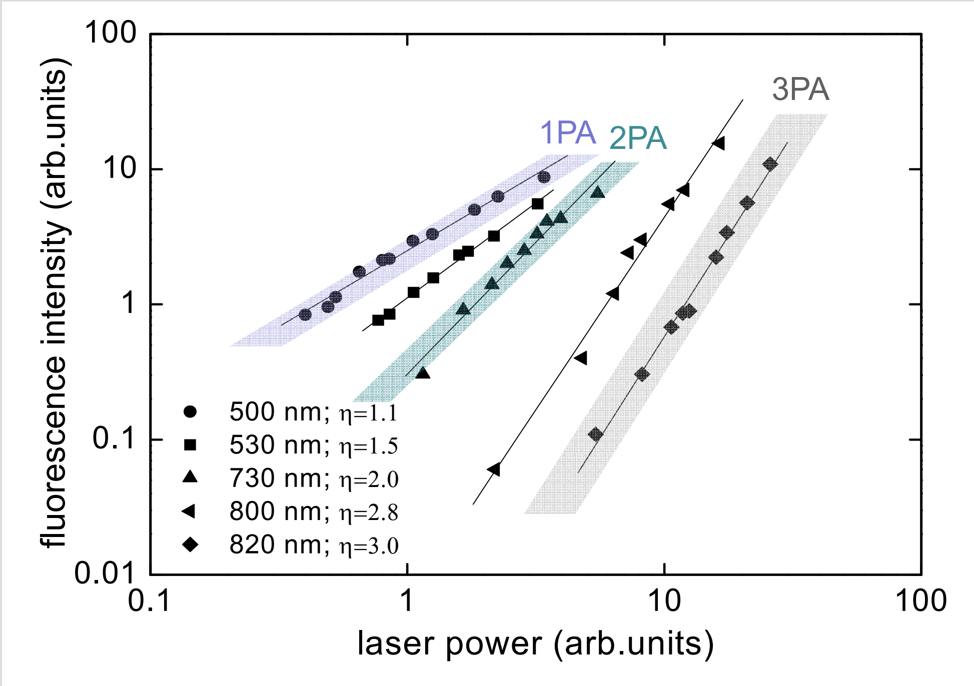
$$\sigma_{12}^{eff} = (14 \pm 2)x10^{-16}\,cm^2$$

