Two Photon Absorption & Carrier Generation in Semiconductors

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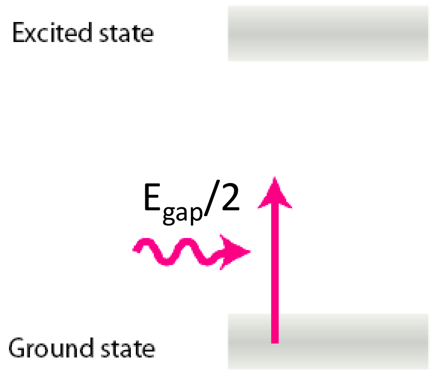
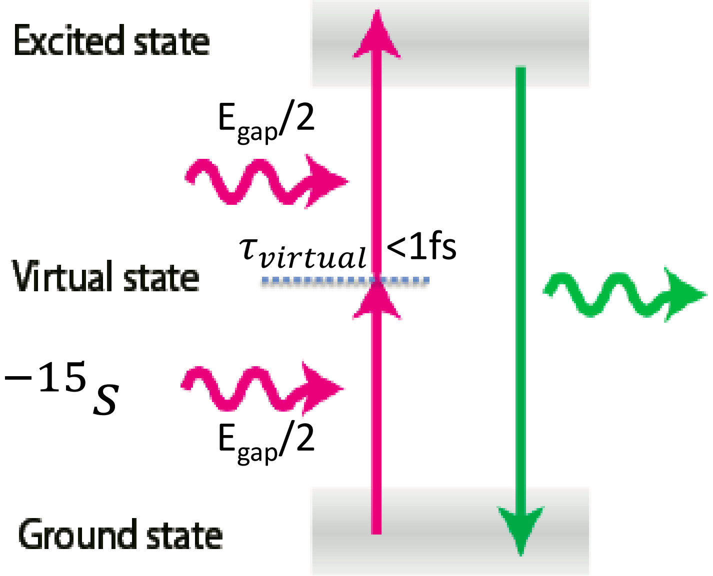
Two Photon Absorption (TPA) Origins

Maria Goppert-Mayer
1963 Nobel Prize in Physics

Conventionally, no excitation if $E_{photon} < E_{gap}$ ~1eV



But if TWO photons arrives in ~ 100 attoseconds



$$\tau_{virtual} \sim \frac{\hbar}{E_{gap}/2} \sim 0.1x10^{-15} s$$

One vs Two-photon Excitation

In 1931, it was predicted theoretically by Maria Goppert-Mayer ,[1], in her PhD dissertation, that due to the time-energy uncertainty principle, forbidden under gap single photon atomic transitions ($h\nu < E_{gap}$) can be possible adding the energy of two almost simultaneous photons ($2h\nu > E_{gap}$) through intermediate "virtual" states. In fluorescent molecules, two photon pumping was demonstrated by Webb et al in 1990 [2] by detection of the fluorescency photon. Refocusing the optics in depth, we can obtain a Z-scan, by Prof. Eric W. Van Stryland and Prof. David J. [3].

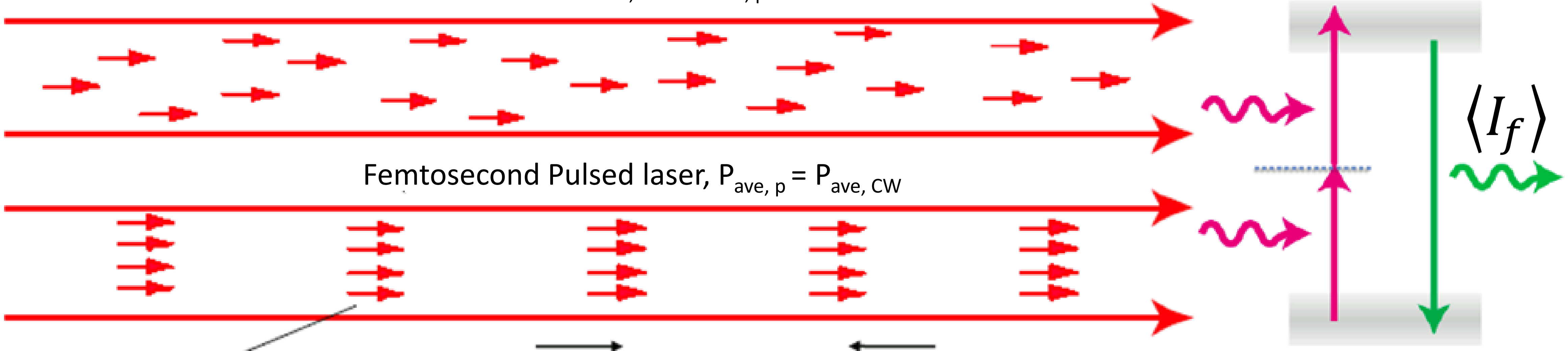
References:

[1] M. Gappert-Mayer, ***Ann. Phys.,*** 1931, **9**, 273-294.
[2] W. R. Zipfel, R. M. Williams, W. W. Webb, ***Nat. Biotechnol***., 2003, **21**, 1369-1377.
[3] M. Sheik-Bahea, A. A. Said, T. H. Wei, D. J. Hagan, E. W. Van Stryland, ***IEEE J. Quantum Electronics***, 1990, **26**, 760-769.

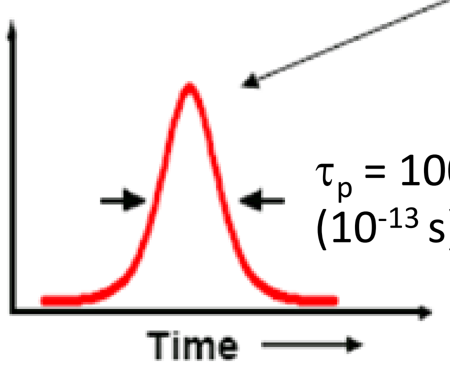
Lasers for Two Photon Absorption (TPA)

CW laser, $P_{ave,\ CW} = P_{ave,\ p}$

Fluorescent Molecule



12.5 ns (f_p= 80 MHz)



τ_p = 100 fs
(10^{-13} s)

$$\langle I_{f,CW} \rangle \propto \sigma_{2PE} P_{ave}^2$$

Fluorescence light Intensity
CW laser excitation

$$\langle I_{f,P} \rangle \propto \sigma_{2PE} \frac{P_{ave}^2}{\tau_P\ f_p}$$

Fluorescence light Intensity
Pulsed laser excitation

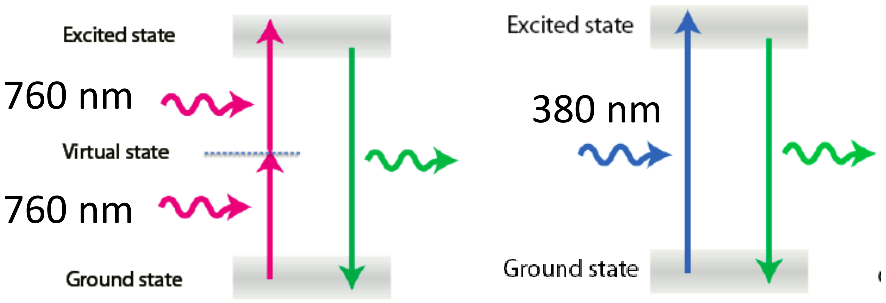
The probability of a two photon absorption is increased by $(1/t_p f_p)=10^5$ for the same average power by using a <u>mode-locked pulsed laser</u>

Two-photon fluorescence excitation and related techniques in biological microscopy, A.Diaspro et al., Quaterly Reviews of Biophysics 38(29) 2005, pp 97-166

Z scan Technique
A Photochemistry Illustrative Example

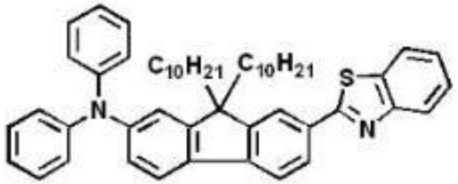
2PA in organic liquids

Ti:Sapphire Laser
200 fs, 760 nm, 76 MHz Rep.Rate
Raw Focusing on Fluorene 3:
Two Photon Absorption Induced Fluorescence
With a second harmonic generator
200 fs, 380 nm, 76 MHz Rep. Rate
One Photon Absorption Induced Fluorescence



SPA signature: a diffraction Rayleigh cone
<u>TPA signature</u>: a single blue dot

Fluorene 3



http://chemistry.cos.ucf.edu/belfield/photophysics

Z scan Technique
Funny and Useful!

