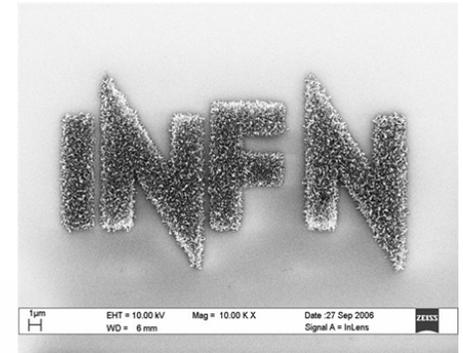


PARIDE



Pixel Array for Radiation Imaging DETector

**INFN
&
University of**

**L'Aquila
Bari
Napoli
Perugia
Roma2**

+ FBK

Large area CNT-Si heterojunction for photodetection

Carla Aramo for PARIDE Collaboration

Micro&Nano for photon detection

MACRO

MICRO

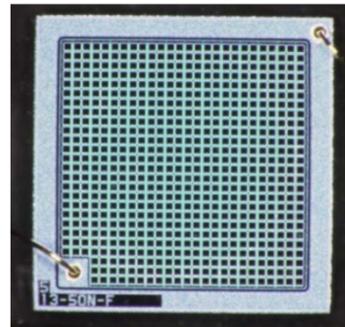
NANO



'30

Issues

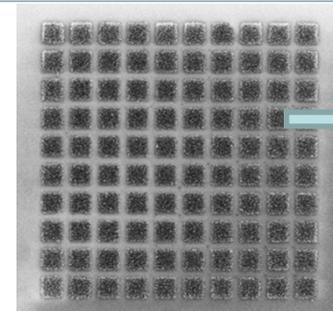
- Intrinsic limit QE < 40%
- high voltage, bulky, fragile
- influenced by B, E fields
- damaged by high-level light
- ageing (eg. He)
- radiopurity



'90

Issues

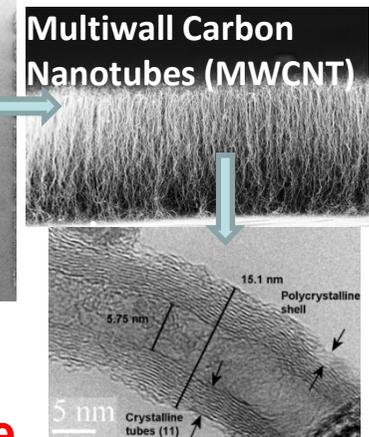
- Dark noise still the most limiting factor → active area
- Low T: perform ideally in the range $100K < T < 200K$
- QE in near-UV region ~ 40% and PDE ~ 30%



Near future

Advantages

- Lower dark noise at room temperature
- Large area and highly segmented → linearity
- Higher QE in the near-UV region



6 C Carbonio 12.0107	7 N Azoto 14.00674
14 Si Silicio 28.0855	15 P Fosforo 30.97376
32 Ge Germanio 72.64	33 As Arsenico 74.9216

from Vacuum to Silicon to Carbon

Carbon Nanotubes (CNTs)

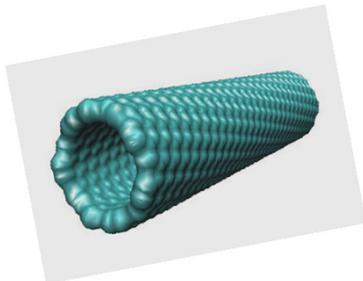
Molecular Nanowires ($d \sim 1 \text{ nm}$, $l \sim 1 \mu\text{m}$)

SWNTs

Single Graphene Sheets ($d \approx 0.7 \div 3 \text{ nm}$, $l \approx \mu\text{-range}$)

$\notin \mathbb{N}$

Semiconductor

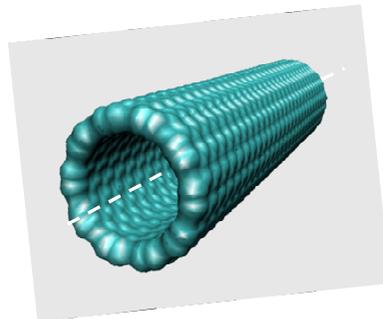


Channel (FETs),
Luminescence

$|n-m|/3$

$\in \mathbb{N}$

Metal



Ballistic Conduction,
 e -wave guides, SETs

MWNTs

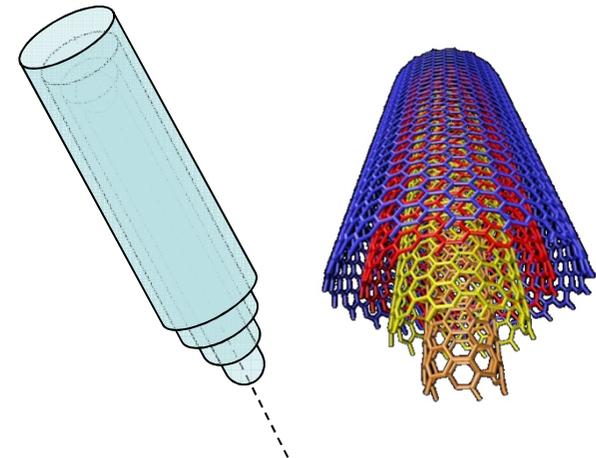
Coaxial graphene sheets

($d \approx 2 \div 100 \text{ nm}$, $l \approx \mu\text{-range}$)

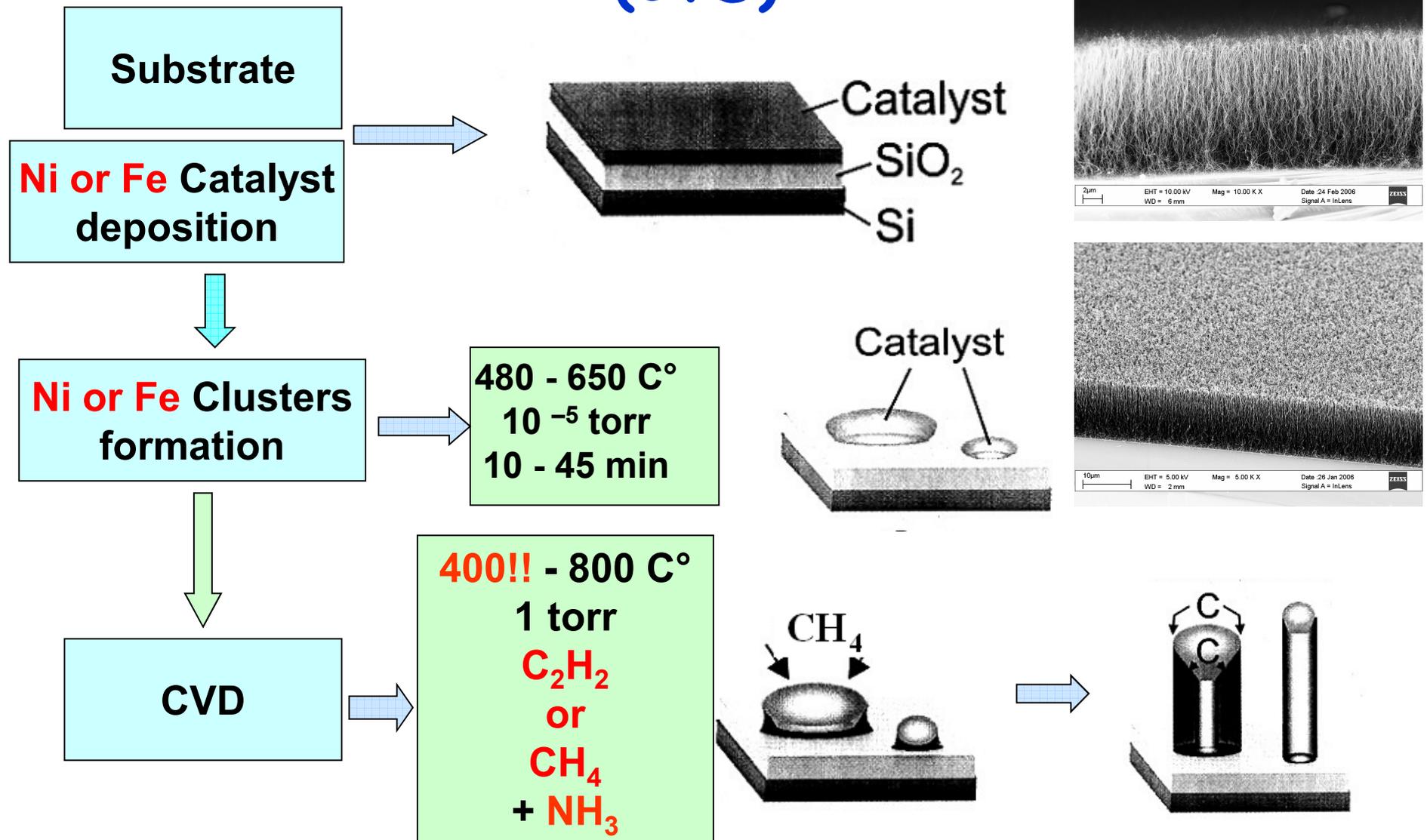
($d^{\text{out}} \approx 20_{\text{AD}}, 100_{\text{CVD}} \text{ nm}$)

Vias

Nanocomposites



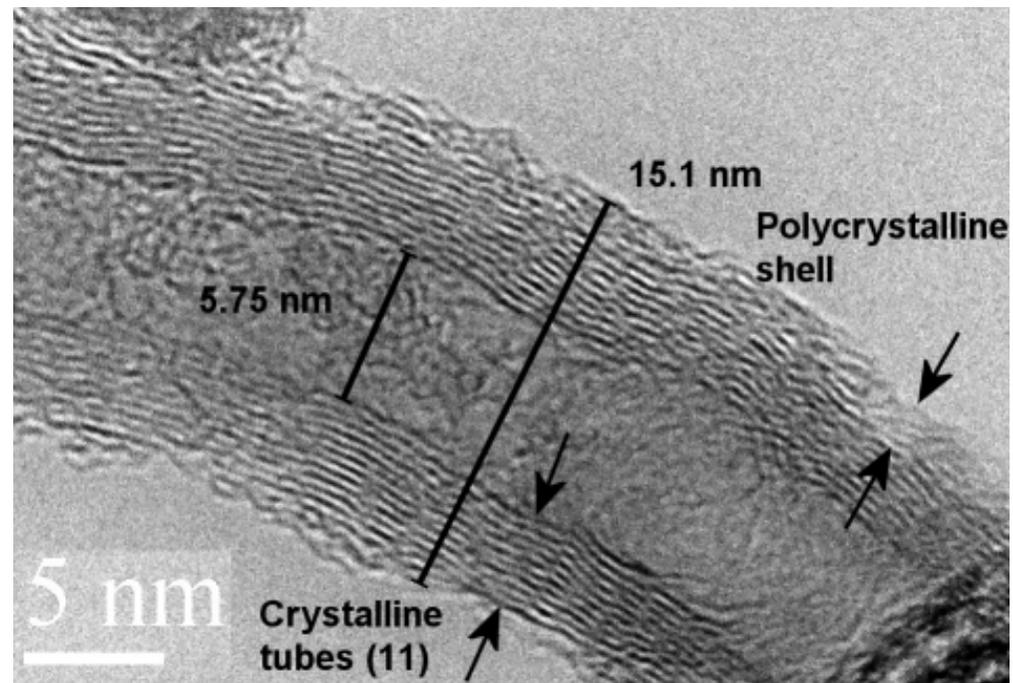
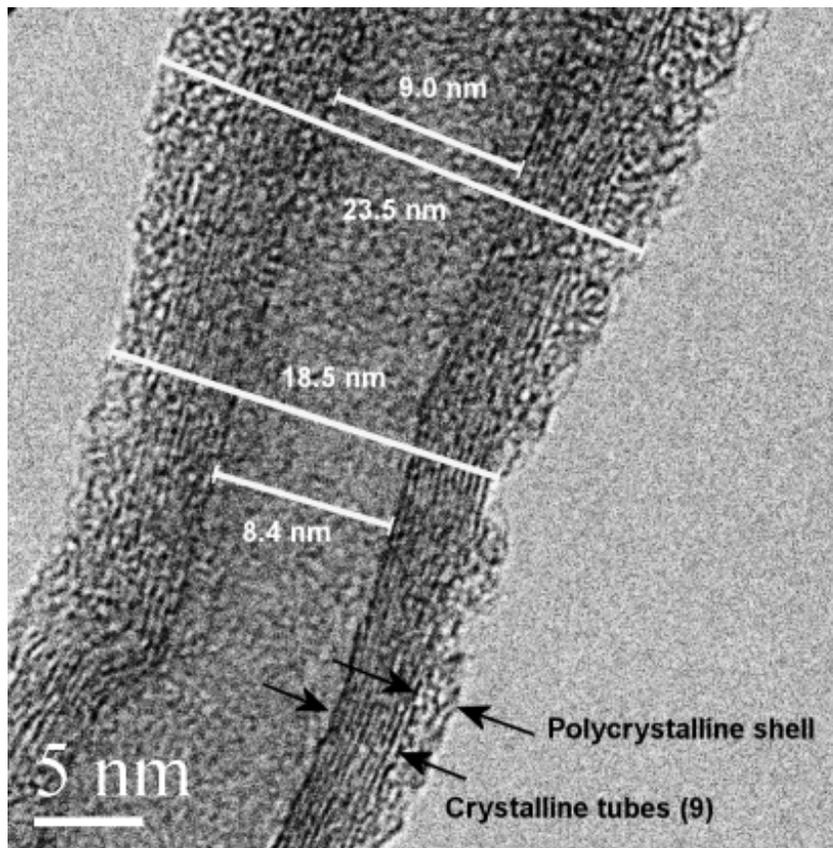
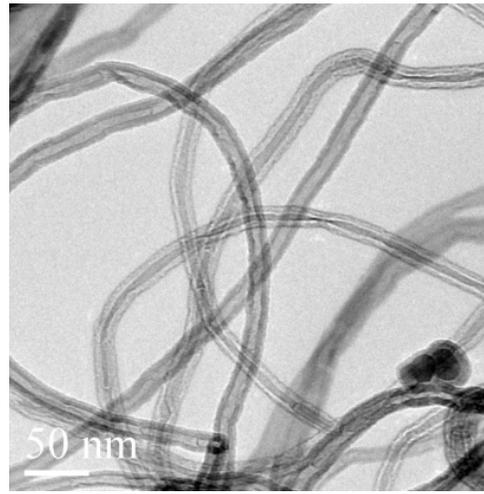
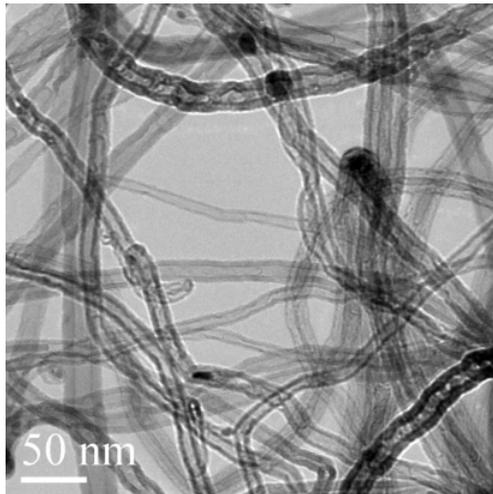
Growth Mechanism of Carbon Nanotubes (CVD)



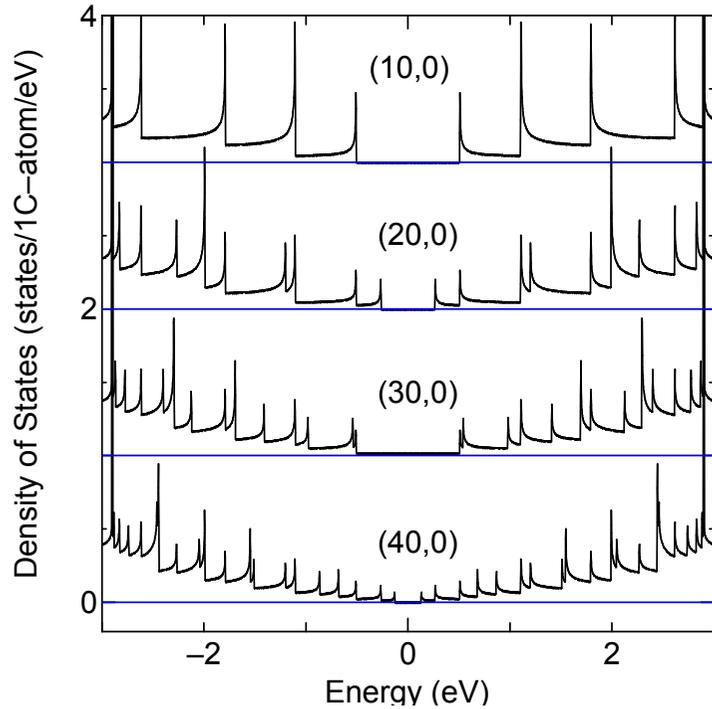
CNT

Characteristics

- External diameter: 15 – 25 nm
- Internal diameter: 5 – 10 nm
- Average number of nanotubes: 10 – 15

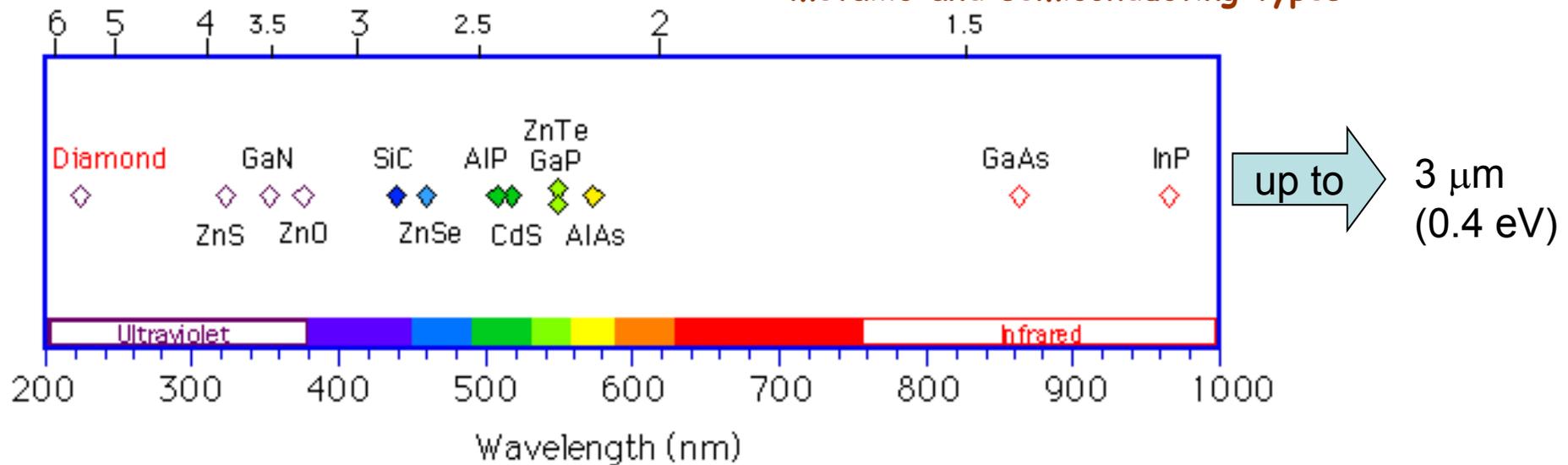


May CNT be used as photodetectors?