$$\tau_{10} = 2.8ns$$

$$t_{21} \approx 50\,fs$$

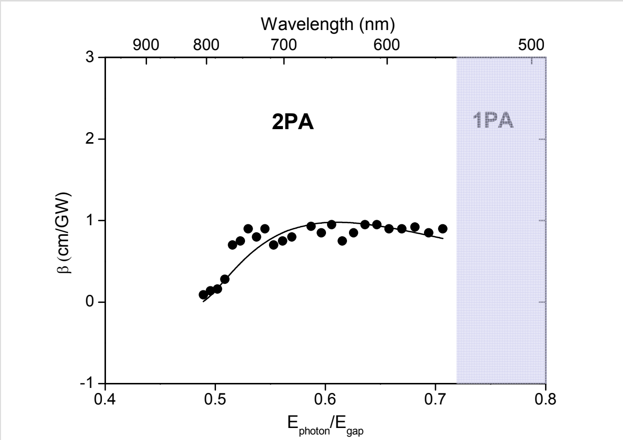
Results: fluorescence

to verify the number of photons in each process



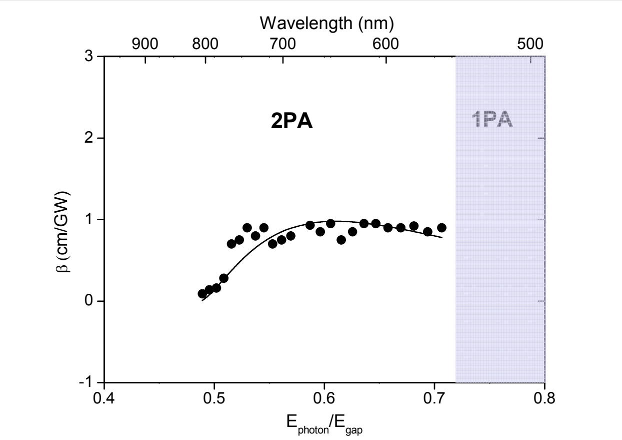
fluorescente at 510 nm

Summary

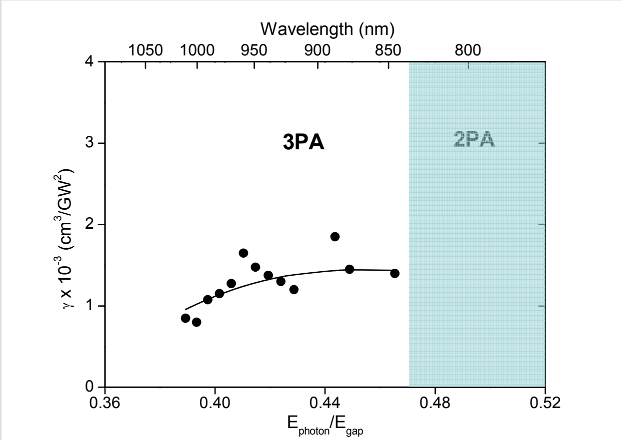


Wavelength (nm)
900
800
700
600
500
2PA
1PA
β (cm/GW)
3
2
1
0
-1
0.4
0.5
0.6
0.7
0.8
E_{photon}/E_{gap}

Summary

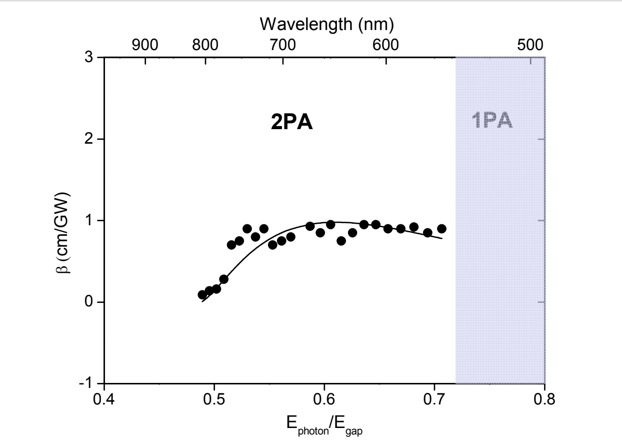


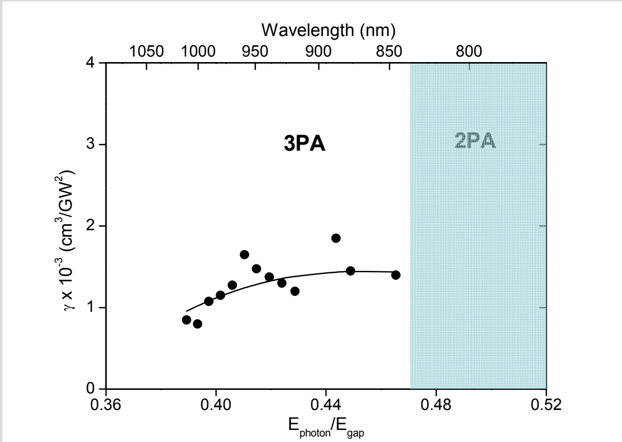
Wavelength (nm)
900
800
700
600
500
3
2
1
0
-1
2PA
1PA
β (cm/GW)
0.4
0.5
0.6
0.7
0.8
E_{photon}/E_{gap}

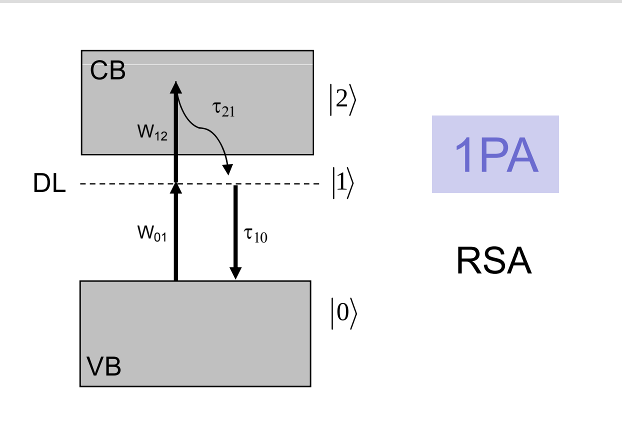


Wavelength (nm)
1050
1000
950
900
850
800
4
3
2
1
0
3PA
2PA
$\gamma \times 10^{-3}$ (cm^3/GW^2)
0.36
0.40
0.44
0.48
0.52
E_{photon}/E_{gap}