SubSurface Laser Engraving (SSLE)

Tipically in BK7 Glass (Borosilicate doped with potassium)

Also with pure quartz (SiO_2)

Pico or FemtoSecond Laser, 1064 nm (SiO_2), 532 nm (BK7)

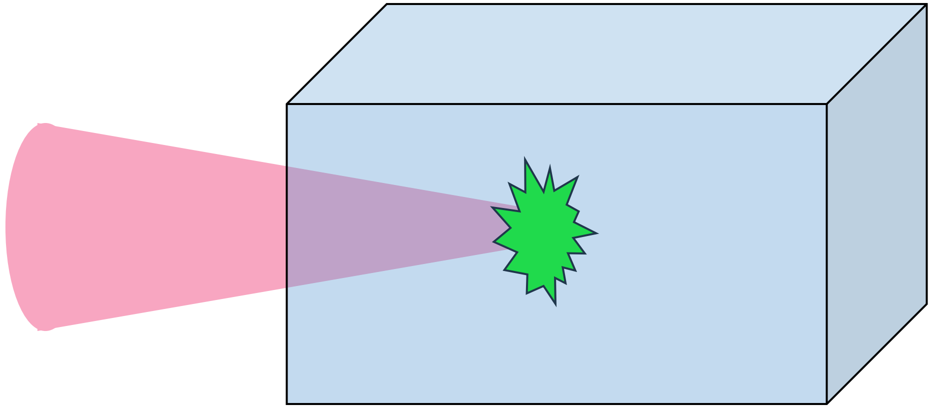
Multi-Photon Absorption

Free electron creation in the focus point

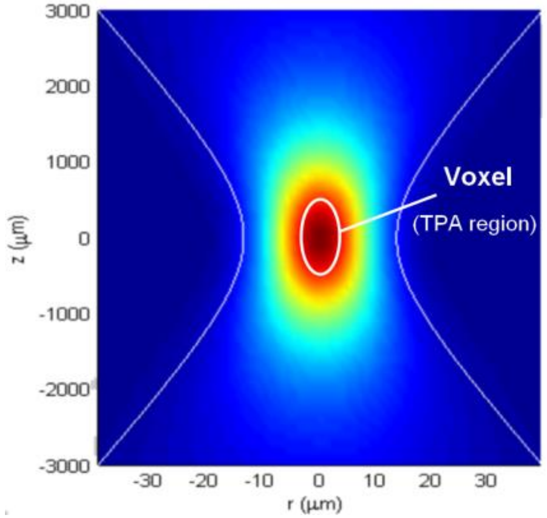
FotoChemistry in Solids:

Index of refraction changes,

Color centers



TJDP-532K Machine (532 nm, BK7 crown glass)
http://www.tianjunlaser.com/



Two-Photon Photopolymerization and 3D Litographic Microfabrication. H.B.Sun and S.Kawata. APS (2004) 170 pp 169-273, Springer-Verlag.
Femtosecond Laser Litography in Organic and Non-Organic Materials, F.Jipa et al., Chap.3, Nanotechnology and Nanomaterials, "Updates in Advanced Litography", ed. by S.Hosaka, INTECH, 2013.

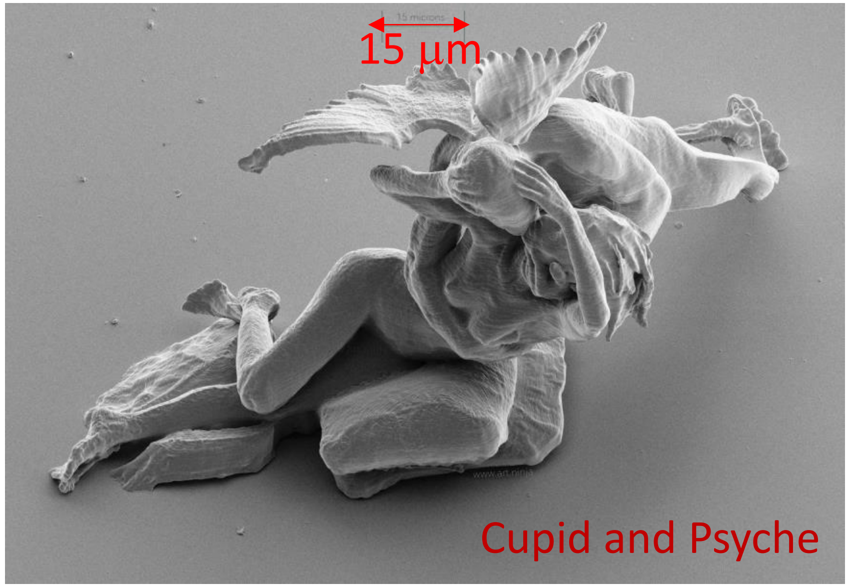
Z scan Technique
3D Litography

3D Litography

If you illuminate a light-sensitive polymer with Ultra Violet wavelengths, it solidifies wherever it was irradiated in a kind of crude lump. It's the process your dentist uses when your filling is glued in with a UV light.

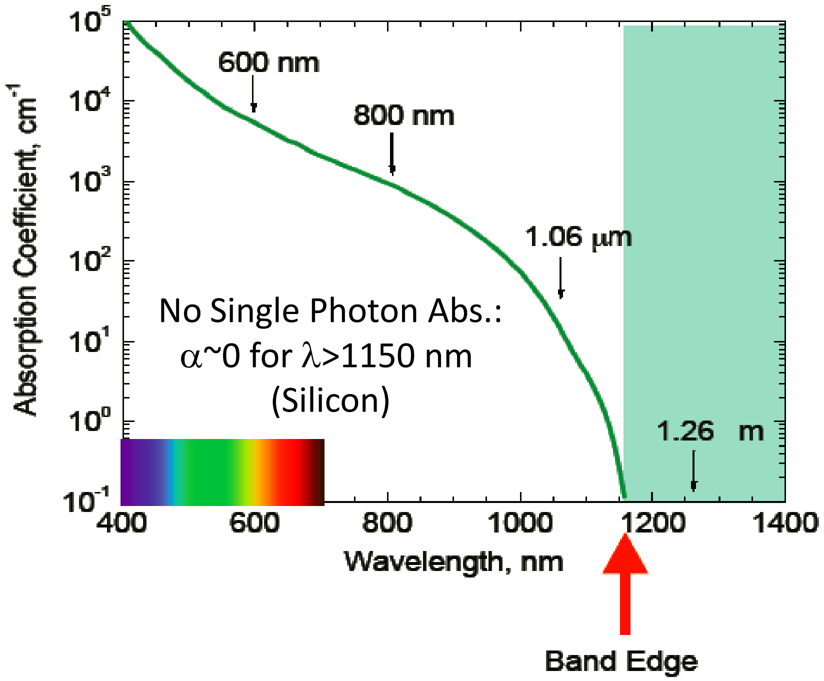
If however you use longer wavelength intense light, and focus it tightly through a microscope, something wonderful happens: at the focus point, the polymer absorbs <u>TWO PHOTONS</u> and responds as if it had been illuminated by UV light, namely it will solidify. <u>This two photon absorption occurs only at the tiny focal point - basically a tiny 3D pixel (called a Voxel).</u> The sculpture is then moved along fractionally by a computer controlled process and the next voxel is created. Slowly, over hours and hours the entire sculpture is assembled voxel by voxel and layer by layer.

Jonty Hurwitz, TPA nano-sculptor,
http://www.jontyhurwitz.com/nano



Two Photon Absorption (TPA)
Silicon

- In solid state, fast (ps) optical excitation generates electron-hole carriers.
- If the laser pulse wavelength is sub-bandgap the material is transparent to the optical pulse.
- Carriers are generated (TPA) by nonlinear absorption at high pulse irradiances by the simultaneous absorption of two photons
- Carriers are highly concentrated in the high irradiance region near the beam focus



$$\frac{dI(r,z)}{dz} = -\alpha I(r,z) - \beta_2 I^2(r,z) - \sigma_{ex} N I(r,z)$$

Optical Absorption Equation

$$\frac{d\Phi(r,z)}{dz} = \beta_1 I(r,z) - \gamma_1 N(r,z)$$

Phase Change by free carriers

$$\frac{dN(r,z)}{dt} = \frac{\alpha I(r,z)}{\hbar\omega} + \frac{\beta_2 I^2(r,z)}{2\hbar\omega}$$