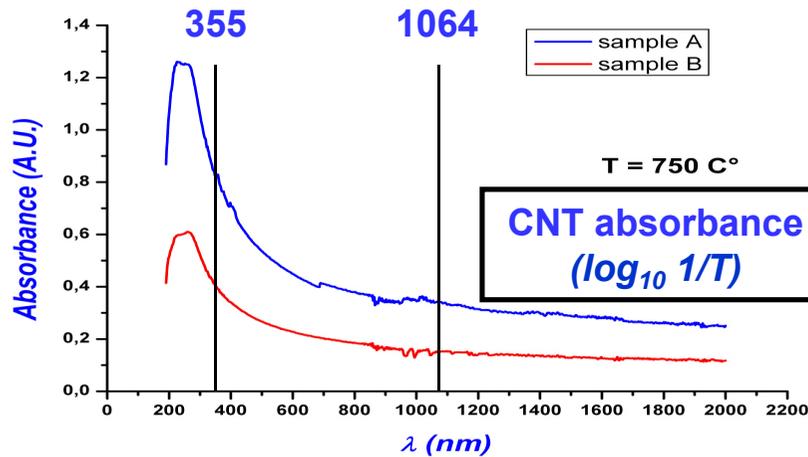
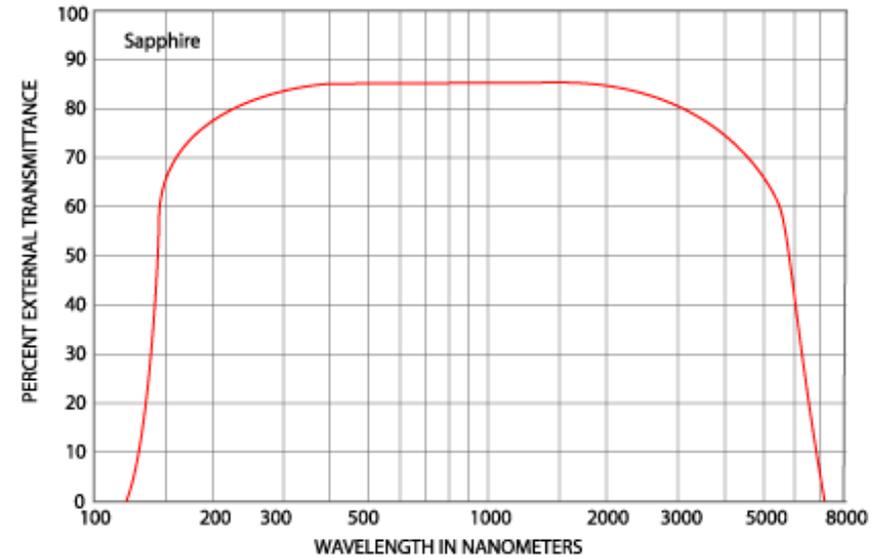
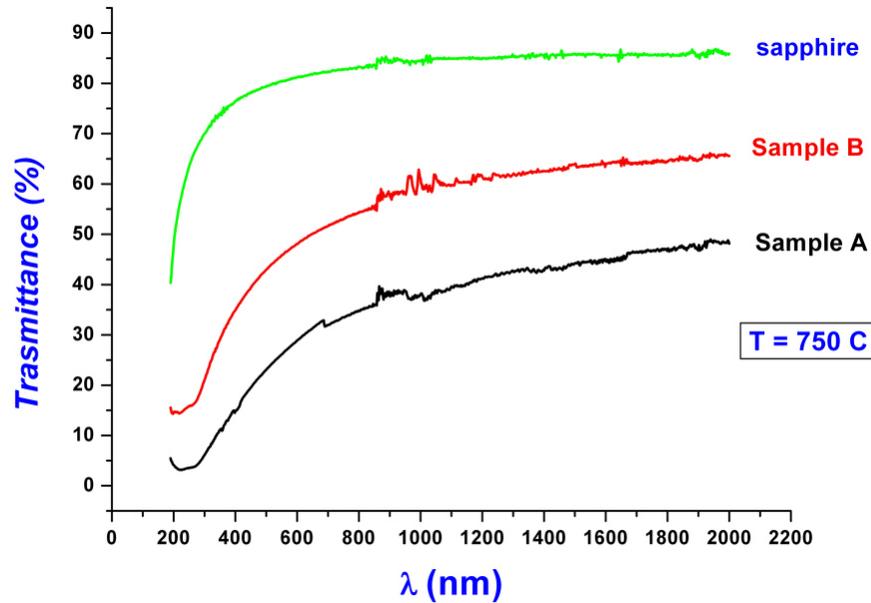


A layer of Multiwall Carbon Nanotubes covers a wide range of diameters and chirality, offering a device sensitive to a wide range of radiation frequencies. In addition the CNT density is very high, allowing, even in a small area, a great number of tubes sensitive to the radiation: $\approx 10^8 - 10^{10}$ MWCNT / 1 mm^2

A multiwall carbon nanotube typically consists of a concentric set of nanotubes of both metallic and semiconducting types



MWCNT photo absorbance

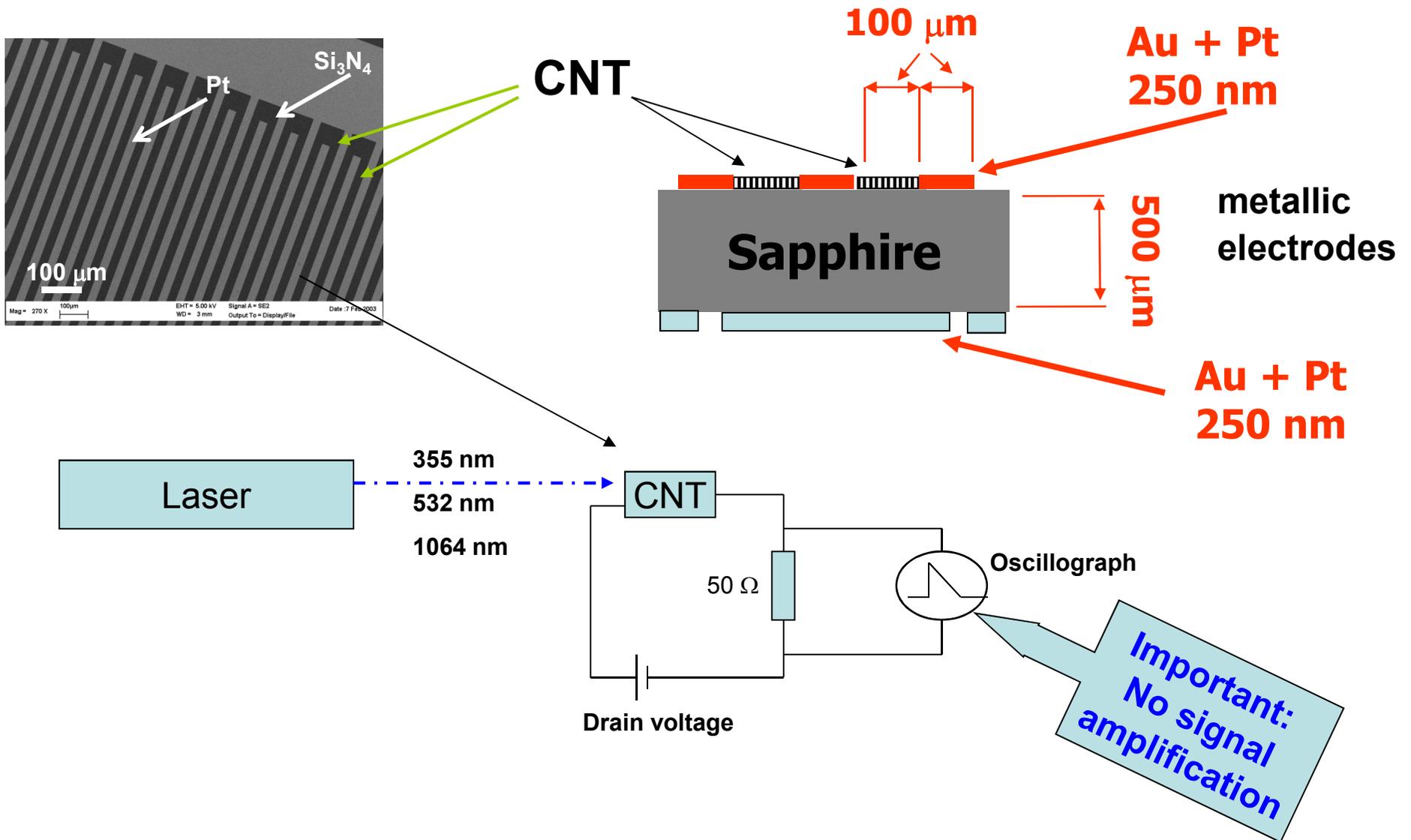


The UV radiation appears more absorbed than the visible and infrared (IR) photons.

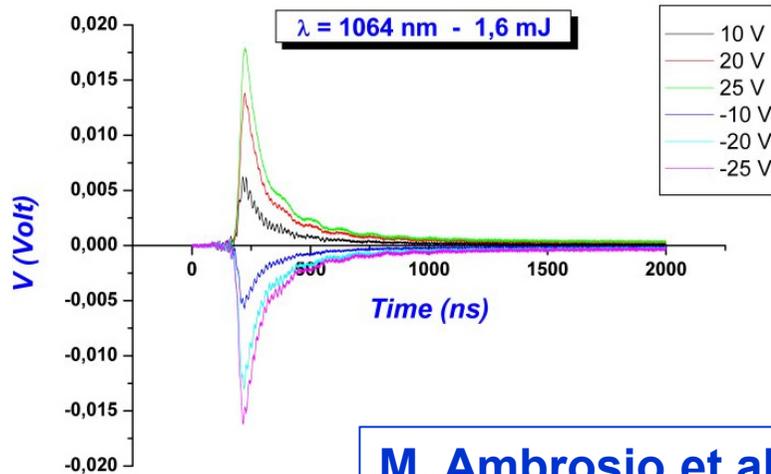
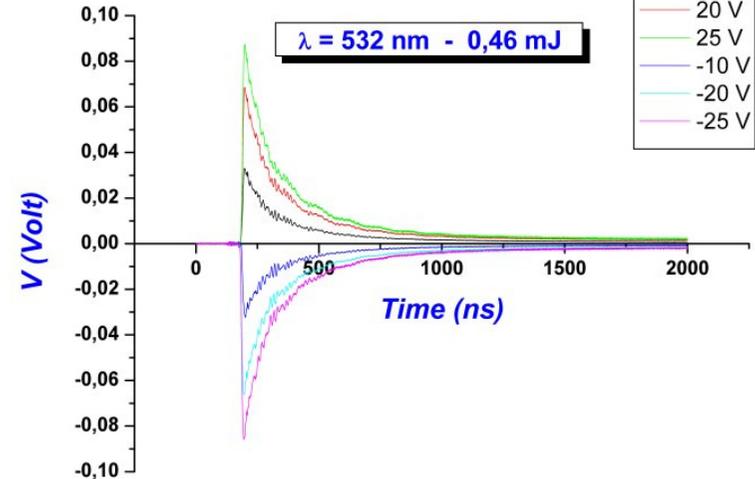
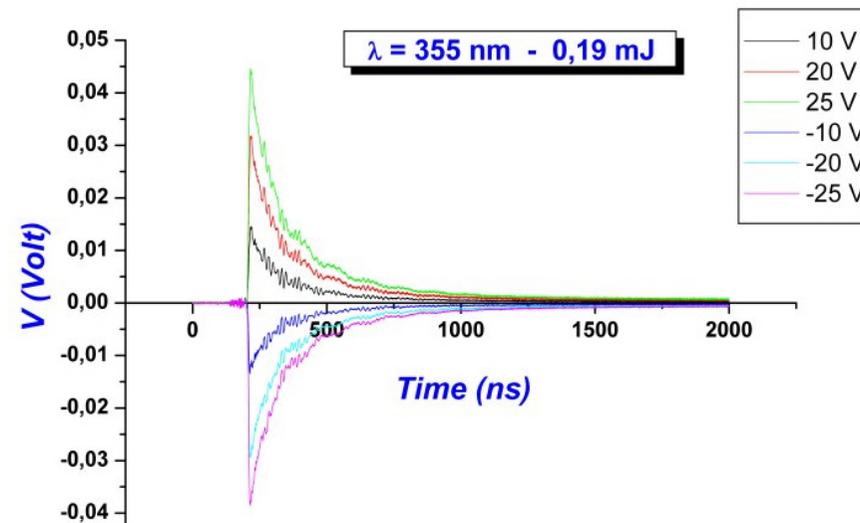


CNTs on sapphire

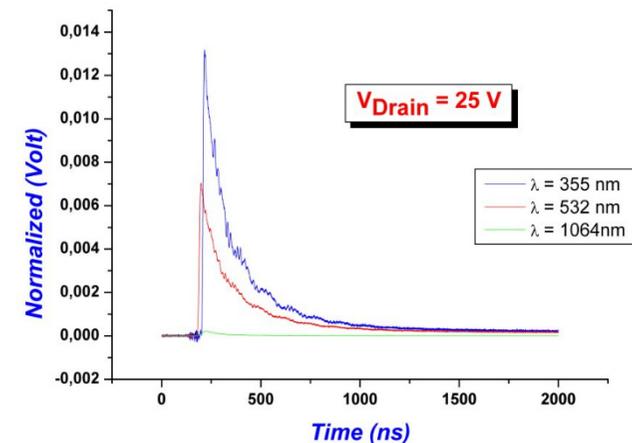
Sapphire prototype detector



Detected Signals on Sapphire



Comparison at different λ

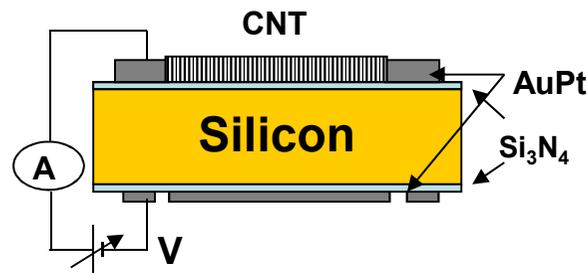
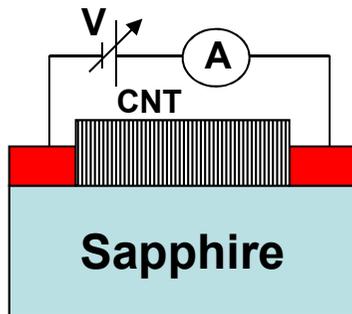
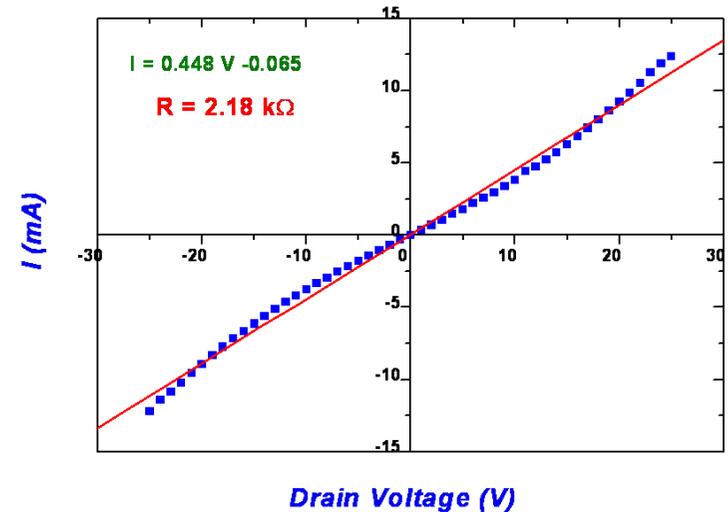


M. Ambrosio et al. VCI 2007

The response of the MWCNT carpet to a monochromatic pulsed light, showing the highest and faster detection efficiency for the UV light. These characteristics make the material highly interesting for UV photodetection.

Main problems with sapphire photodetector

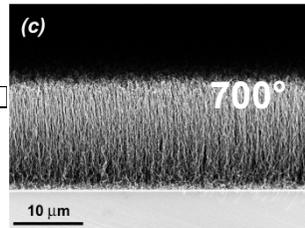
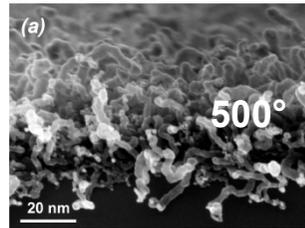
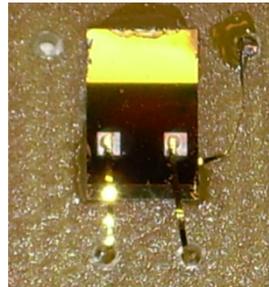
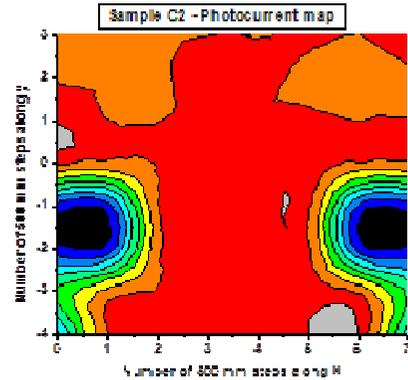
1. High dark current between metallic electrodes;
2. Low level of photocurrent (no signal amplification can be applied without amplifying the dark current also).