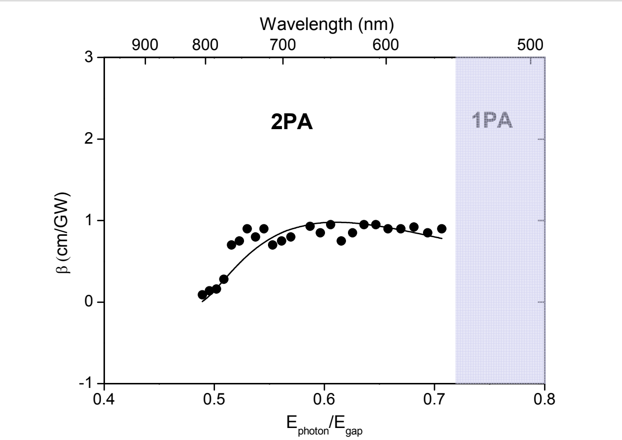
Summary

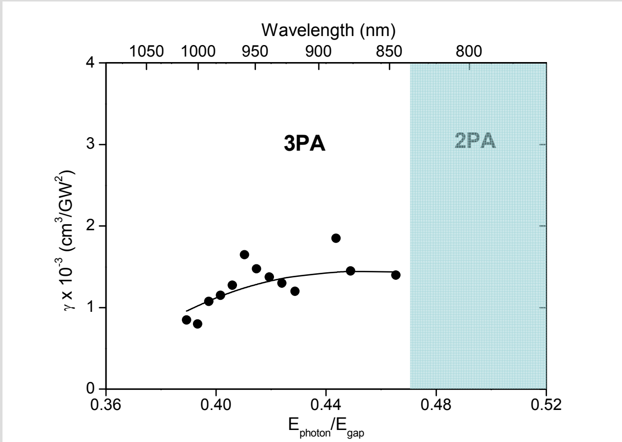

Wavelength (nm)
900 800 700 600 500
2PA
1PA
β (cm/GW)
E_{photon}/E_{gap}

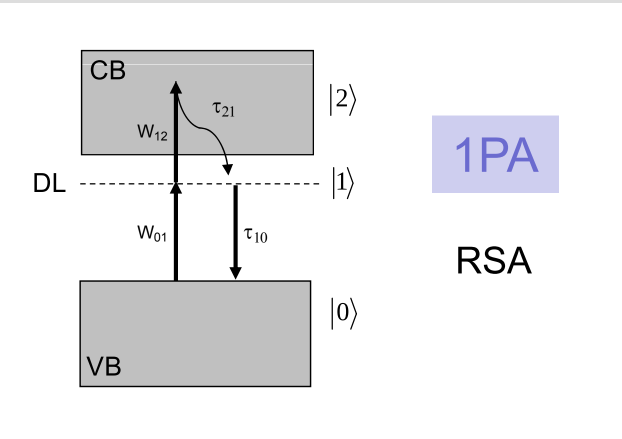

Wavelength (nm)
1050 1000 950 900 850 800
3PA
2PA
γ x 10⁻³ (cm³/GW²)
E_{photon}/E_{gap}

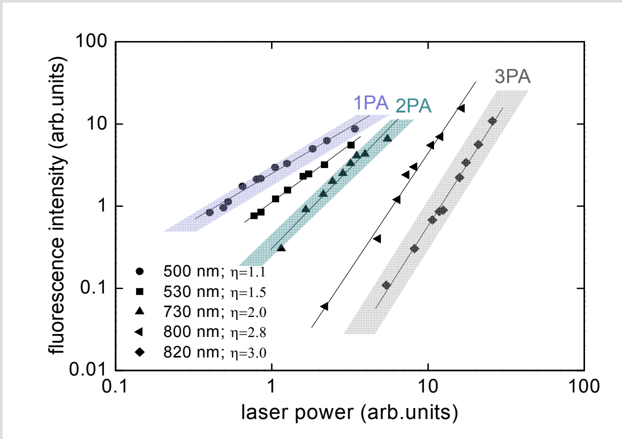

CB
$|2\rangle$
τ_{21}
W_{12}
DL
$|1\rangle$
W_{01}
τ_{10}
$|0\rangle$
VB
1PA
RSA

Summary


Wavelength (nm)
900 800 700 600 500
2PA
1PA
β (cm/GW)
E_{photon}/E_{gap}


Wavelength (nm)
1050 1000 950 900 850 800
3PA
2PA
$\gamma \times 10^{-3}$ (cm^3/GW^2)
E_{photon}/E_{gap}