Carrier Generation equation

And free carrier absorption (σ_{ex}) is neglible if we avoid heavily doped volumes (<1E20 cm^{-3}), so the TPA carrier generation goes as:

$$\frac{dN_{2P}(z)}{dt} = \frac{\beta_2}{2\hbar\omega} I^2(z,t) \quad ; \quad I(z) = \frac{I_o}{1+\beta_2 I_o z}.$$

$$N_{2P}(z) = \frac{\beta_2}{2\hbar\omega}\int_{-\infty}^{\infty} I^2(z,t)\,dt$$

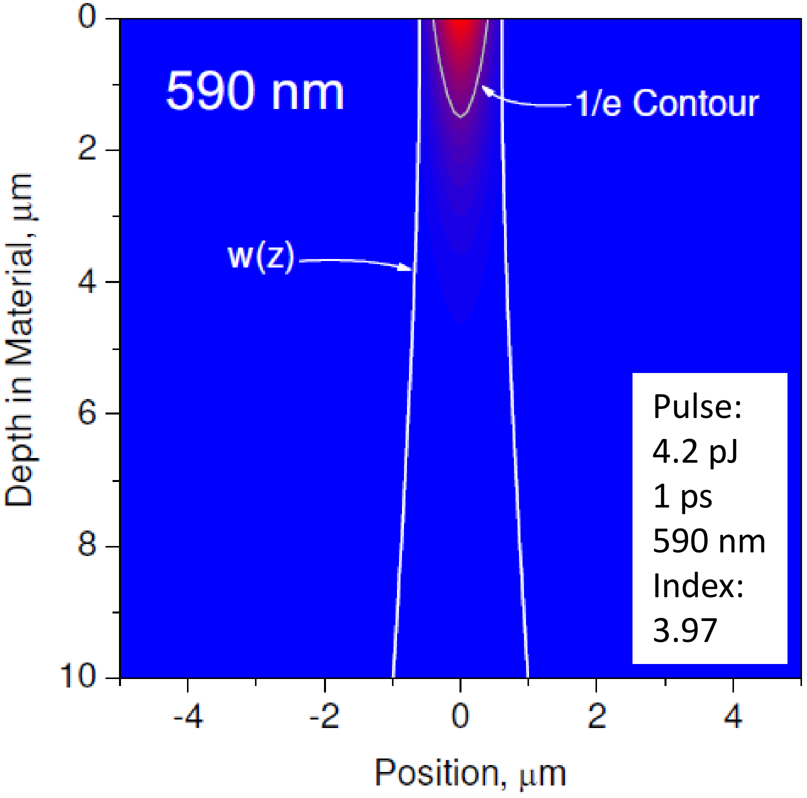
Subbandgap Laser-Induced Single Event Effects: Carrier generation via two photon absorption. D.M.McMorrow et al. IEEE Transactions on Nuclear Science, 49 (6) Dec 2002, pp 3002-3007.
Laser Simulation of Single-Event Effects: A state of the art review, S.Buchner, Army Research Laboratory, ARL-CR-185

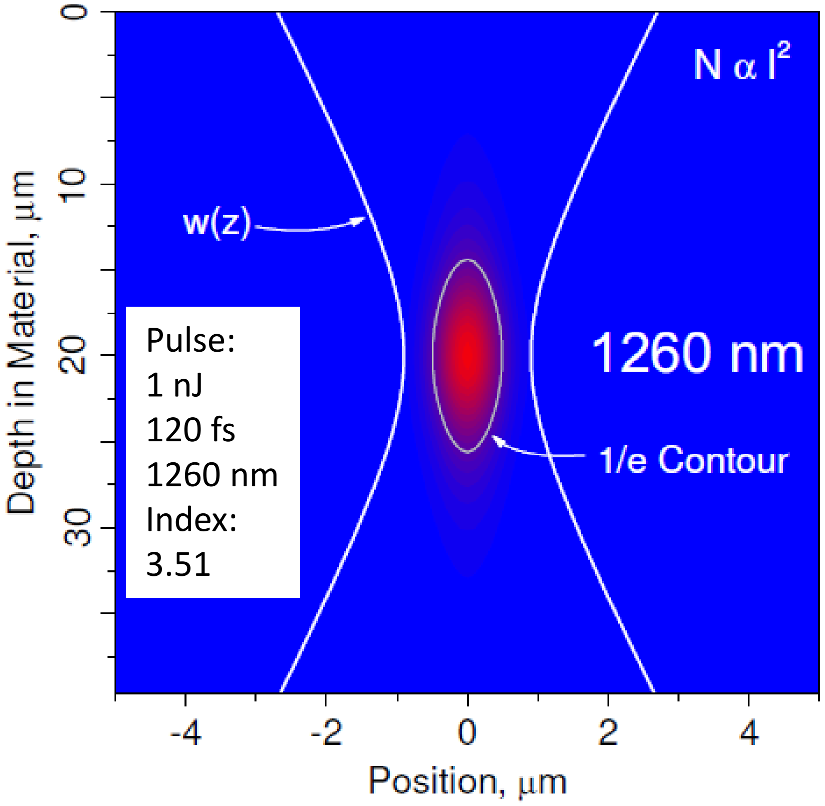
TPA in Silicon
Gaussian Pulse (TM00)
Femtosecond Laser

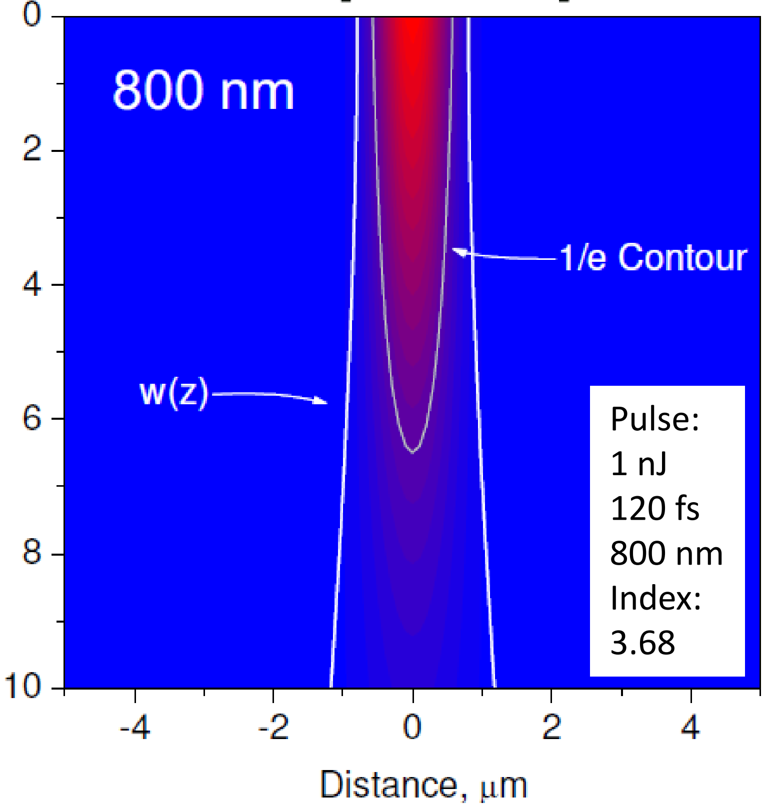
All of them focused to a diameter of 1.2 μm

$$I(r,\, z) = \frac{2P}{\pi w^2}\, \exp(-2r^2/w^2)$$

$$w(z) = w_o \left[1 + \left(\frac{\lambda z}{\pi w_o^2 n}\right)^2\right]^{1/2}$$







$$N_{1P}(z) = \frac{\alpha}{\hbar\omega}\, \exp(-\alpha z) \int_{-\infty}^{\infty} I_o(z,\, t)\, dt$$

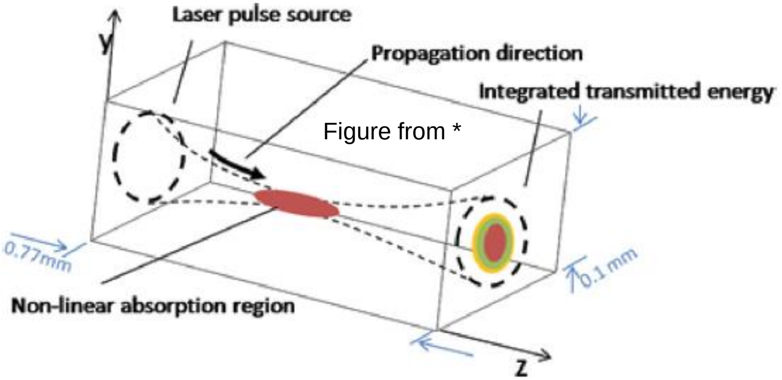
$$N_{2P}(z) = \frac{\beta_2}{2\hbar\omega} \int_{-\infty}^{\infty} I^2(z,\, t)\, dt$$

$$N_{1P}(z) = \frac{\alpha}{\hbar\omega}\, \exp(-\alpha z) \int_{-\infty}^{\infty} I_o(z,\, t)\, dt$$

Single Event Effect Induced by Two-Photon Absorption: Overview and Current Status. D.M.McMorrow et al. **RADECS 2004**, Madrid, Spain, 22nd-24th September , 2004.