The use of a layered silicon substrate permits to collect the generated photocurrent through the silicon avoiding the surface dark current.

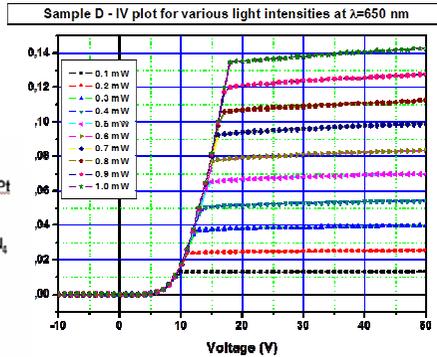
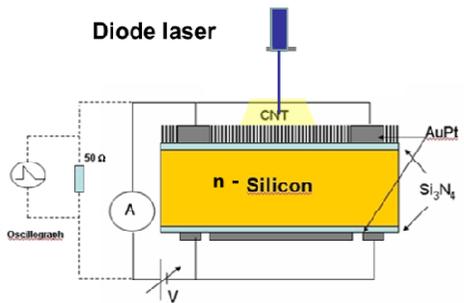
Solution: create a MWCNT-Si photosensitive heterojunction in which CNTs film converts photons into electrons and the layered silicon substrate amplify the signal by means of a secondary discharge.

The results of first MWCNT-Si device



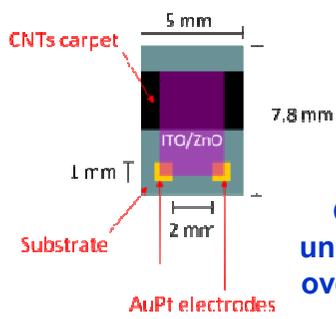
Electrical analysis of carbon nanostructures/silicon heterojunctions designed for radiation detection, NIM A 629, 377–381 (2011)

Innovative carbon nanotube – silicon large area photodetector, 2012 JINST 7 P08013 2012.



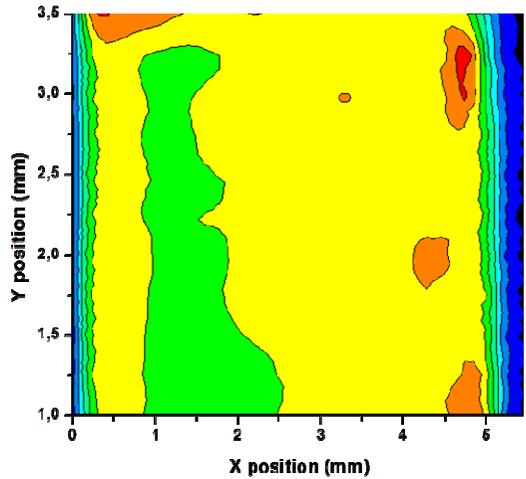
Ni Catalyst deposition

Coating

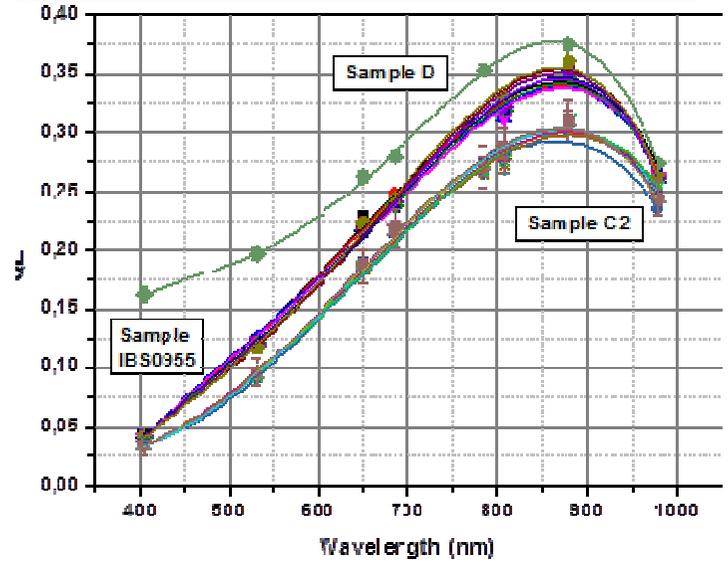


Great uniformity over large area

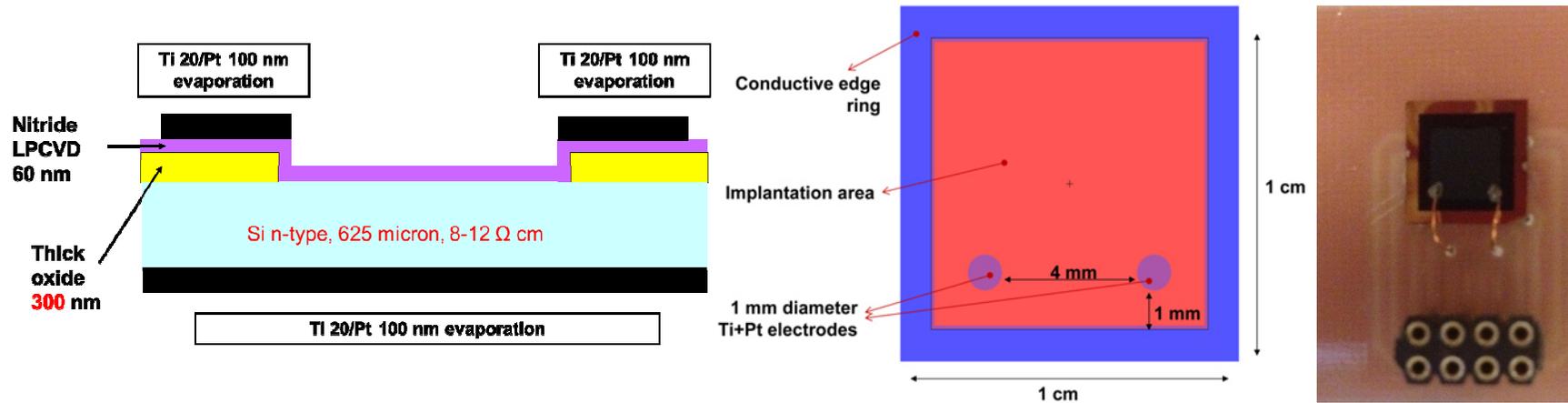
Map of sample IBS0955



QE comparison for samples C2_500°, D_700° and IBS0955_500°

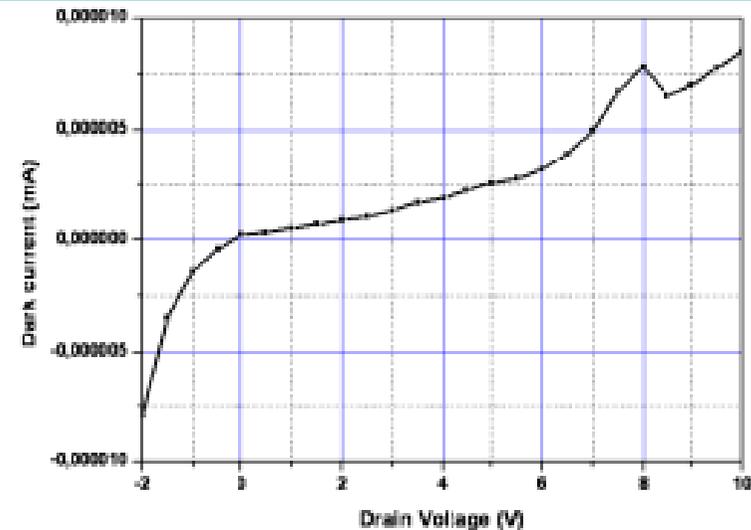
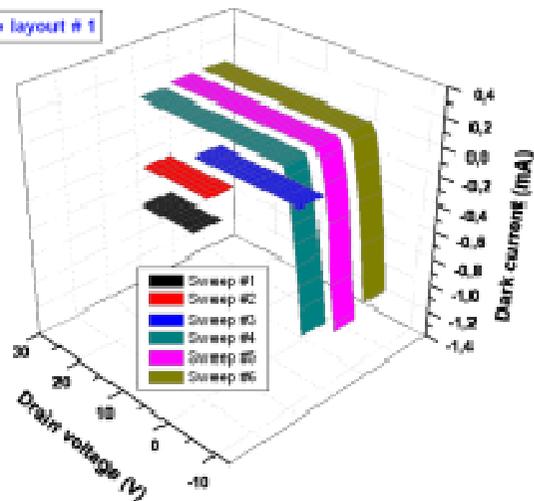


The FBK substrate

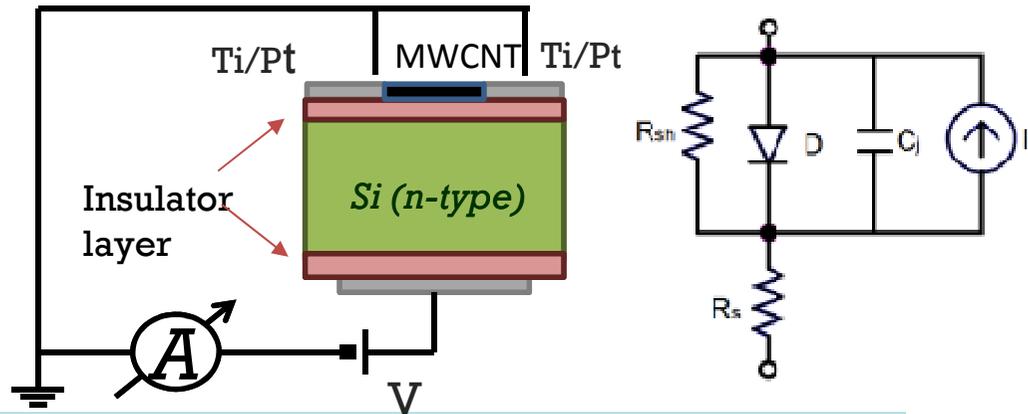


The device substrate has the structure of a Metal-Insulator-Silicon (MIS) photodiode: applying an external electrical field between the two electrodes on the front and on the back of silicon layer and operating voltage sweeps the dark current assumes a straighten behavior typical of a diode .

Substrate layout # 1

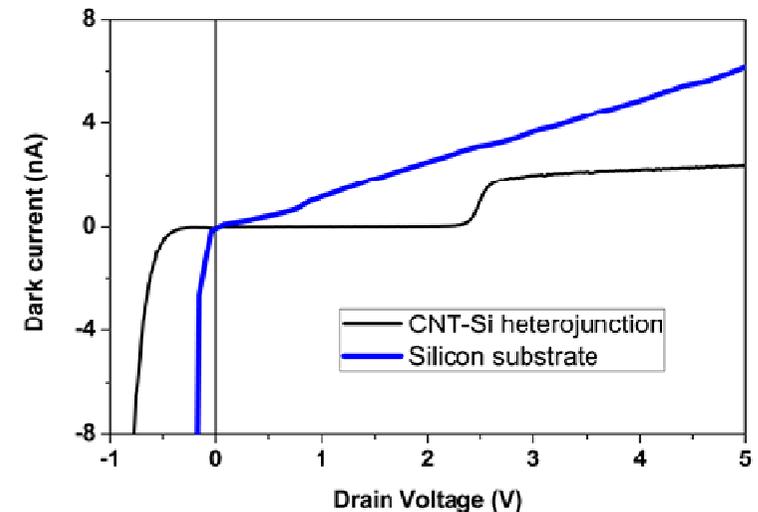
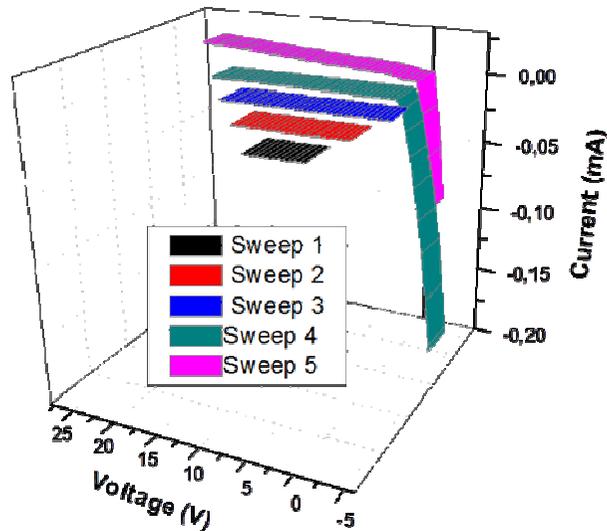


Large area MWCNT-Si radiation detector



An electrical field can be applied between the two sides of device obtaining a large area photodiode whose equivalent circuit is depicted in figure, where D is the device, I_L is the photocurrent generated by the incident light, C_j is the heterojunction capacity, R_{sh} the shunt resistance and R_s the circuit resistance.

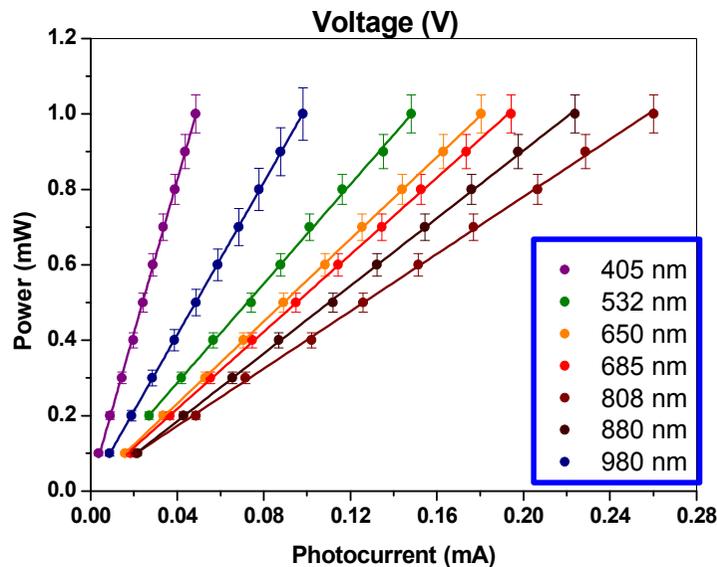
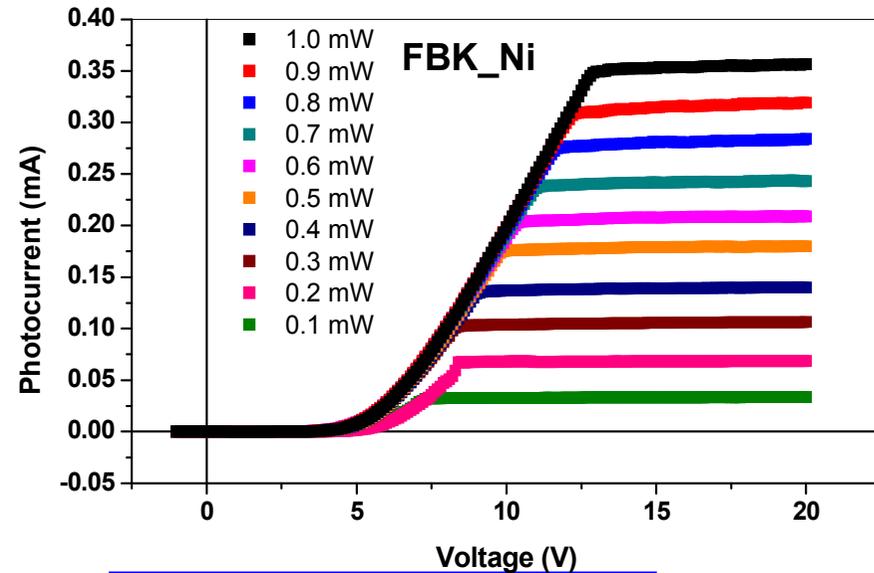
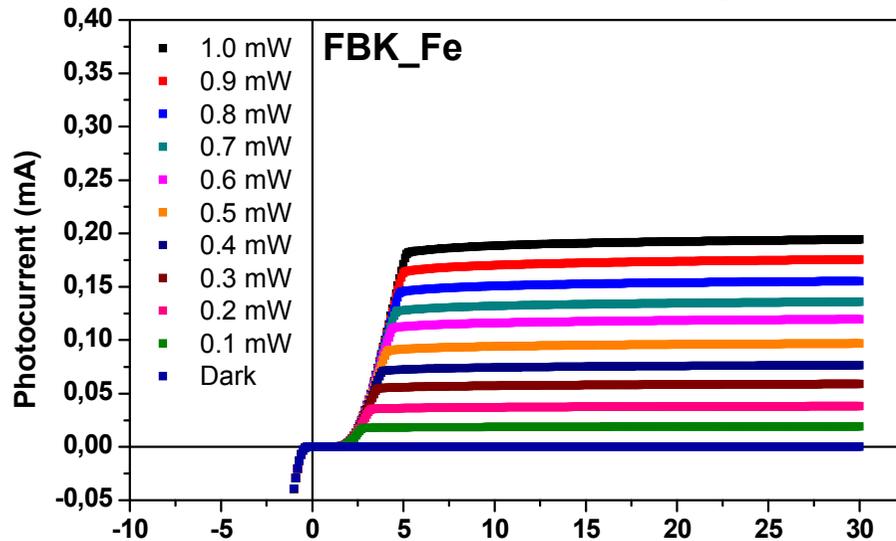
The application of voltage sweeps to the device generates the heterojunction between CNT and silicon substrate: the dark current measured between electrodes in the front and in the back of device assumes a threshold behavior.