CB
VB
DL
W_{12}
W_{01}
τ_{21}
τ_{10}
$|2\rangle$
$|1\rangle$
$|0\rangle$
1PA
RSA


1PA 2PA
3PA
500 nm; η=1.1
530 nm; η=1.5
730 nm; η=2.0
800 nm; η=2.8
820 nm; η=3.0
fluorescence intensity (arb.units)
laser power (arb.units)

Acknowledgments

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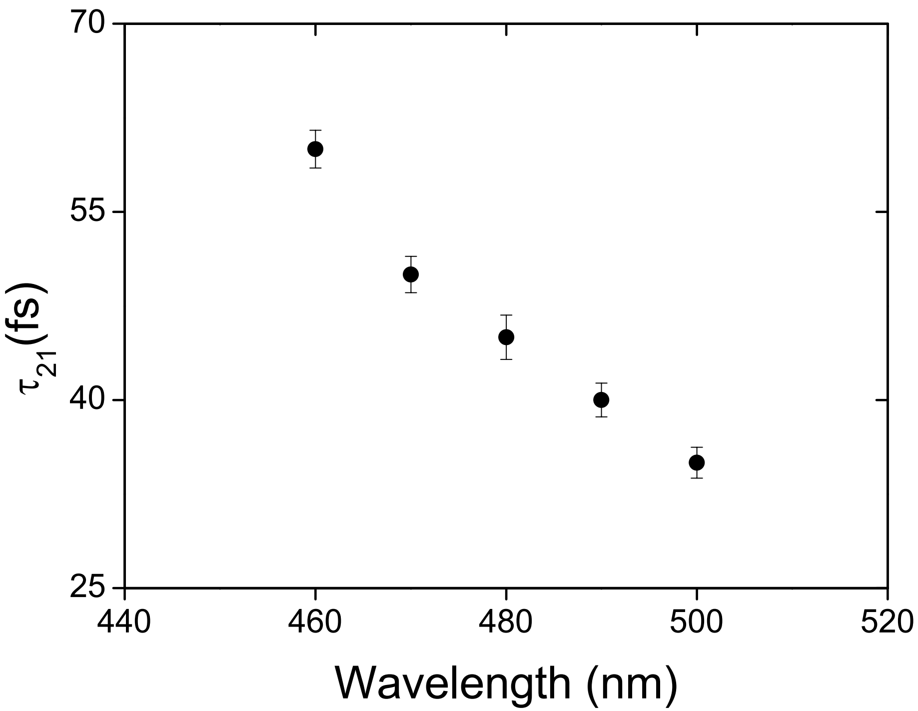
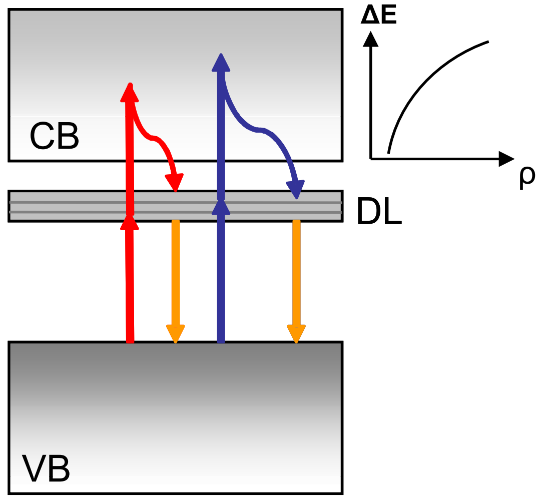
Results