Instituto de Física de Cantabria

TPA in Silicon
Gaussian Pulse (TM00)
Femtosecond Laser



Gaussian Intensity Profile

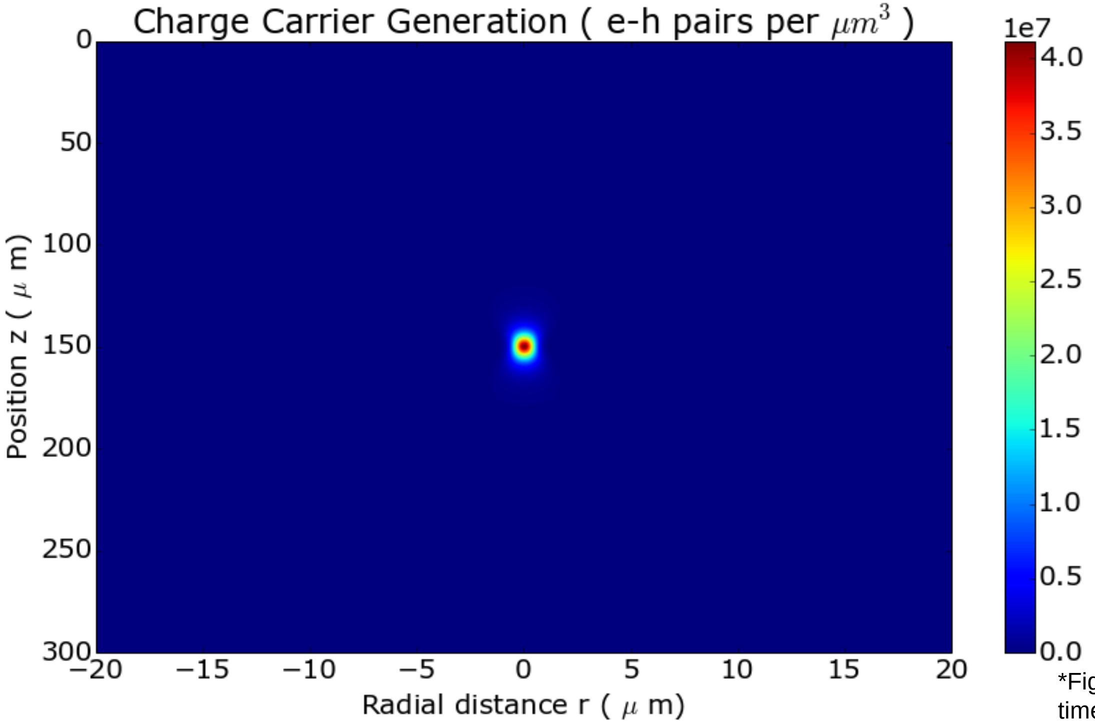
$$w(z)^2 = w_o^2\left[1+\left(\frac{\lambda z}{\pi w_o^2 n}\right)^2\right]$$

$$I(r,z) = \frac{2P}{\pi w(z)^2}\exp\frac{-2r^2}{w(z)^2}$$

τ = 120 fs
β= 0.3 cm/GW
Power = 1.e-9/120e-15 W
λ = 1300 nm
w_0 = 0.947e-6 m
n (Si) = 3.51
z_0 = 150e-6 m

TPA absorption (negligible attenuation)