The heterojunction is characterized by a well defined threshold at 2.4 V completely absent in the substrate without CNTs. Dark current is about 2 nA for a surface of 0.5 cm^2

Photocurrent and linearity

685 nm light emitting diode (LED) laser

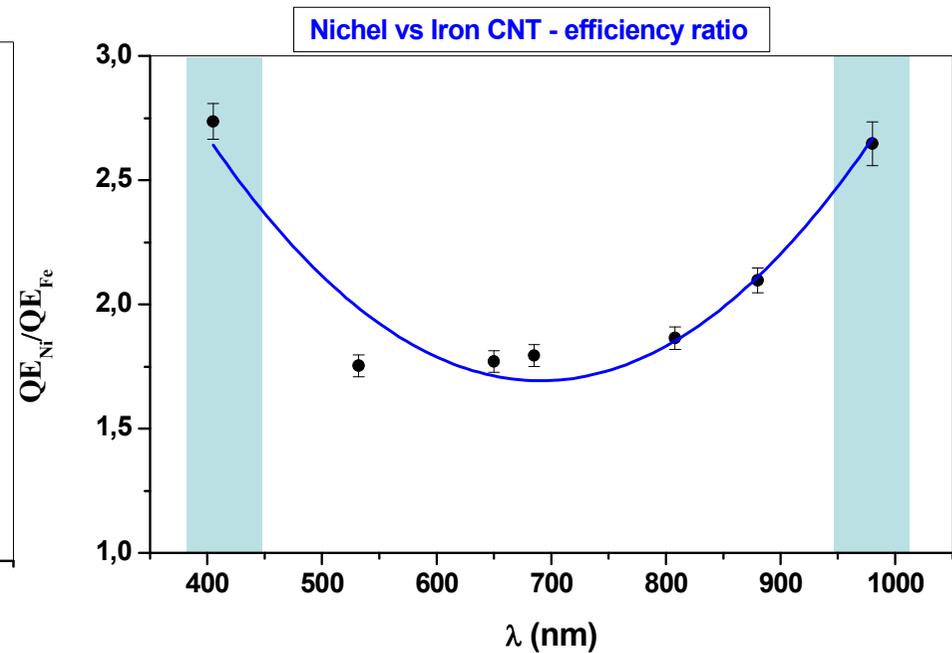
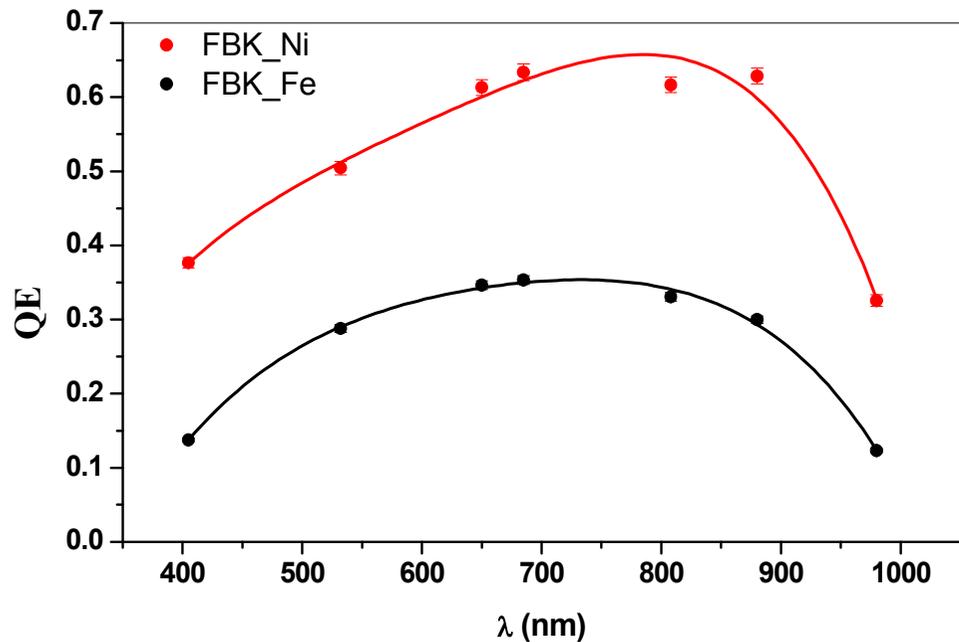


- Room temperature
- No electronics
- No signal amplification
- Long and stable plateau
- Linearity I vs P
- Threshold
- No saturation observed
- Uniformity on all the CNT surface
- Long term stability (>8 years)
- Breakdown @ >100 V

Quantum Efficiency

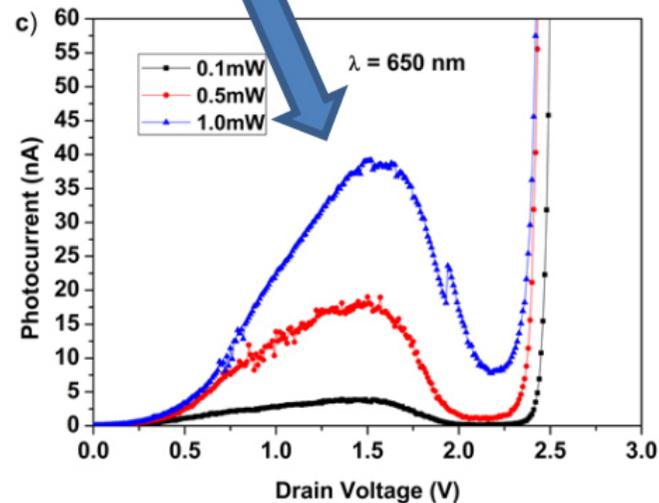
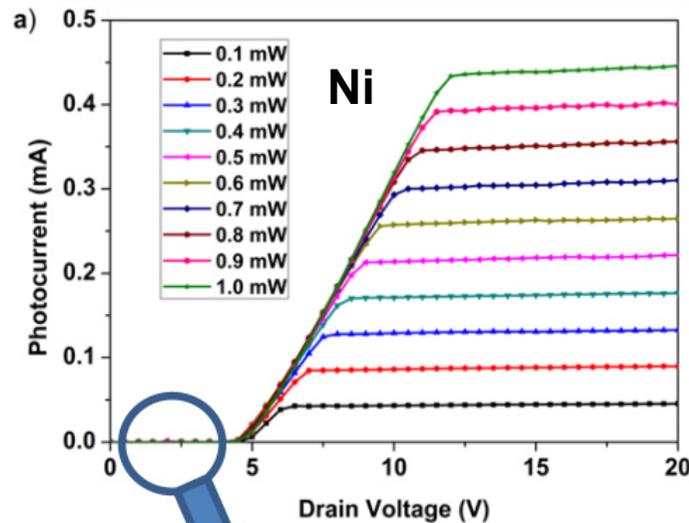
The quantum efficiency (QE) of the device was estimated as the ratio of the number of electrons collected at plateau (I) to the number of incident photons according to the relation:

$$QE \pm \sigma_{QE} = \frac{hcl}{\lambda eP} \pm \frac{hc}{e} \sqrt{\left(\frac{1}{\lambda P}\right)^2 \sigma_I^2 + \left(\frac{I}{\lambda^2 P}\right)^2 \sigma_\lambda^2 + \left(\frac{I}{P^2 \lambda}\right)^2 \sigma_P^2}$$



Quantum efficiency of CNTs with Ni catalyst appears much higher than that of CNTs with Fe catalyst, especially in the UV and IR regions.

Tunnel effect



The shape of the current-voltage curve presents a negative differential resistance and resembles that of a resonant tunneling junction.

These observations clearly indicate that impinging light and, as a consequence, the photogenerated charges play a fundamental role in the heterojunction behaviour.

The current shape similarity with that of typical resonant tunneling junction suggests that a kind of electronic resonance process induced by the photogenerated charges may be present.

Resonant tunnel-like current is generated only under light radiation and it is function of the wavelength as well as of the power intensity

Aramo C. et al: *Beilstein J. Nanotechnol.* 2015, 6, 704–710

Study of heterojunction MWCNT-Si



Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima



Electrical analysis of carbon nanostructures/silicon heterojunctions designed for radiation detection

A. Tinti^{a,*}, F. Righetti^a, T. Ligonzo^a, A. Valentini^a, E. Nappi^a, A. Ambrosio^b, M. Ambrosio^c, C. Aramo^c, P. Maddalena^b, P. Castrucci^d, M. Scarselli^d, M. De Crescenzi^d, E. Fiandrini^e, V. Grossi^f, S. Santucci^f, M. Passacantando^f

^a INFN, Sezione di Bari, Via Amendola 173, 70126 Bari, Italy

^b CNR-SPIN U.O.S. di Napoli e Dipartimento di Scienze Fisiche, Università degli Studi di Napoli Federico II, Via Cintia 2, 80126 Napoli, Italy

^c INFN, Sezione di Napoli, Via Cintia 2, 80125 Napoli, Italy

^d Dipartimento di Fisica, Università degli Studi di Roma Tor Vergata, Via della Ricerca Scientifica 1, 00133 Roma, Italy

^e INFN, Sezione di Perugia e Dipartimento di Fisica, Università degli Studi di Perugia, Piazza Università 1, 06100 Perugia, Italy

^f Dipartimento di Fisica, Università degli Studi dell'Aquila, Via Vetoio 10, 67100 Coppito, L'Aquila, Italy

**Nuclear Instruments
and Methods in
Physics Research A
629 (2011), 377-381**

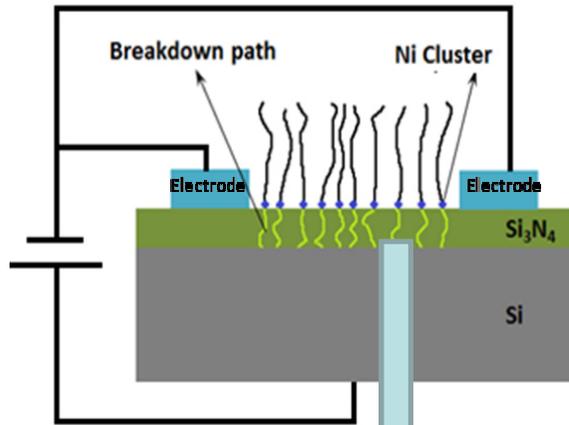
ABSTRACT

A new class of radiation detectors based on carbon nanostructures as the active photosensitive element has been recently developed. In this scenario the optimization of the device, both in dark and on light irradiation, is a crucial point. Here, we report on electrical measurements performed in dark conditions on carbon nanofibers and nanotubes deposited on silicon substrates. Our experimental results were interpreted in terms of a multistep tunneling process occurring at the carbon nanostructures/silicon interface.

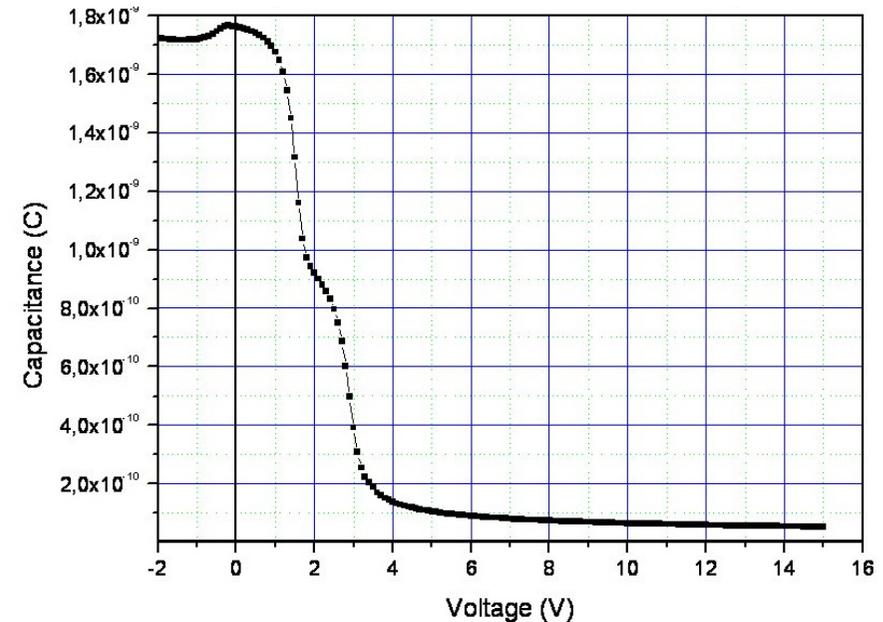
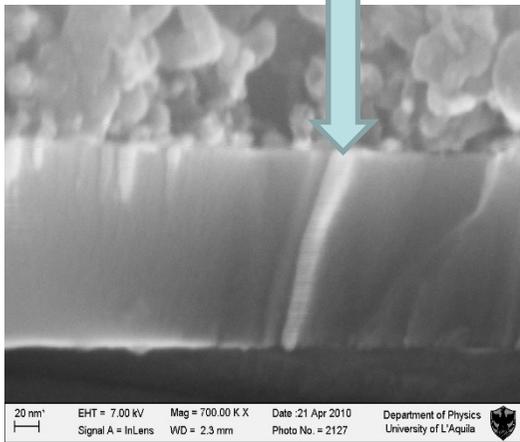
Keywords:
Carbon nanotubes

© 2010 Elsevier B.V. All rights reserved.

Key point → how the hetero-junction creates?

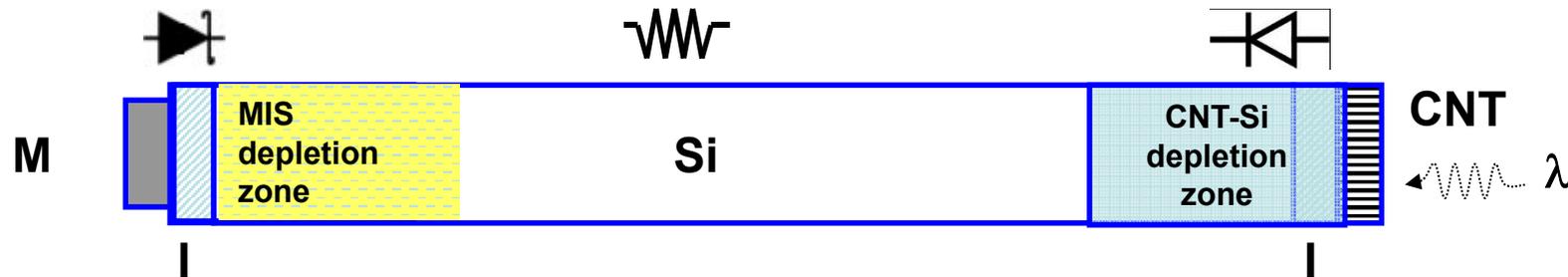


The presence of the insulator is bypassed by the formation of permanent low resistivity conduction channels which are created as an external voltage is applied to the device thanks to the peculiar characteristics of the nanotubes to appear as uni-dimensional elements.



The C-V plot indicates the change of capacitance at the same drain voltage threshold of 2.4 V. The MWCNTs act as a semi-metallic layer, the silicon substrate is n-doped and the nitrite layer is the pierced insulating dielectric.

Phototransistor-like device configuration



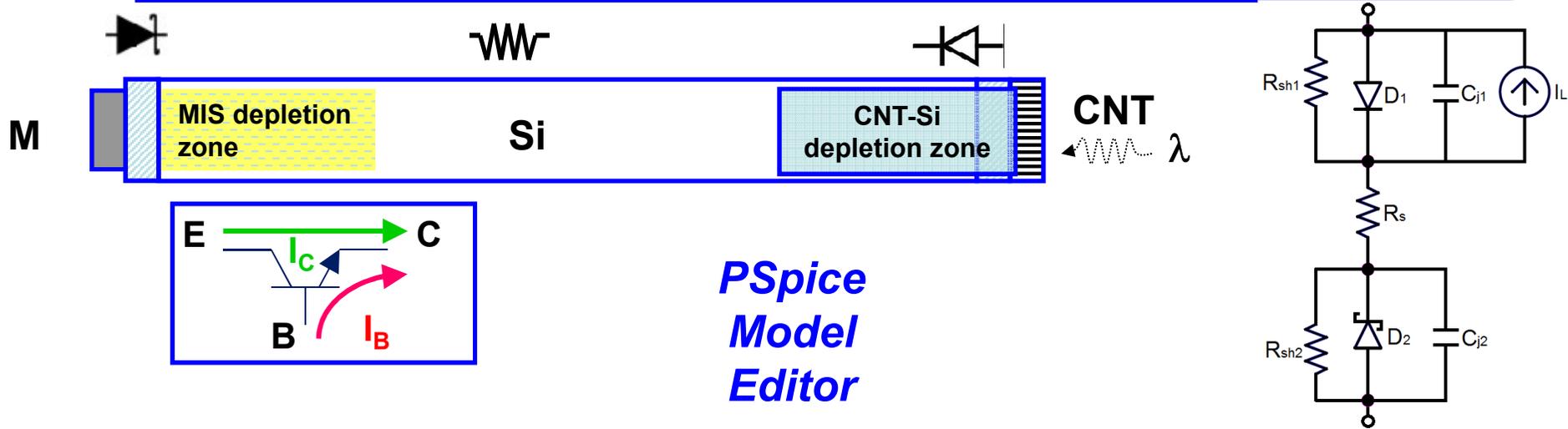
Depletion regions amplitude depends on the applied voltage. The silicon bulk, 625 μm thick, can be considered as a passive resistance except in the depletion areas.

The MIS depletion area cannot be reached from optical radiation because of the presence of metal.