Dynamics

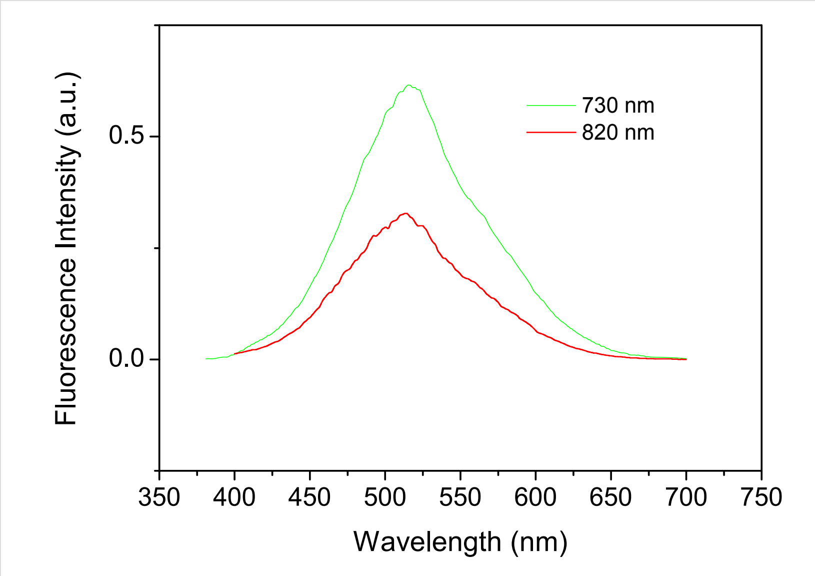




$$\rho(E) = \rho_0 \exp\left(\frac{E - E_{CBM}}{m_c}\right)$$

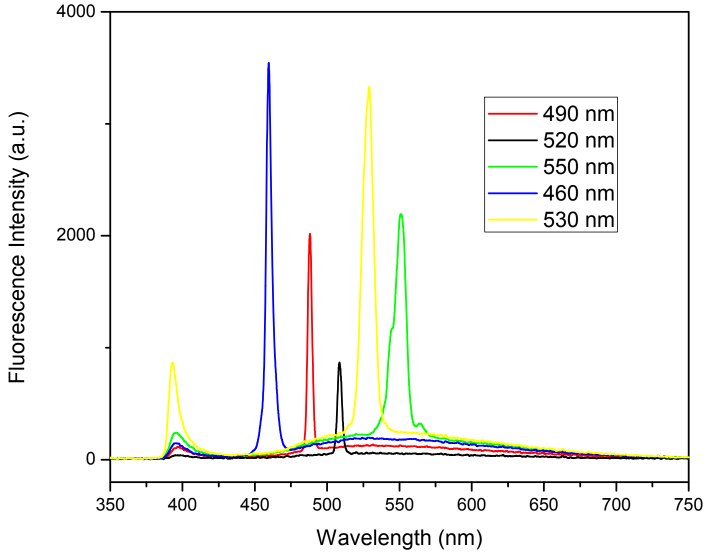
The relaxation time to donor levels increase when the energy differences between the excited impurity states and the edge conduction band decrease.

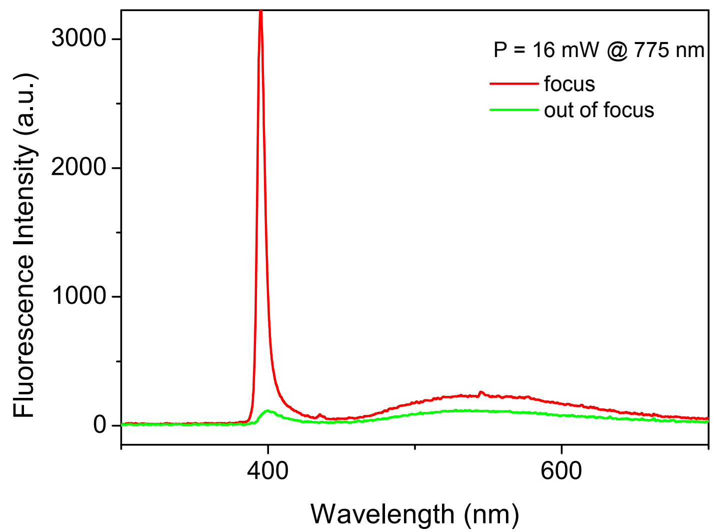
Results



Fluorescence Intensity (a.u.)
0.5
0.0
350
400
450
500
550
600
650
700
750
Wavelength (nm)
730 nm
820 nm

Results


4000
2000
0
Fluorescence Intensity (a.u.)
350
400
450
500
550
600
650
700
750
Wavelength (nm)
490 nm
520 nm
550 nm
460 nm
530 nm


3000
2000
1000
0
Fluorescence Intensity (a.u.)
400
600
Wavelength (nm)
P = 16 mW @ 775 nm
focus
out of focus