$$n_{TPA}(z) = \frac{\beta\tau}{2\hbar\omega} I(r,z)^2$$

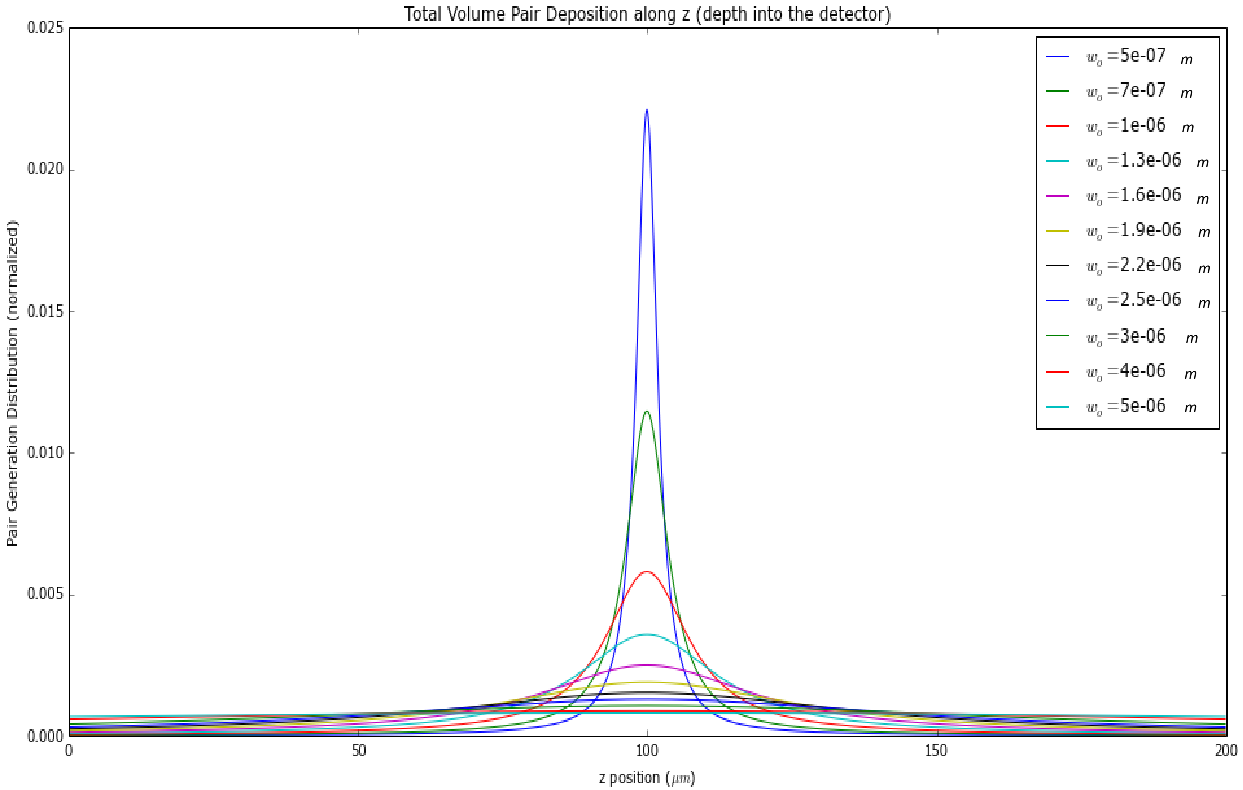


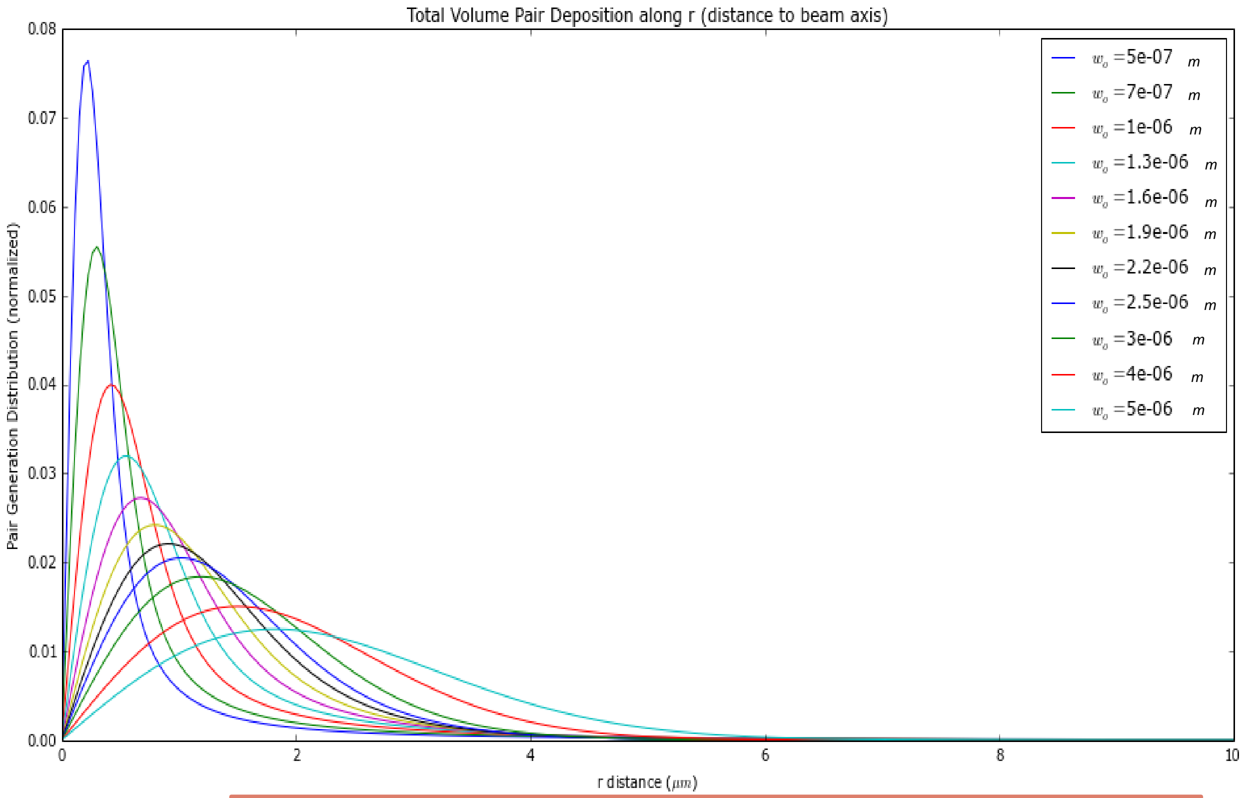
TOTAL NUMBER OF PAIRS GENERATED IN THE DETECTOR

$$N_{total} = \int_{z=200\mu m}^{z=0\mu m}\int_{r=60\mu m}^{r=0\mu m}\int_{\phi=2\pi}^{\phi=0} n_{TPA}(z,r) r d\phi dr dz$$

*Figure from "Non-linear absorption of 1.3-um wavelength femtosecond laser pulses focused inside semiconductors: Finite difference time domain-two temperatura model combined computational study" I.B.Bogatyrev ,D.Grojo, P.Delaporte, S.Leyder, M.Sentis, W.Marine, T.E.Itina J.Appl.Phys. 110, 103106 (2011)

Focus effect on depth/radial distribution





DEPTH PROFILE OF PAIR DEPOSITION

$$N_z(z) = \int_{r=60\mu m}^{r=0\mu m} \int_{\phi=2\pi}^{\phi=0} n_{TPA}(z,r) r dr d\phi dr dz$$

τ = 120 fs

β= 0.3 cm/GW

λ = 1300 nm

n (Si) = 3.51

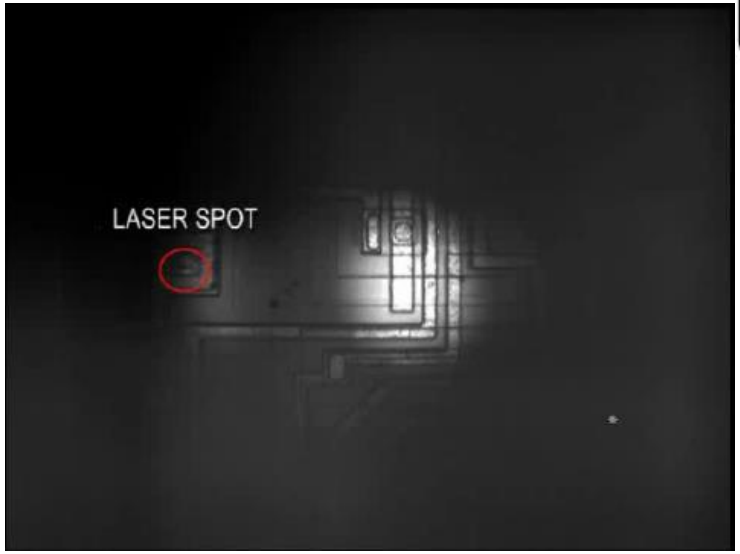
RADIAL PROFILE OF CHARGE DEPOSITION

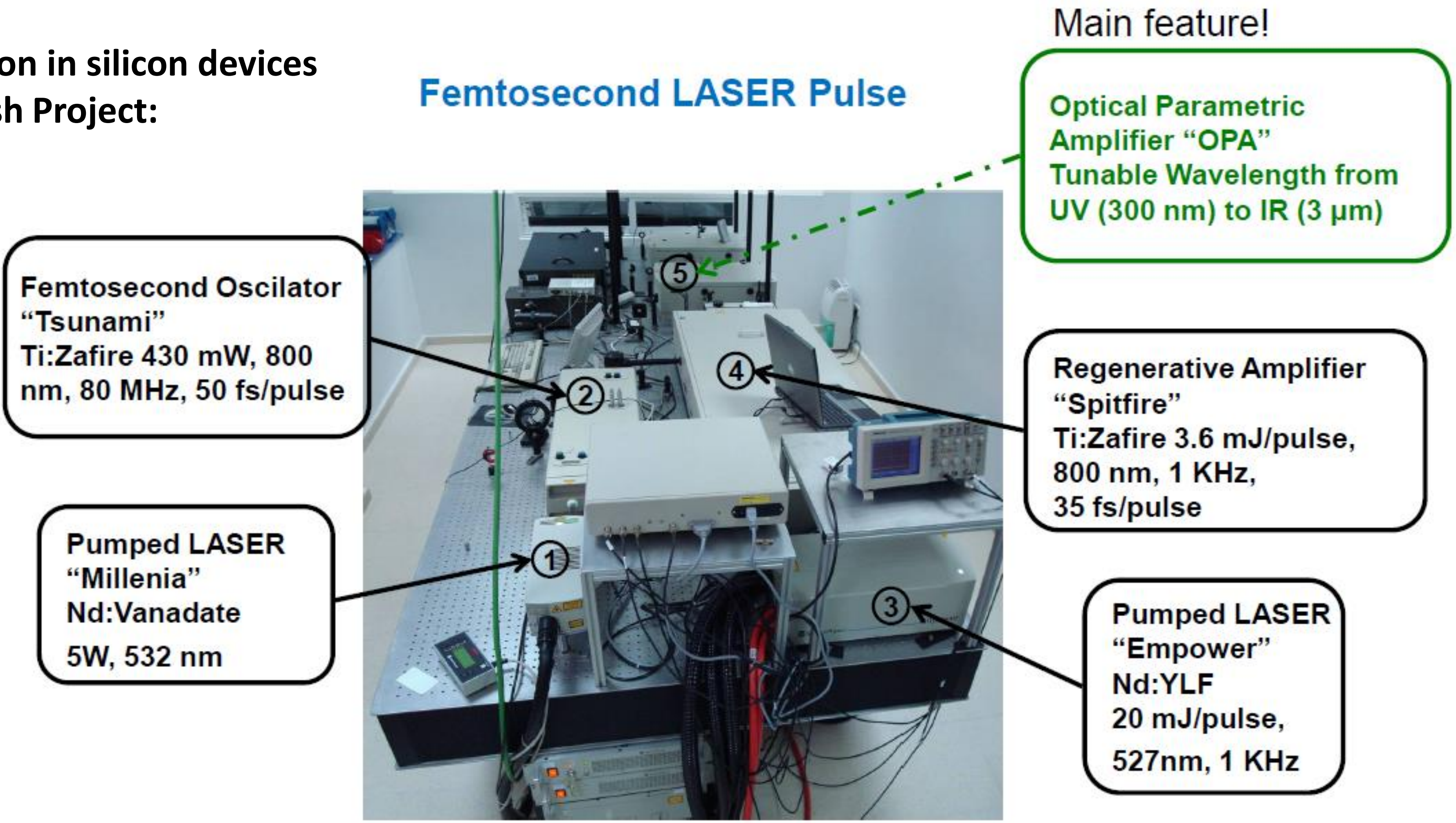
$$N_r(r) = \int_{z=200\mu m}^{z=0\mu m} \int_{\phi=2\pi}^{\phi=0} n_{TPA}(z,r) r dr d\phi dr dz$$

Z-scan for SEE in microelectronics

We had experience of TPA absorption in silicon devices (microelectronics) during the Spanish Project:

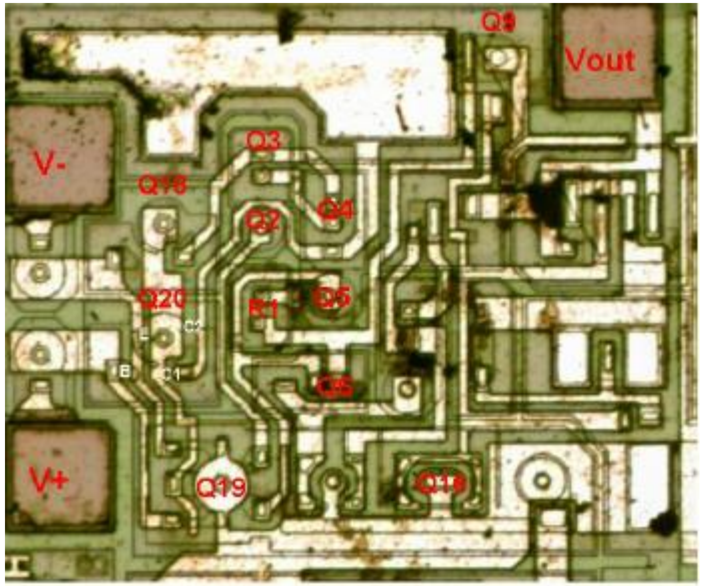
EMULASER PN-PROFIT
PNE-034/2006 ,
31/12/2006 – 10/09/2009

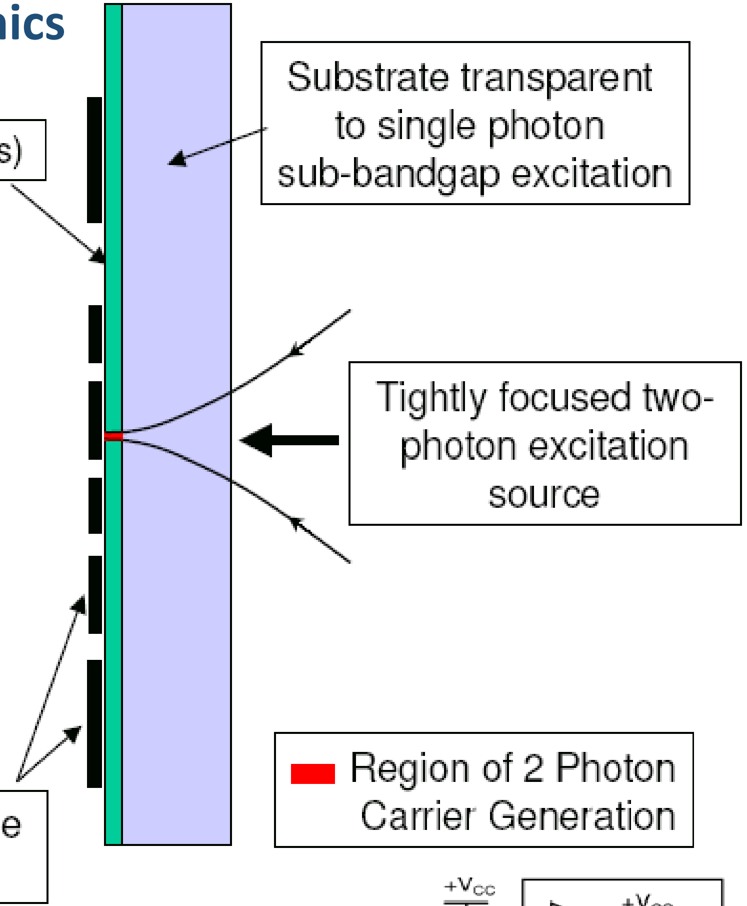


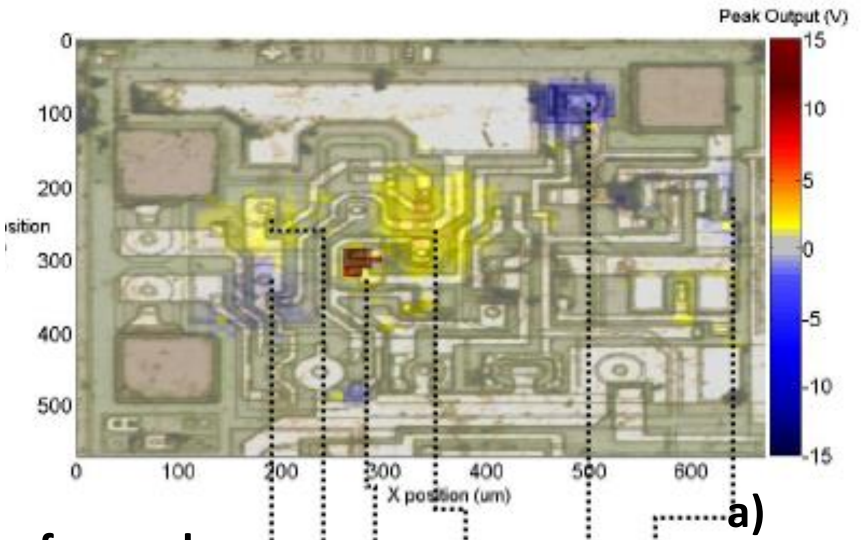


TPA Laser @ University Complutense of Madrid

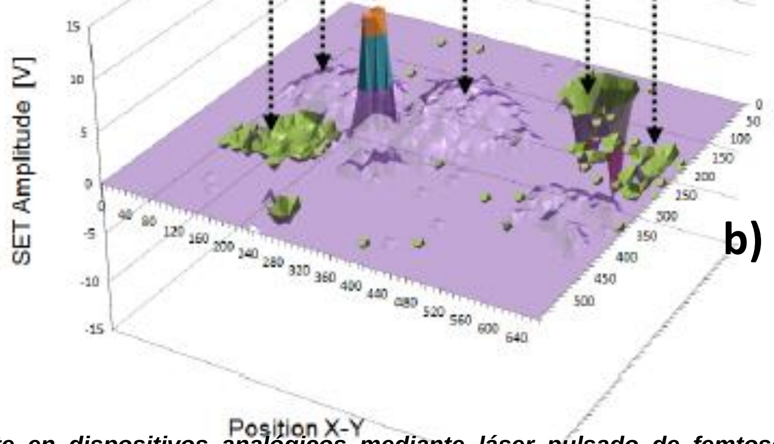
Z-scan for SEE in microelectronics

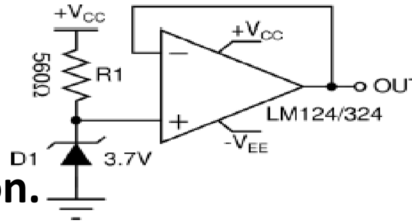






Laser focused 50 μm under the chip surface





Example of a 2D (a) and a 3D (b) SEE sensitive map after a Laser scan over a LM324 opamp, voltaje follower configuration.

Laser Parameters: λ=1300 nm, E_p=0,6 nJ

Emulación de los efectos de la radiación ionizante en dispositivos analógicos mediante láser pulsado de femtosegundo sintonizable. Tesis Doctoral, Isabel López Calle, Facultad de Física, Universidad Complutense de Madrid., Madrid 2010

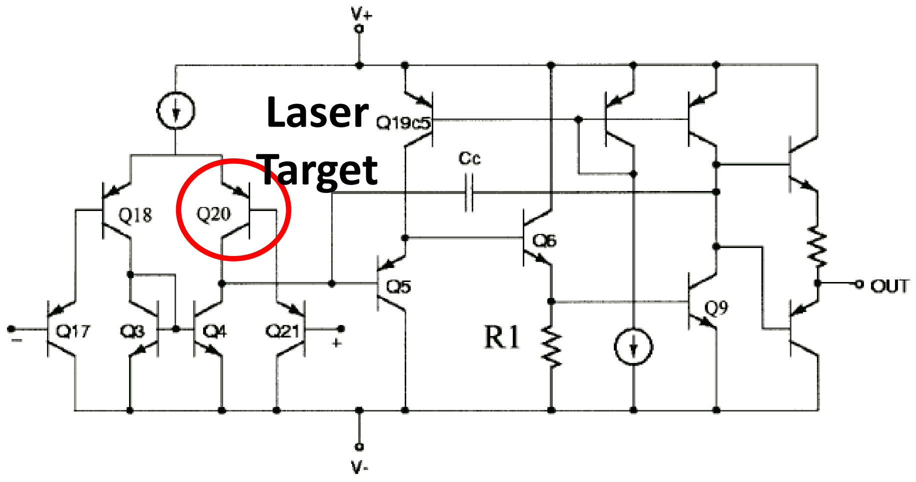
Two Photon Absorption (TPA) Backside Pulsed laser tests in the LM324. I.López-Calle et al. Proceedings of RADECS'09, Bruges, Belgium, September 2009