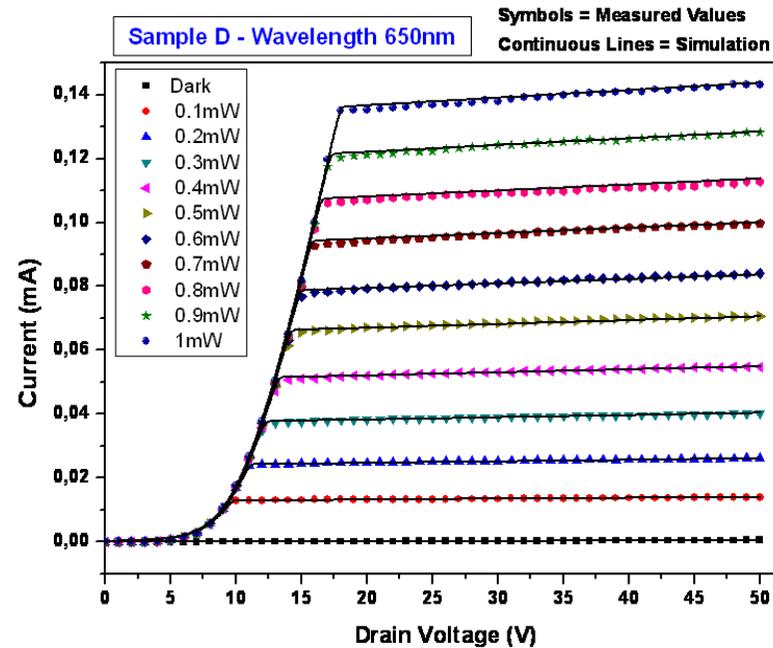
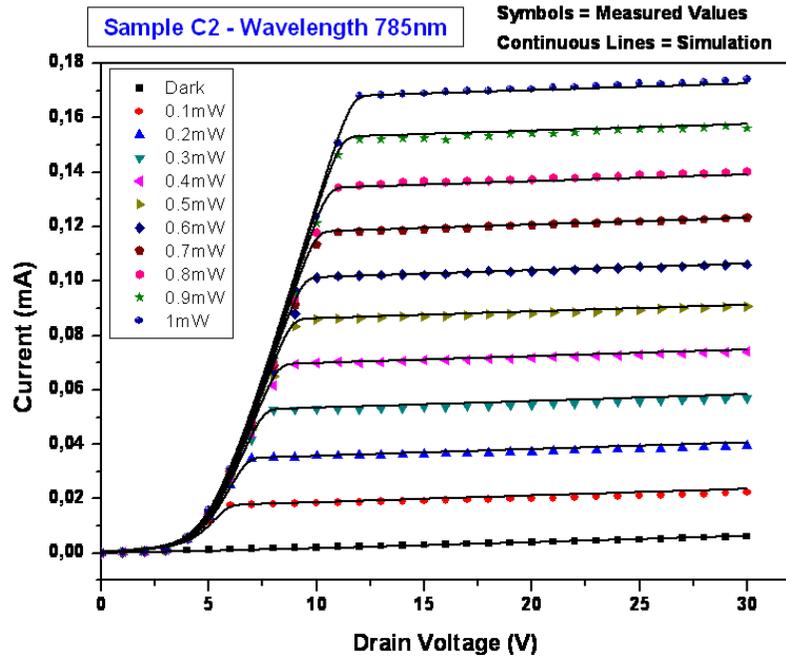
Instead the depletion area created by the CNT-Si heterojunction can be reached and activated by the radiation due to the nanotube characteristics. A light's fraction is absorbed inside CNT and converted in hole-electron pairs or excitons. The other reaches silicon and is converted inside.

The described device can be considered as a “**phototransistor**”

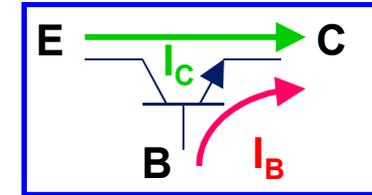
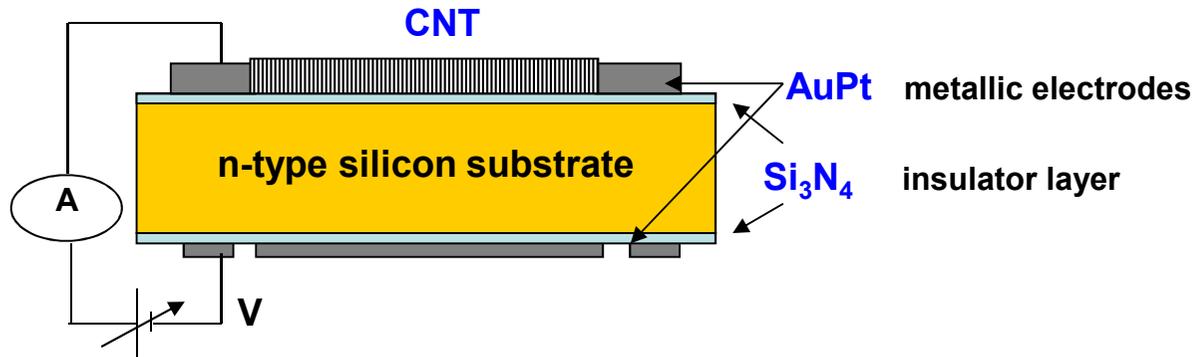
The p-n-p Phototransistor



*PSpice
Model
Editor*

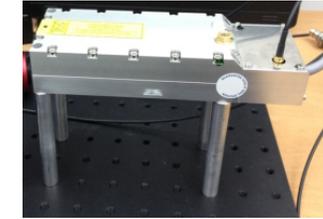


Key points of a CNT-Si photodetector



1. A large area photodetector build by simply growing CNTs on a n-type silicon substrate;
2. a CNT-Si heterojunction is created applying voltage sweeps to the electrodes;
3. the device presents a great uniformity over all the sensitive area;
4. QE and photoresponsivity are comparable to that of commercial photodiode;
5. charge collected presents a great linearity and saturation effects have been observed;
6. photosensistivity extends in a wide wavelength range, from UV to IR;
7. the device can be used both to detect continuous and pulses light;
8. at present no amplification – can detect light pulses with large number of photons ($>10^4$)

355 nm pulsed laser



Teem Photonics

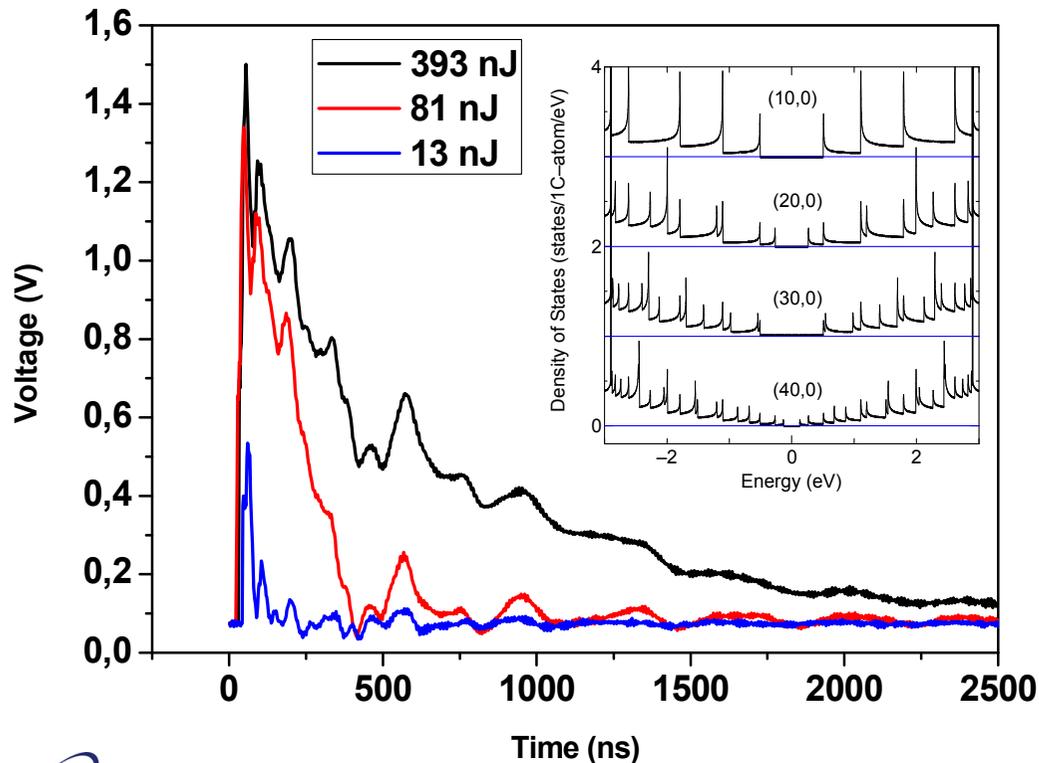
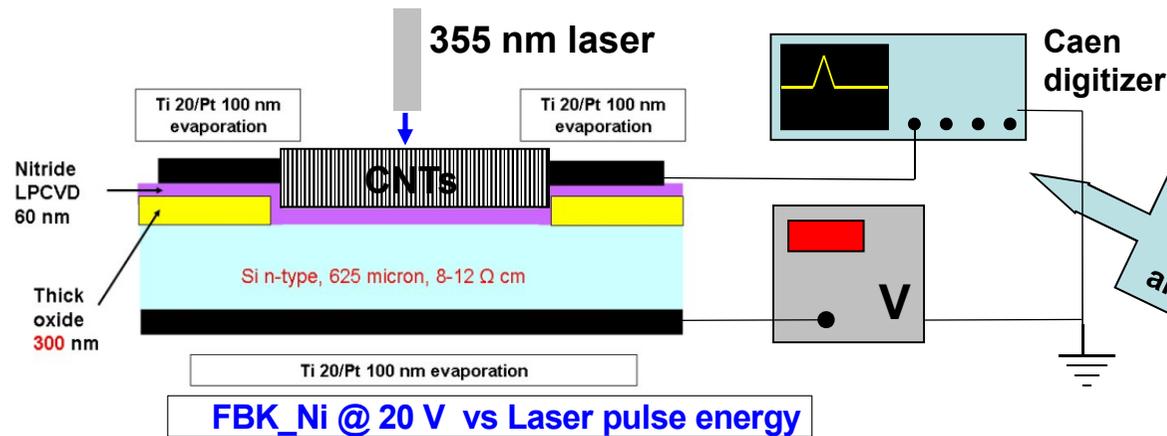
Q switched laser

mod. STV-01E-030

2 microJ at 355 nm

1KHz trigger

pulse width < 400 ps



The signal of Ni catalysed CNT-Si detector due to pulsed signals @ 355 nm is fast, with a rise time of few ns.

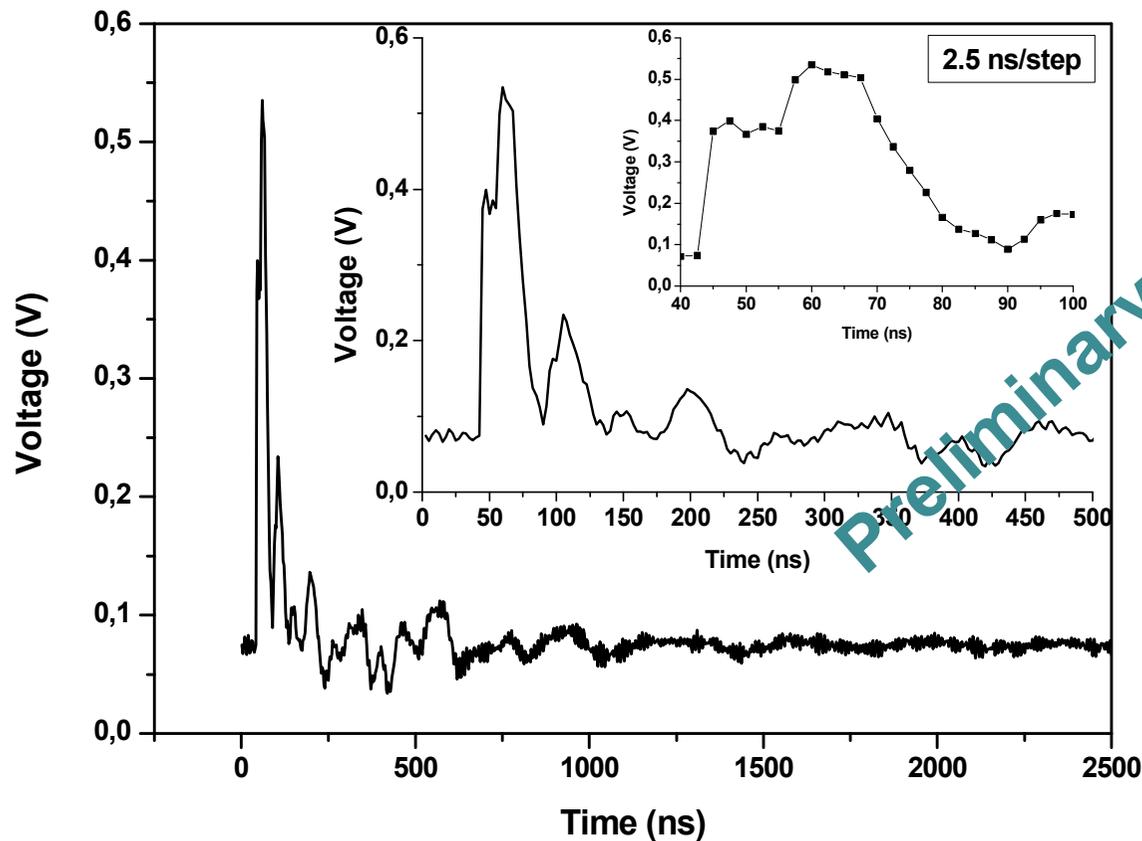
The signal peak, charge and length depend from the laser intensity.
At 13 nJ the peak is ≈ 0.5 V over 50Ω .

As the laser energy beam increases the photodetector signal height raises and its duration becomes larger.

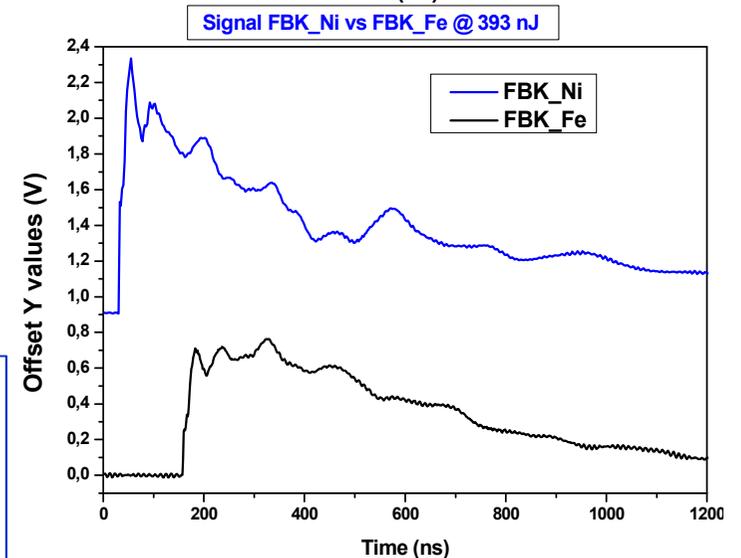
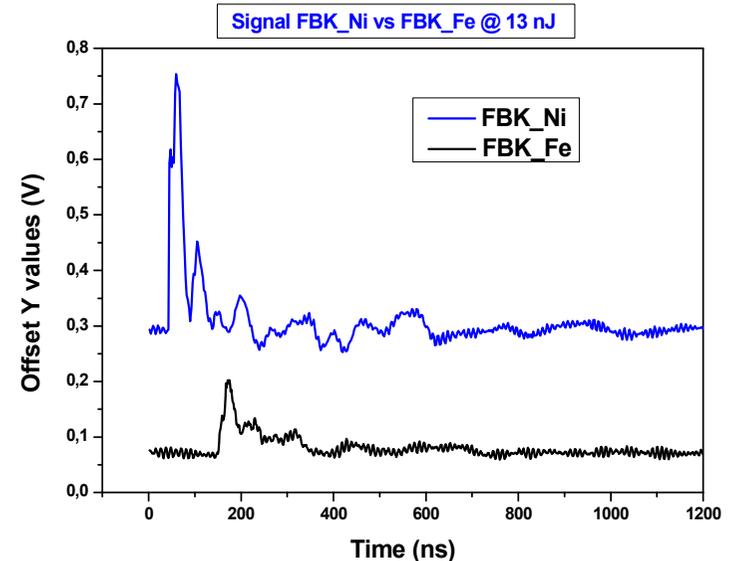
The great signal duration at higher energy probably is due to long time decay metastable states of CNTs excited by beam, whose peaks are well visible as the energy increases.

Details of photodetector signal

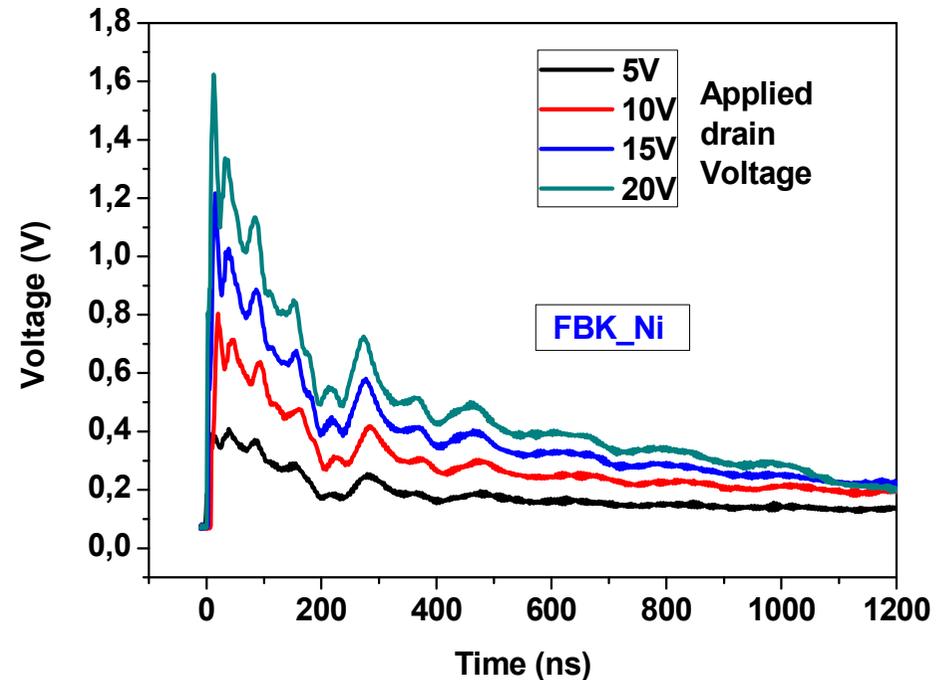
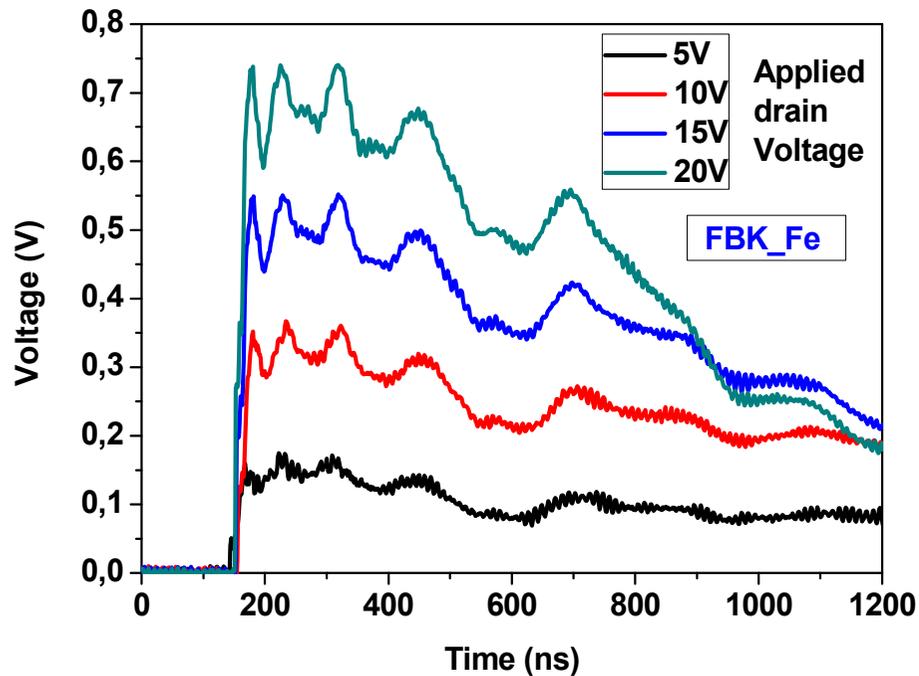
FBK_Ni @ 20 V - Laser energy = 13 nJ



- Rise time of first peak less than 2.5 ns @ 13 nJ
- Signal from Ni catalysed CNTs larger and faster than Fe
- As the laser power increases the number of secondary delayed peaks increases also enlarging the photodetector signal



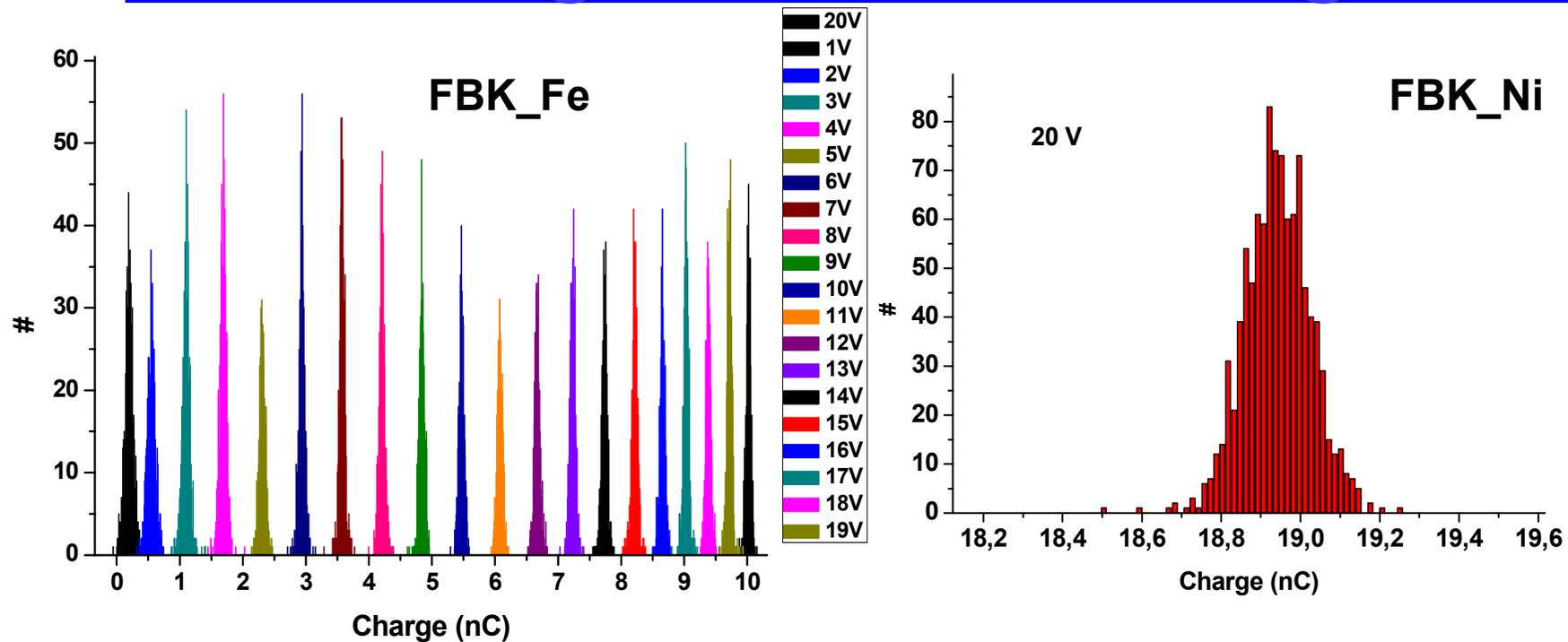
Signal vs drain voltage @ 81nJ



The signal shape strongly depends from the CNT catalyst and from the laser light intensity, less from the drain voltage applied.

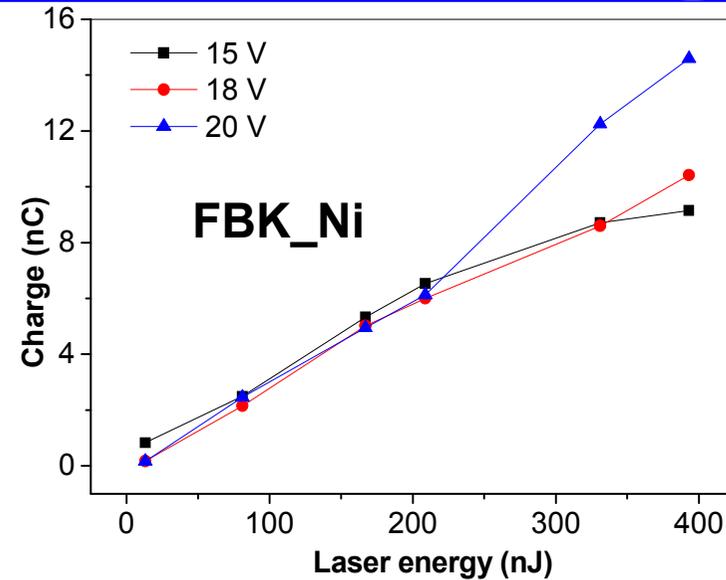
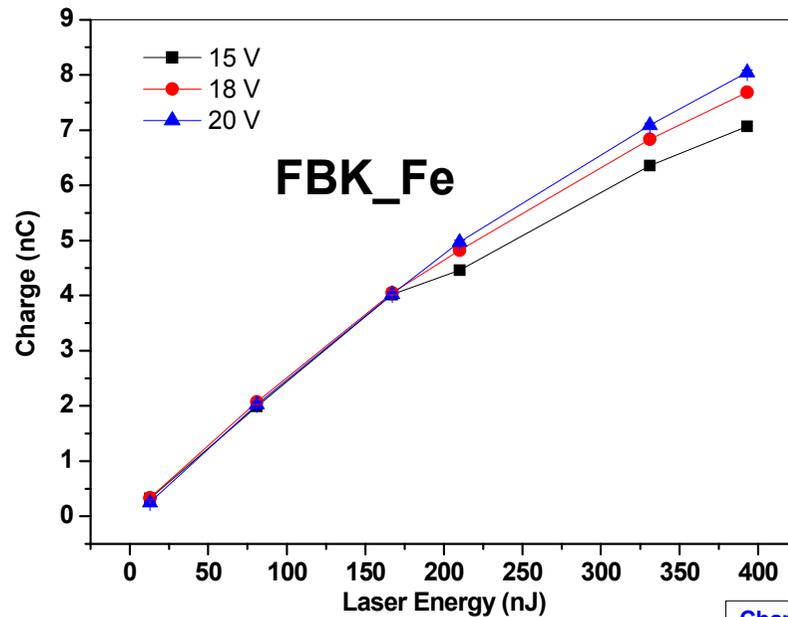
The incident radiation excites CNTs energetic levels and the drain voltage collects the generated charge. **No saturation effects have been observed.**

Charge vs drain voltage

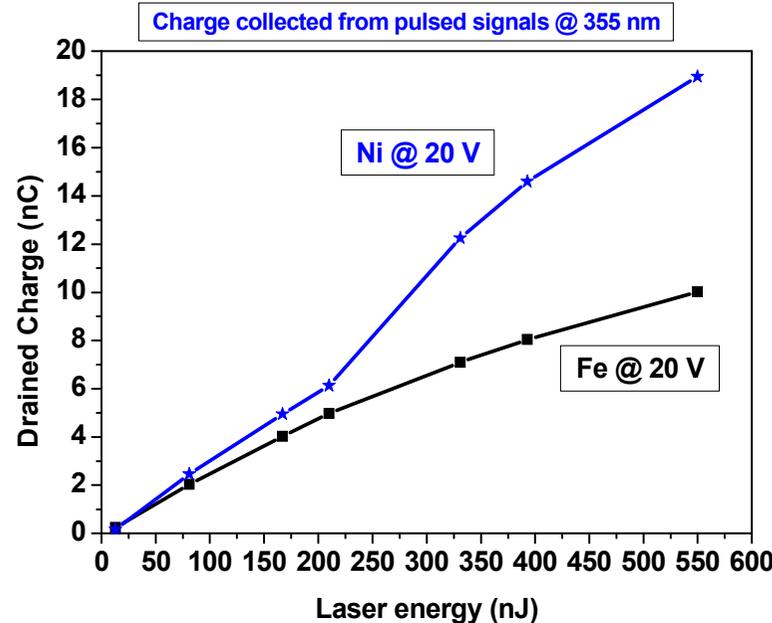


- The MWCNTs with Ni catalyst shows an higher charge generation than Fe.
- In spite of the high light intensity the photodetector charge is linear for all drain voltage range and not in saturation.
- The charge distributions are well separated for all drain voltage range.

Charge vs voltage and laser energy



- For higher intensities a higher drain voltage is requested to avoid the charge recombination inside CNTs.
- For Ni_CNTs it is need a bigger drain voltage due to a bigger amount of charge generated inside the device



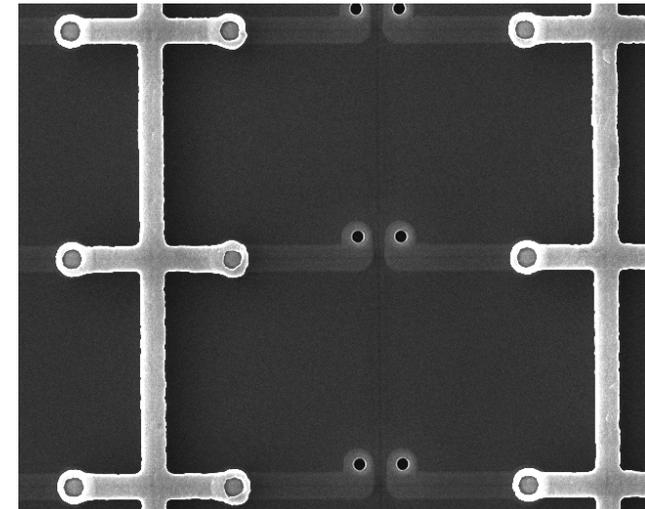
The collected charge from pulsed signals increases quite linearly with laser energy, that means no saturation effects are observed.

SiPM_CNT Project

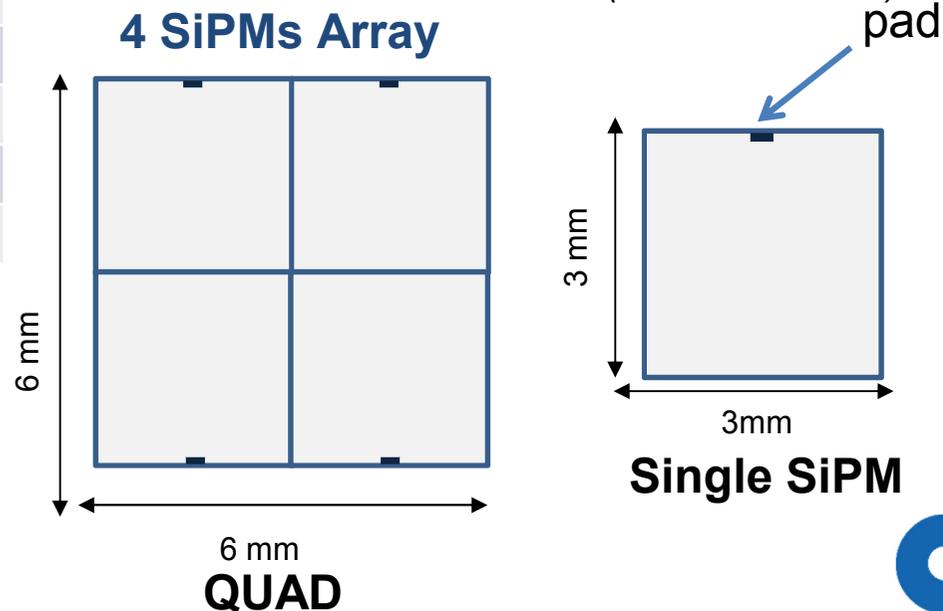
- SiPMs based on FBK NUV-HD technology and specifically designed for CNT

Typical specs of the standard NUV-HD SiPMs

Typical Parameters (@ room T)	NUV-HD CS = 25 μ m
Cell Size	25 μ m
Fill Factor	73%
Breakdown Voltage	26.5 V
Max PDE	50%
Peak PDE λ	410 nm
DCR (20°C)	< 150 kHz/mm ²
DiCT	25%
DeCT + AP	2%

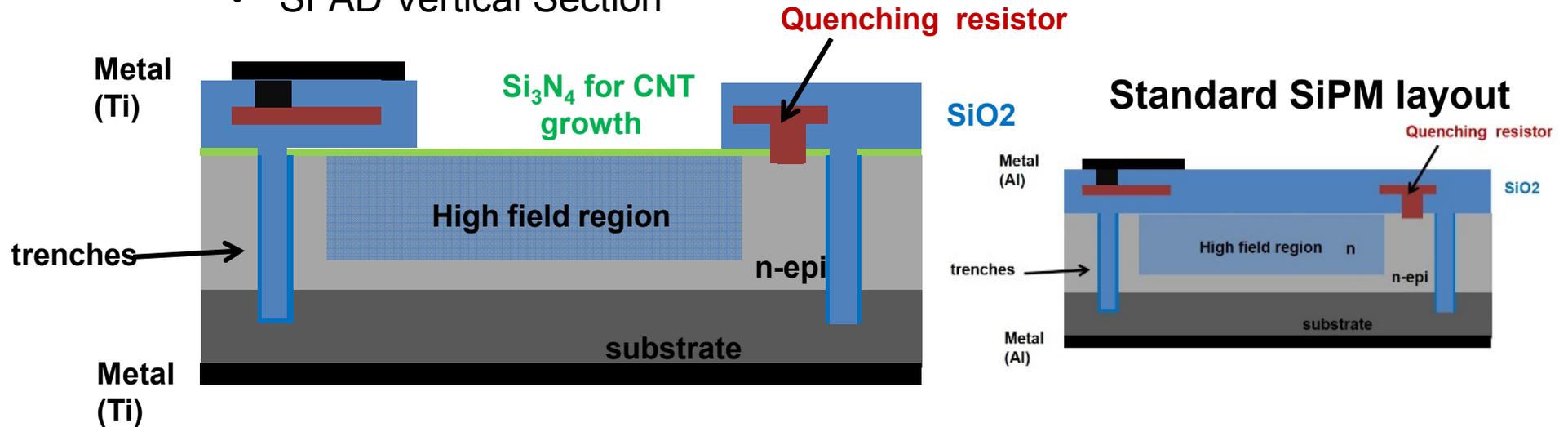


SEM Image of the single cell composing the SiPM (cell size = 25 μ m)



SiPM_CNT Layout

- SPAD Vertical Section



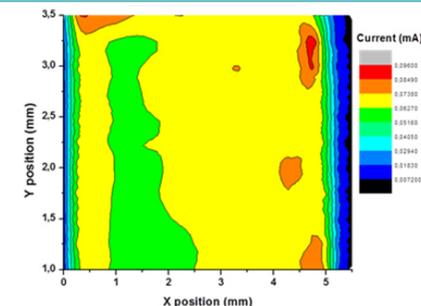
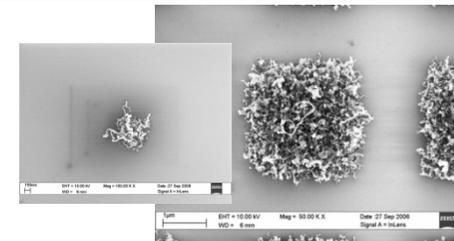
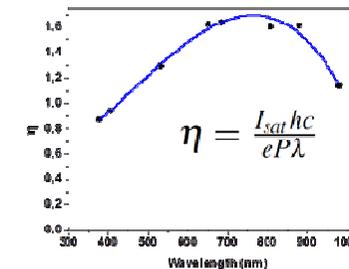
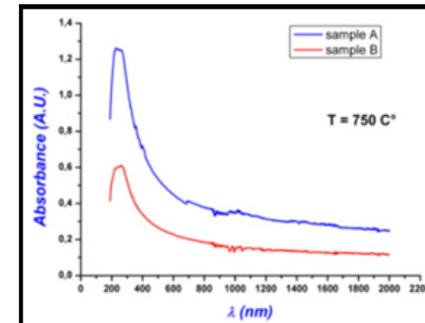
- Spad Size = $30 \times 30 \text{ um}^2$
- SPAD Fill factor = 78%