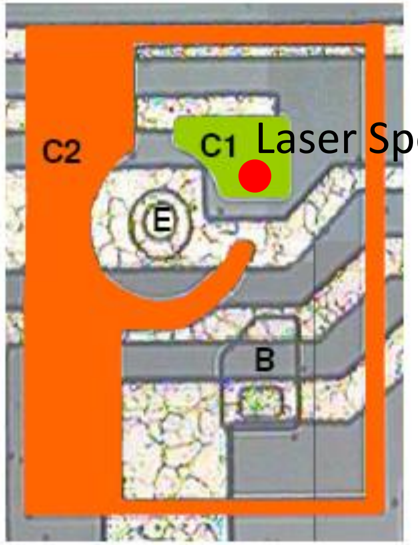
Z-scan for SEE in microelectronics

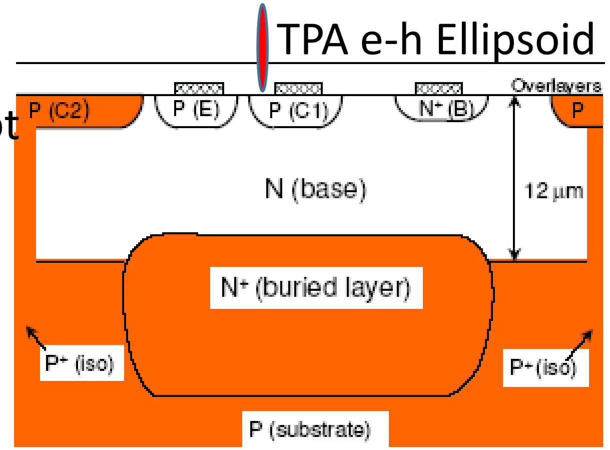
LM124 Op-Amp Inverting configuration, gain of 20

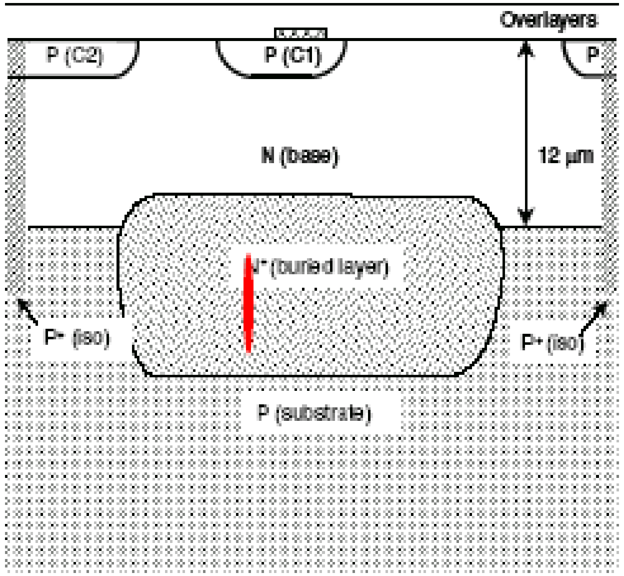
V_{dd}= ±15 V, V_{in}=5 mV

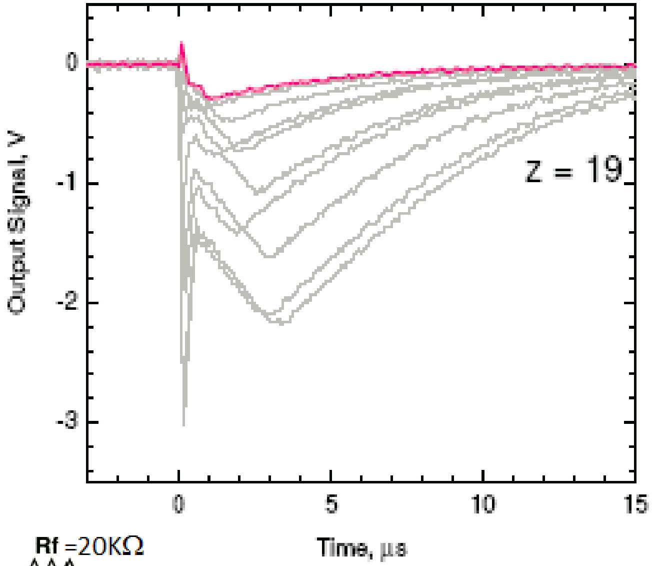
1260 nm, 120 fs pulsed laser, focused on Q20

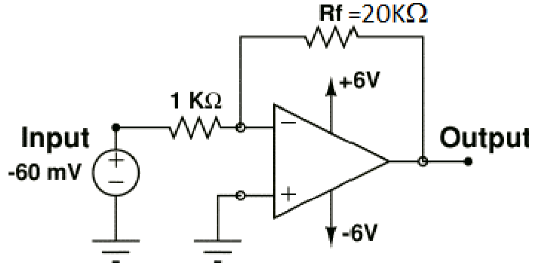






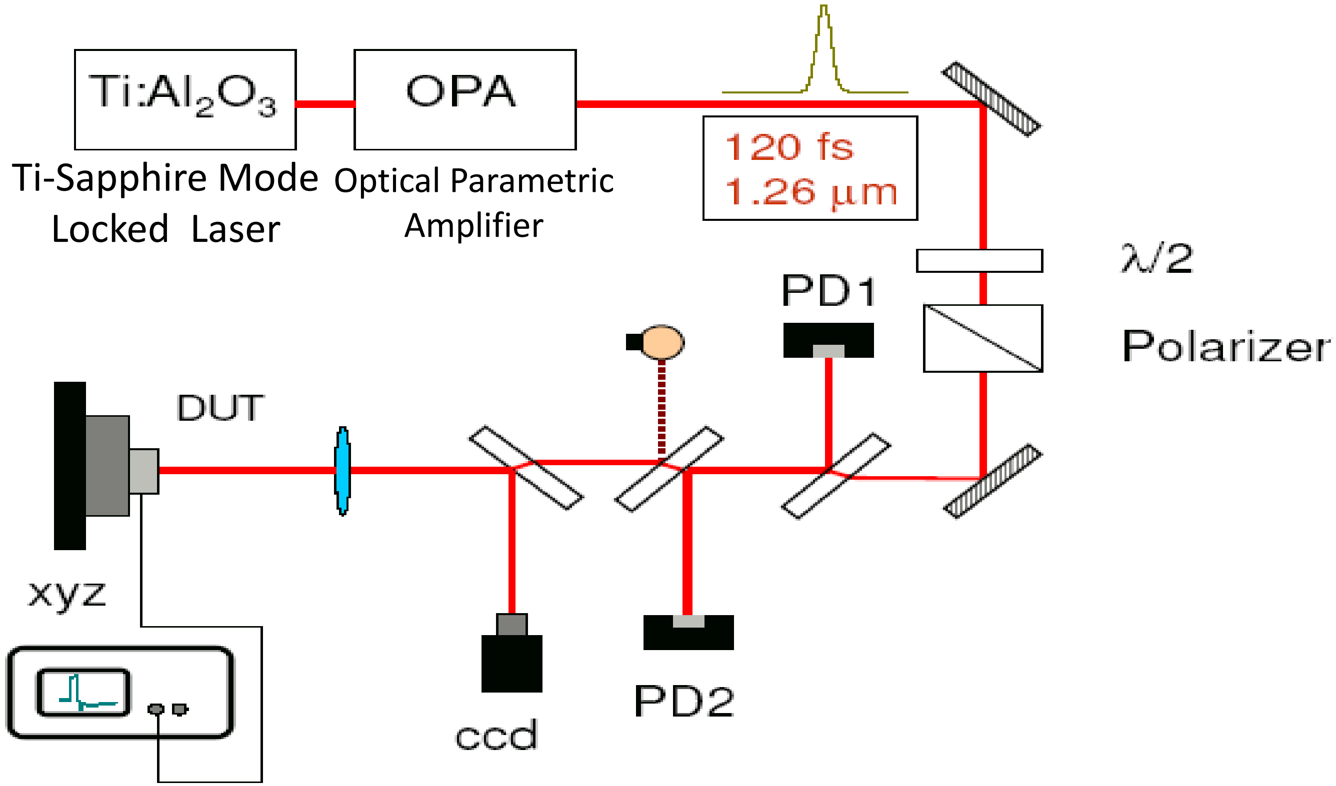






2004 Proceedings of the 6th International Workshop on Radiation Effects on Semiconductor Devices for Space Applications. D.M.McMorrow et al. **RASEDA 2004**, Tsukuba, Japan, 6th-8th October, 2004.