SiPM_CNT

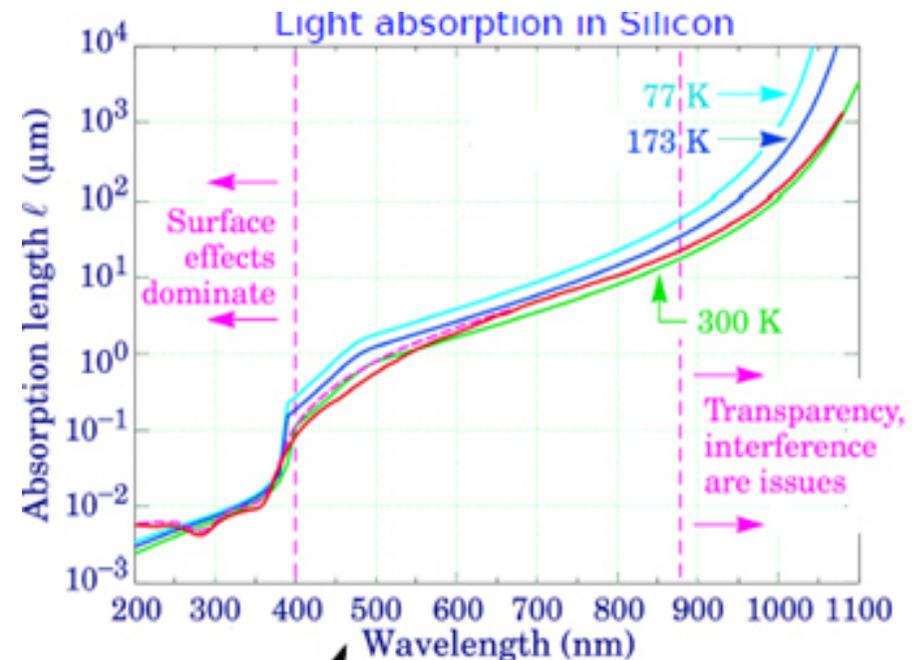
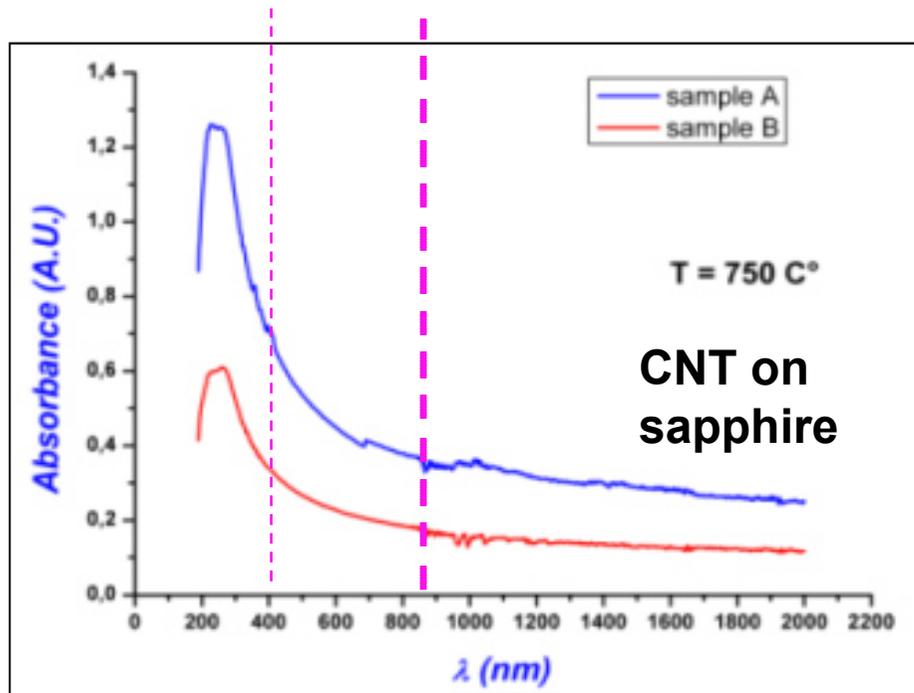
- **New SiPM structure** for high temperature tolerance
- **The CNT grown** directly on the device surface
- **Sensitive surface for every pixel** constituted by MWCNT-Si hetero-junction

Main Scientific Challenges

- ✓ **Increase quantum efficiency** due to the photoresponsivity enhancement from MWCNT absorbance, in the wavelength range 200-400 nm.
- ✓ **Decrease dark noise** thanks to hetero-junction MWCNT/Si → increase the device area (cm² or more VS mm² SiPM).
- ✓ **MWCNT bottom-up technology** → cells (pad or strip) hundreds of nm up to mm VS 15 μm up to 100 μm for SiPM.
- ✓ **Maximizes the sensitive area for photo-conversion and its uniformity** → Electric field applied via a conductive coating.



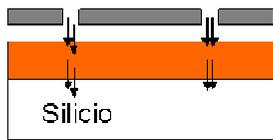
Light absorption



The CNT layer absorbs UV photons producing photoelectrons that can be transferred by means of an external electrical field inside the depletion zone of the hetero-junction. Therefore the charge induces a photocurrent drained out by the applied voltage.

Nanolithography and patternization

Electron beam exposure



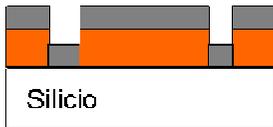
GDSII mask design



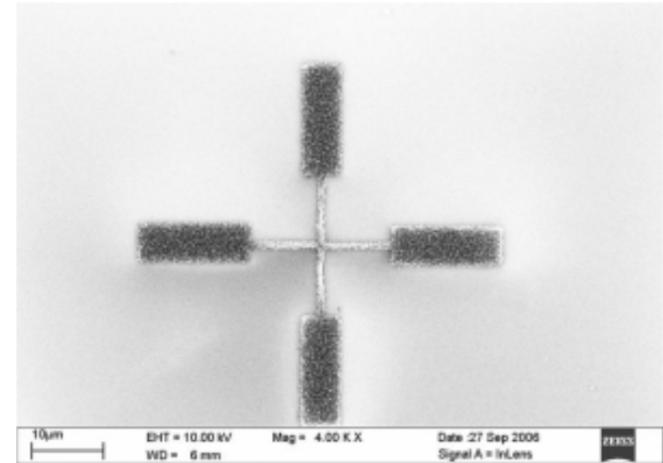
After developing



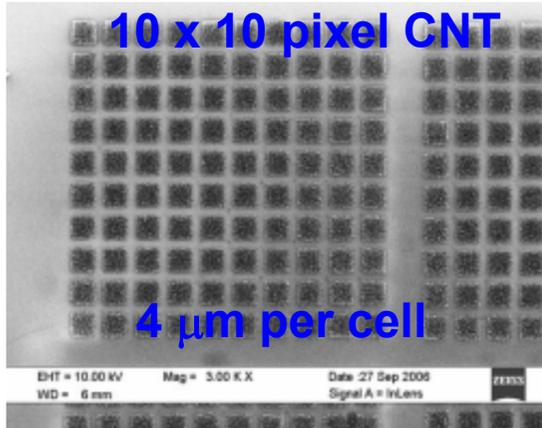
Nichel film deposition



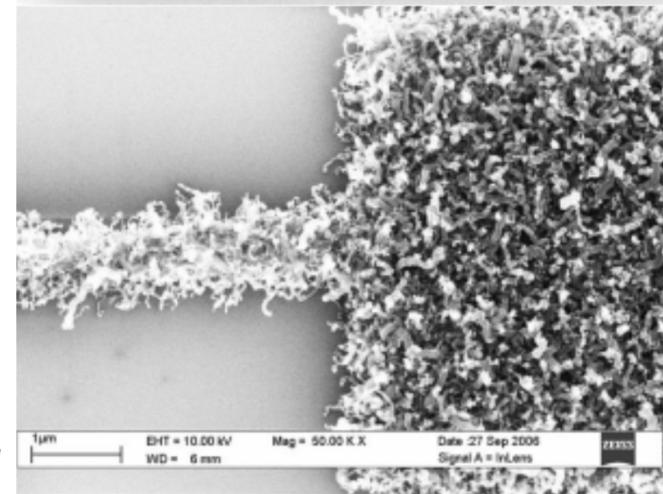
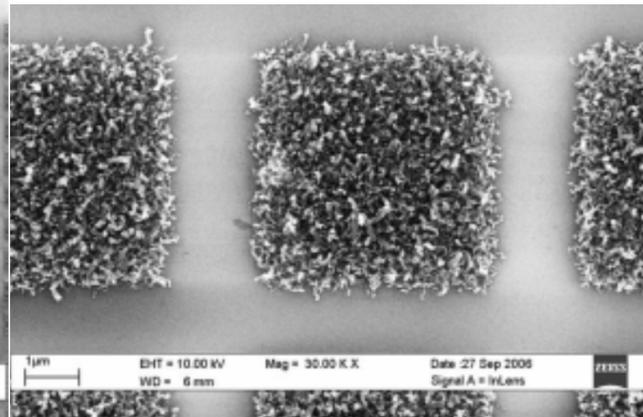
Aceton bath



10 x 10 pixel CNT

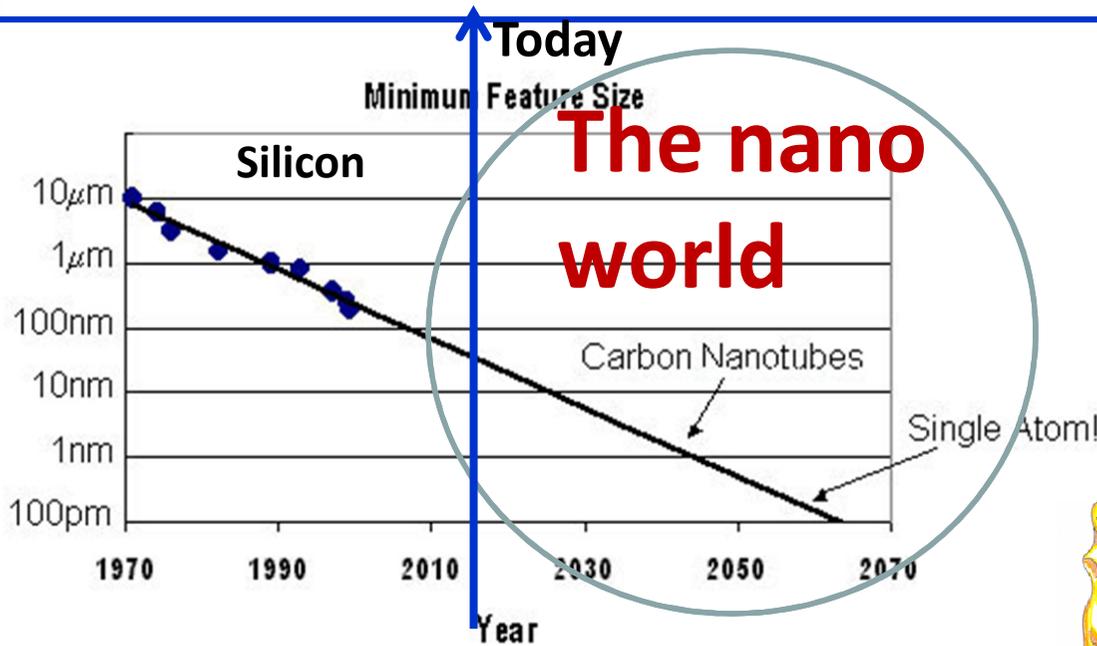


4 µm per cell



Nano-pixelled photocathodes sensitive to the UV radiation may be obtained by means of nanolithography in a very cheap and easy way!

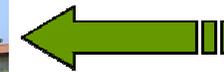
From micro to nano world



How long we can continue with the Moore's law?



Top - Down



Bottom - Up



The ability of silicon semiconductor to efficiently conduct electric current is lost at a few nanometers → carbon nanotubes.

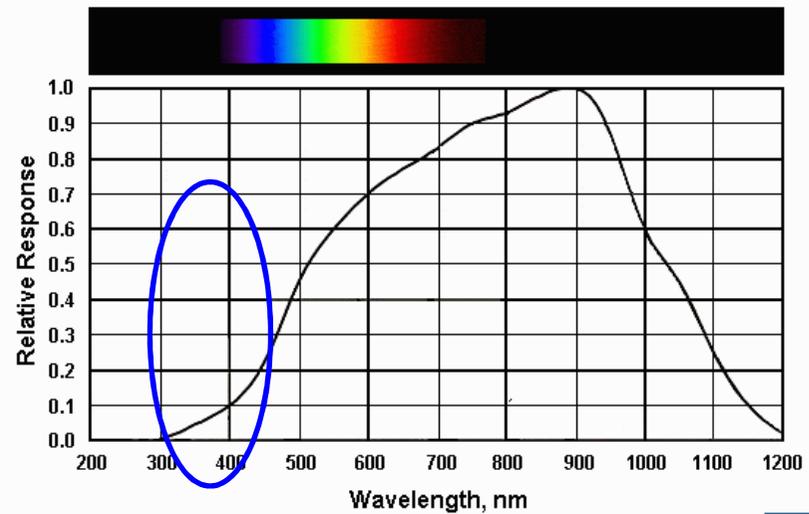
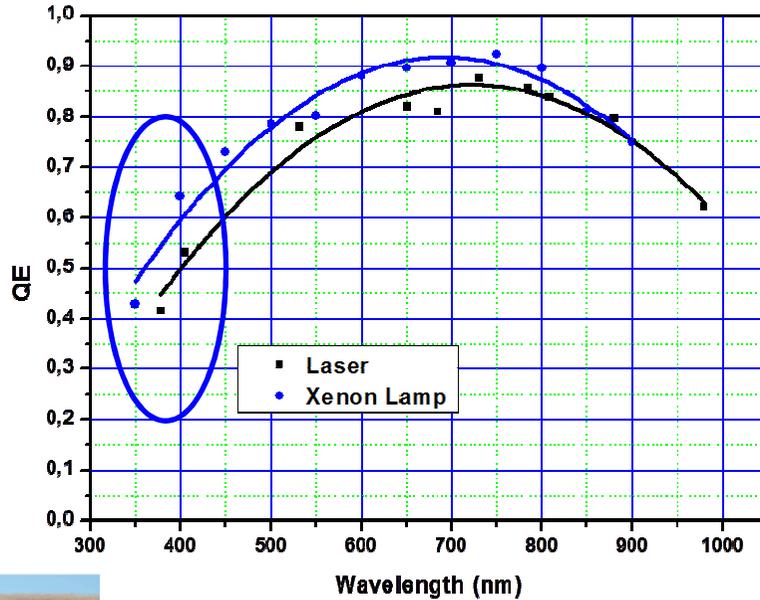
Building nano - detecting micro

PARIDE

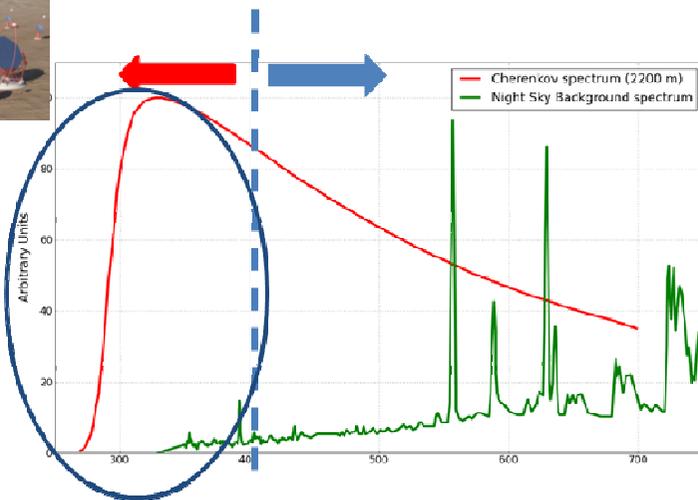


Thank you for attention

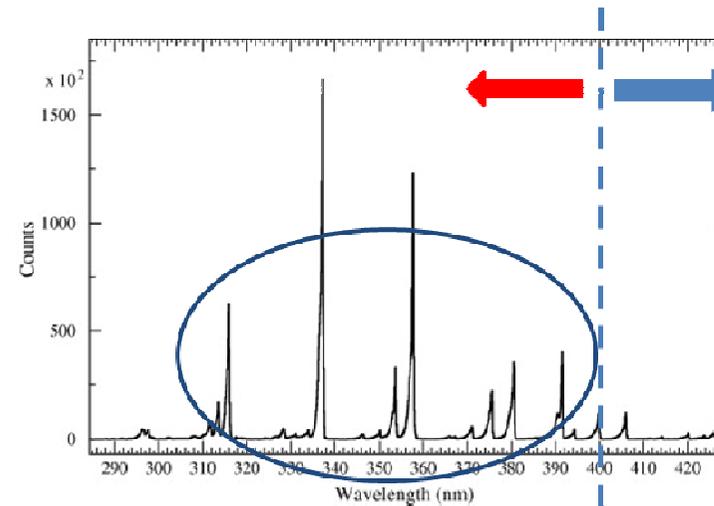
Why a MWCNT-Si photodetector?



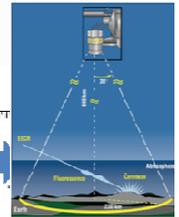
Typical Silicon Photodiode Spectral Response



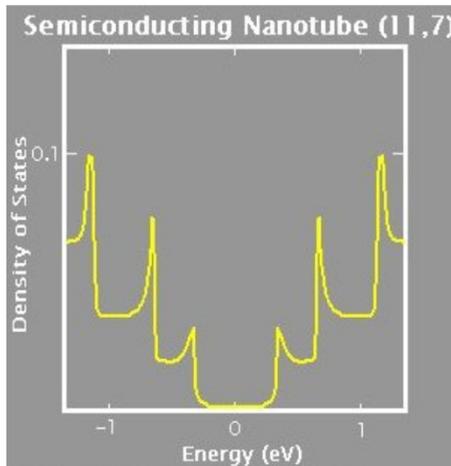
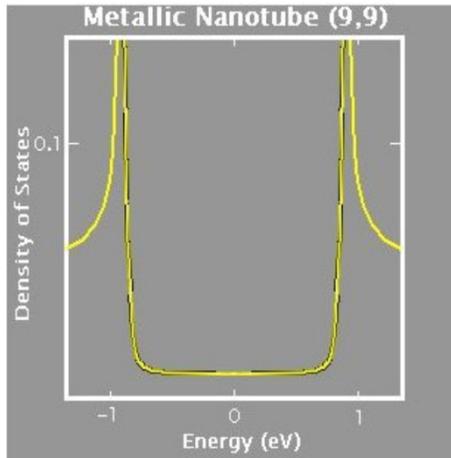
Cherenkov radiation



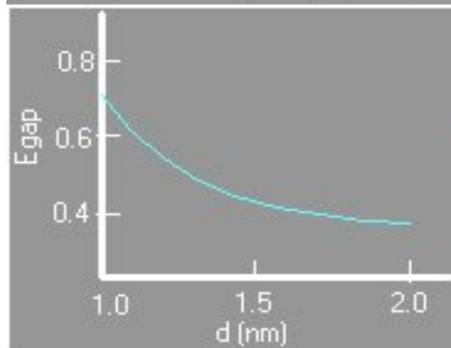
Fluorescence radiation



CNT energetic levels



Above: The Van Hove singularities in metallic and semiconducting nanotubes. Adapted from [19].