General Layout and Availability



Example: PULSBOX
Turn-key Smart Laser Source for Pulsed Laser Stimulation (1300 nm)
From PULSCAN (www.pulscan.com)
< 100.000 €

Conclusions

- Laser TPA regularly used in TPA fluorescence microscopy
- Laser TPA used since 10 years ago for SEE studies in Silicon microelectronics
- There is industrial equipment for Pulsed Laser interaction in TPA mode
- **Now, we propose to use Laser TPA as a new TCT technique:**

TPA-TCT

See the presentation about the experimental demonstration of TPA-TCT*: **"TPA-TCT, A novel Transient-Current-Technique based on the Two Photon Absorption Process",*** P.Castro, M.Fernández, J.González, R.Jaramillo, M.Moll, R.Montero, F.R.Palomo, I.Vila, 25th RD50 General Meeting, November 19th-21st, 2014, CERN
https://indico.cern.ch/event/334251/session/1/contribution/35/material/slides/0.pptx

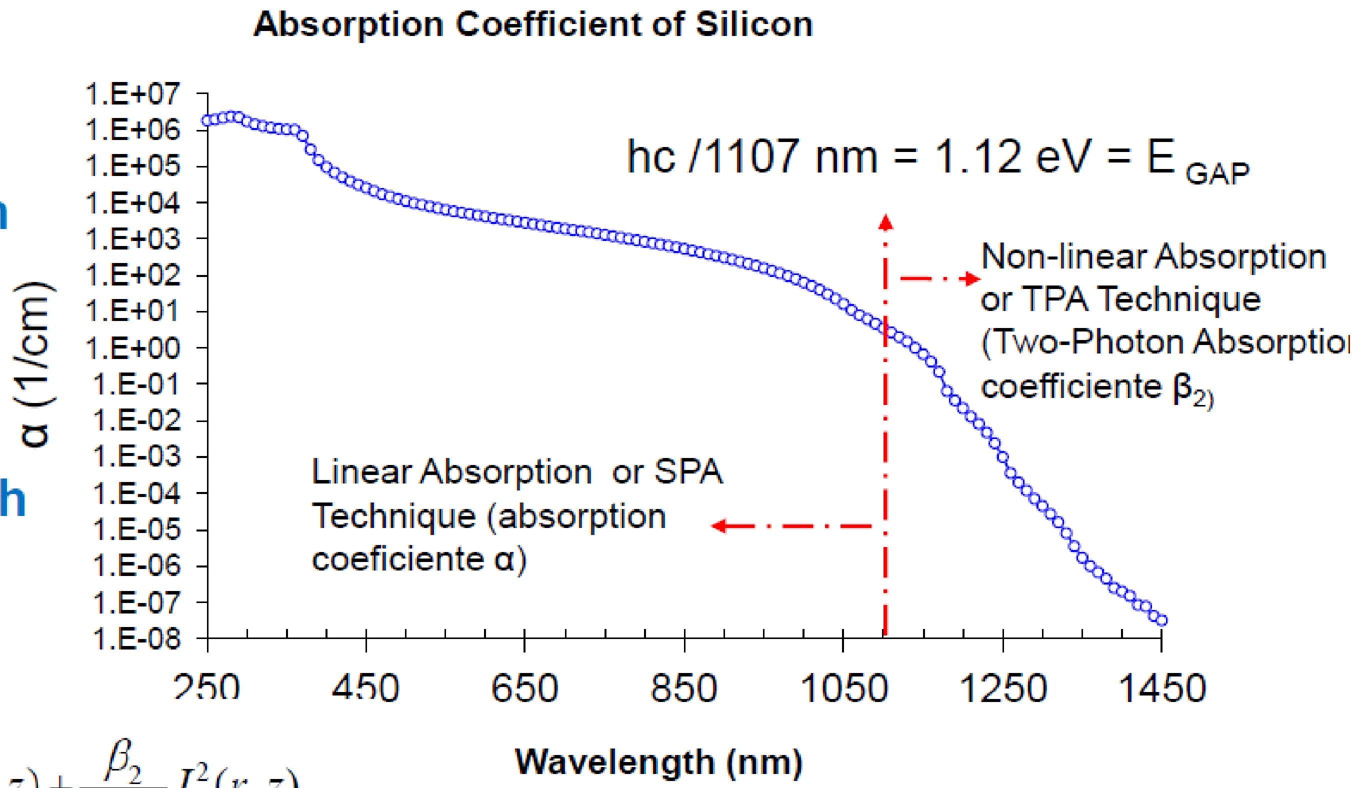
Thanks for your attention

fpalomop@cern.ch

Regarding to the wavelength

↓

We can use both SPA & TPA techniques



$$\frac{dN(r,z)}{dt} = \frac{\alpha}{E\gamma} I(r,z) + \frac{\beta_2}{2 \cdot E\gamma} I^2(r,z)$$

Technique	Adventages	Disadventages
SPA (Single Photon Absorption)	α is well known.	Low penetration
TPA (Two Photon Absorption)	Any level of penetration	β_2 is not well Known. Under investigation

Energy band-gap dependence of two-photon absorption, E.W.Van Stryland et al. Optics Letters, 10(10) Oct.1985, pp.490-492

Two photon absorption coefficient β_2

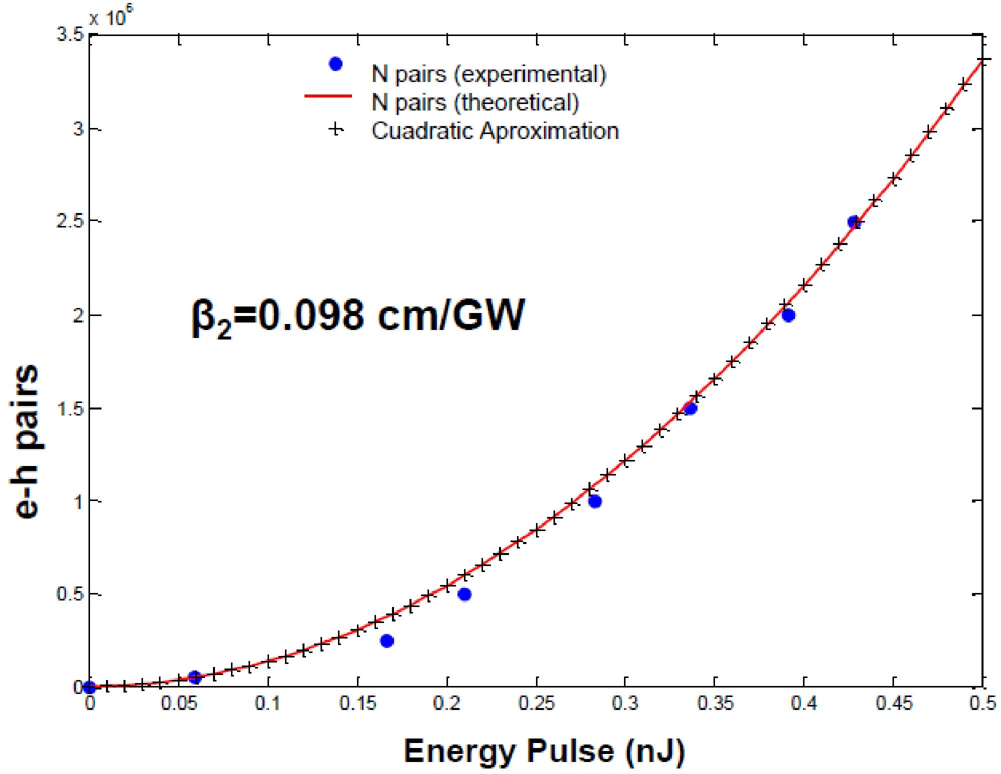
Carrier generation equation:

$$\frac{dN(r,z)}{dt} = \frac{\alpha I(r,z)}{\hbar\omega} + \frac{\beta_2 I^2(r,z)}{2\hbar\omega}$$

β_2 is defined as the probability of simultaneous absorption of two photons.

It differs from linear absorption α in that the absorption depends on the square of the light intensity, thus it is a nonlinear optical process.

$$N_{pairs} = \frac{T^2 \cdot \beta_2 \cdot d}{2E_\gamma \cdot s \cdot t} E_p^2$$



Reference	Li07	Br07	Xu99	Mo92	Bo86
β_2 (cm/GW)	0.55	1.6	1	0.1	1
Type	Exp.	Exp.	Exp.	Teo.	Exp

Effective value of β_2 = meassured at device output signal

Emulación de los efectos de la radiación ionizante en dispositivos analógicos mediante láser pulsado de femtosegundo sintonizable. Tesis Doctoral, Isabel López Calle, Facultad de Física, Universidad Complutense de Madrid., Madrid 2010