Semiconductors nanotubes show interesting fluorescence properties in the region of close infrared (from ~ 1 to $\sim 15 \mu\text{m}$) tied to their electronic characteristics. Nanotubes of type $n-m=3p$ with p entire positive or null are metallic conductors.

All the others are semiconductors whose gap is function of the diameter, and are approximated from the function:

$$E_{\text{gap}} = 2 y_0 \text{acc}/d$$

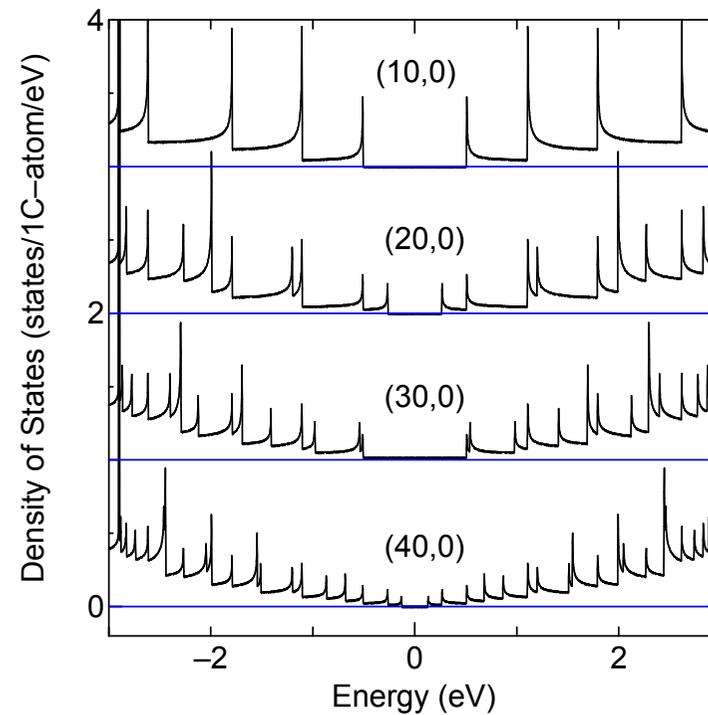
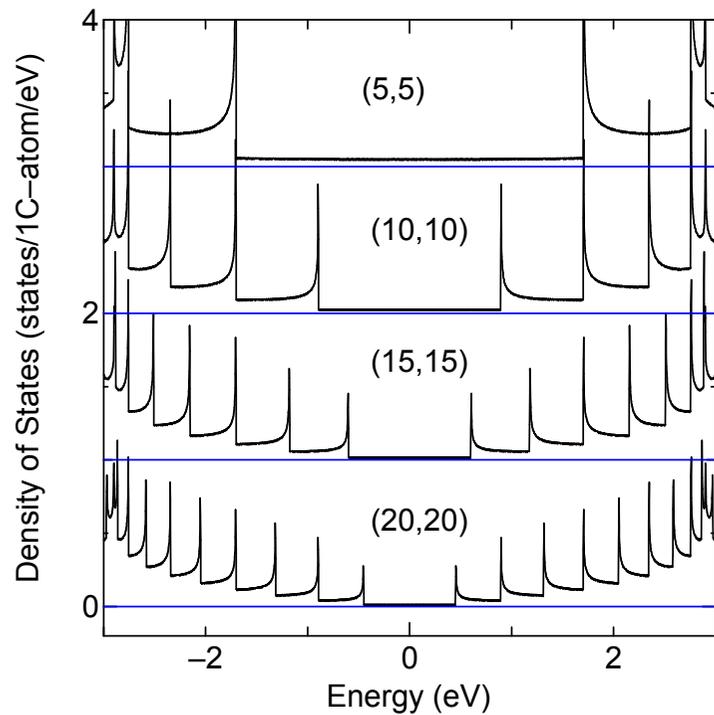
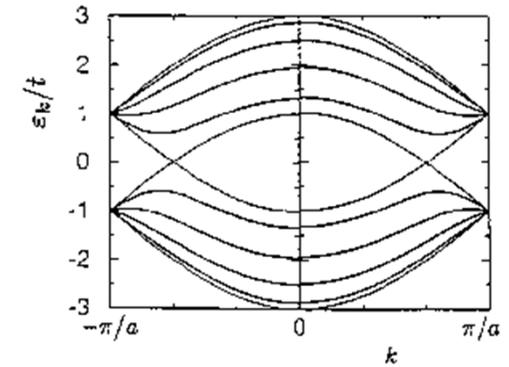
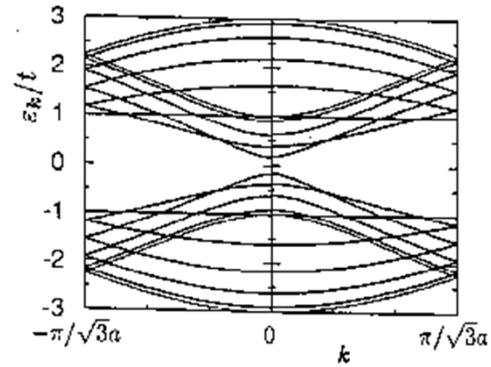
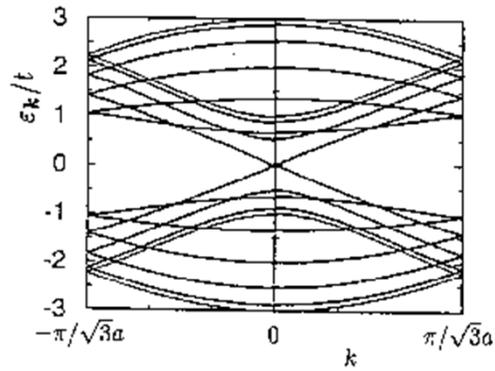
where $y_0=0.1$ eV, $\text{acc}=0.142$ nm and d is the diameter. This implies that for the Single Wall CNT the fundamental gap varies from 0.4 to 0.7 eV.

Multi Wall CNT instead present a wider range of energy gap.

Metallic Zig-zag

Semiconducting Zig-zag

Armchair



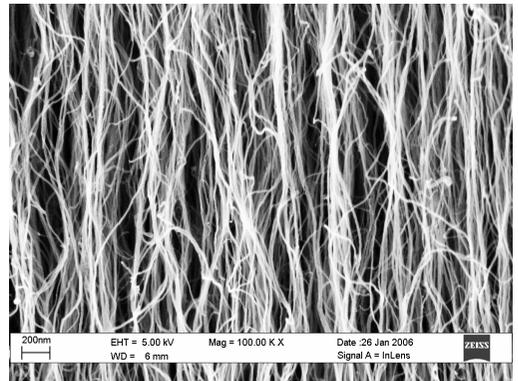
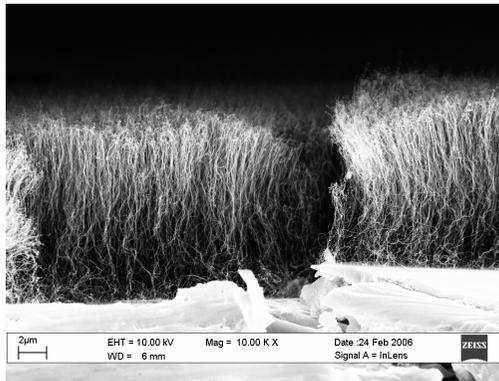
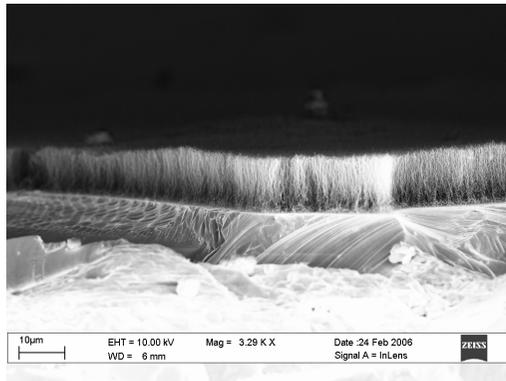
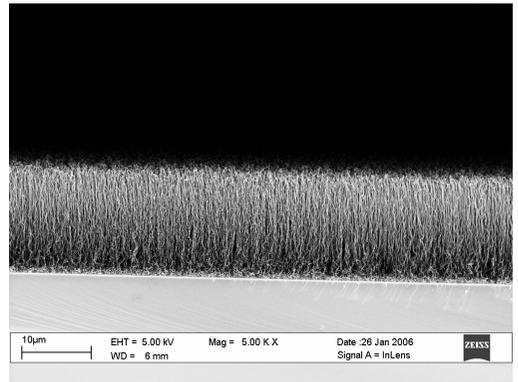
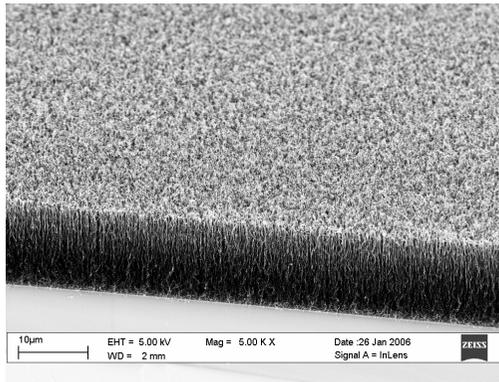
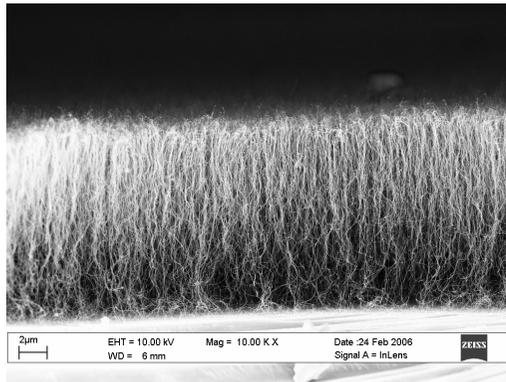
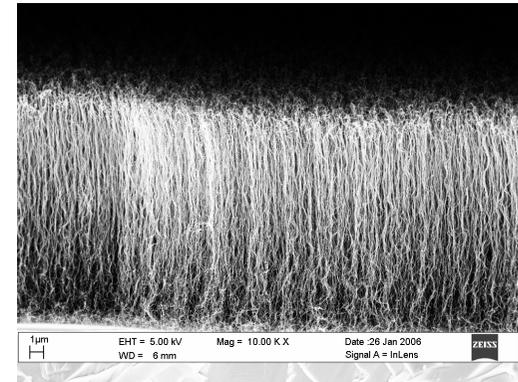
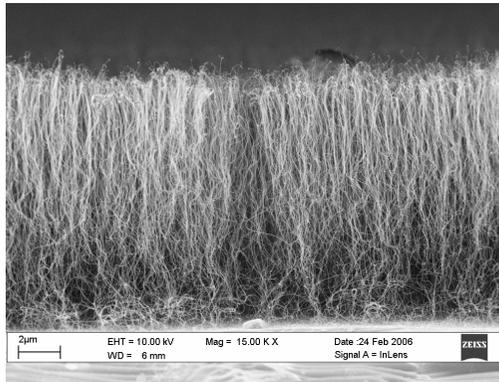
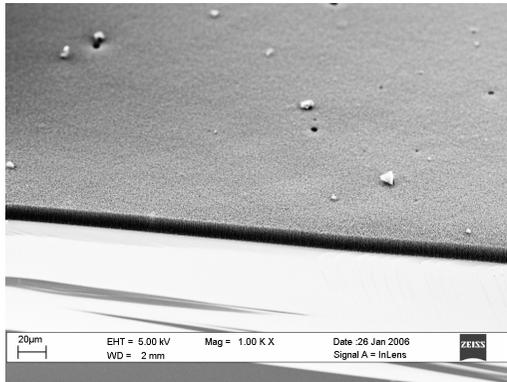
$$c \cdot K = 2\pi m$$

$$m = 1 \dots q$$

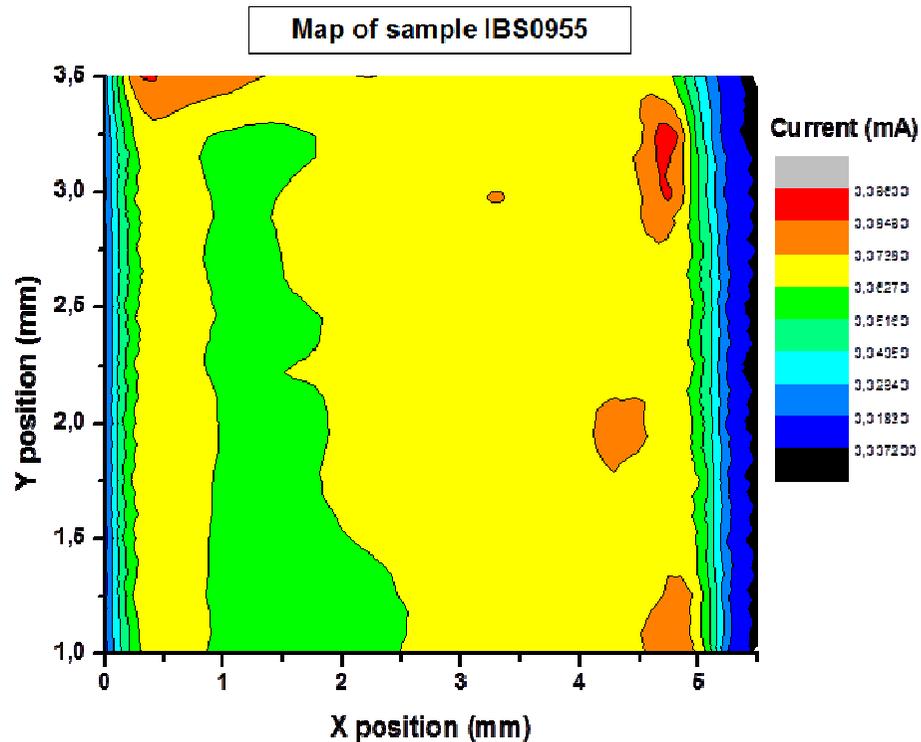
A multiwall carbon nanotube typically consists of a concentric set of nanotubes of both metallic and semiconducting types

R. Saito, G. Dresselhaus and M.S. Dresselhaus, *Physical Properties of Carbon Nanotubes*, Imperial College Press (2003)

SEM Images



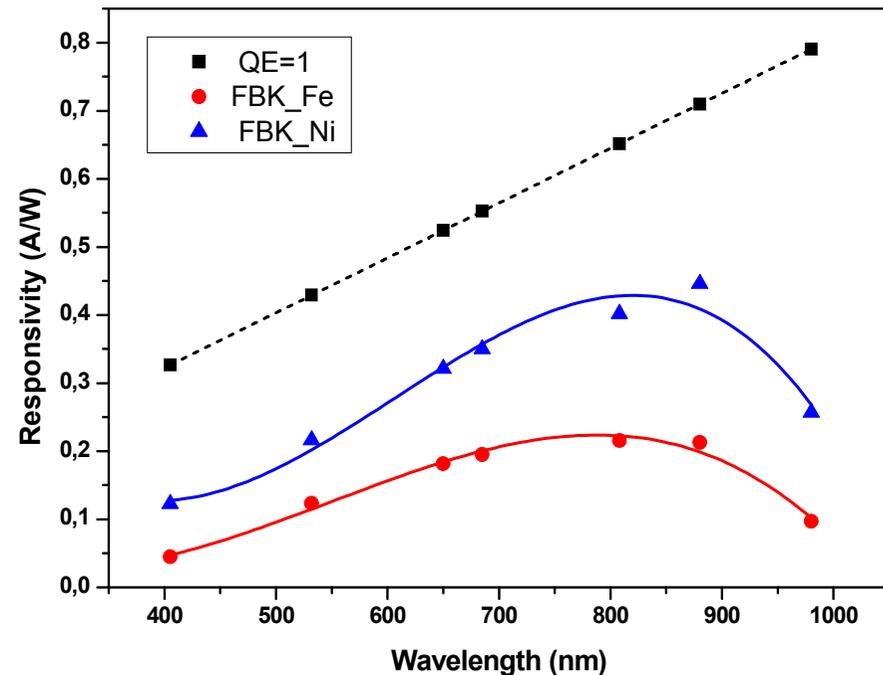
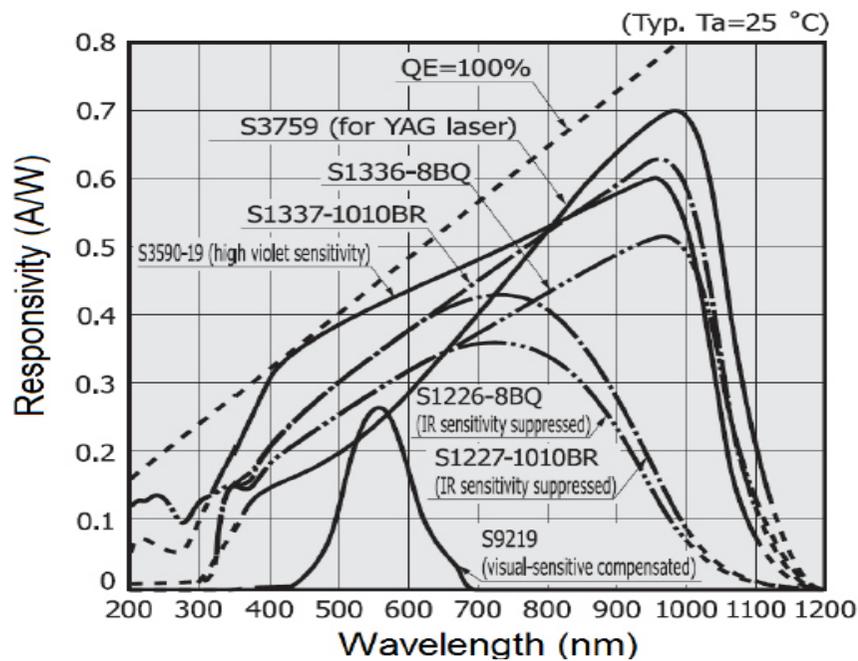
Great uniformity over large area



The electric field applied uniformly over the entire CNT surface plays a fundamental rule making uniform the charge electrodes.

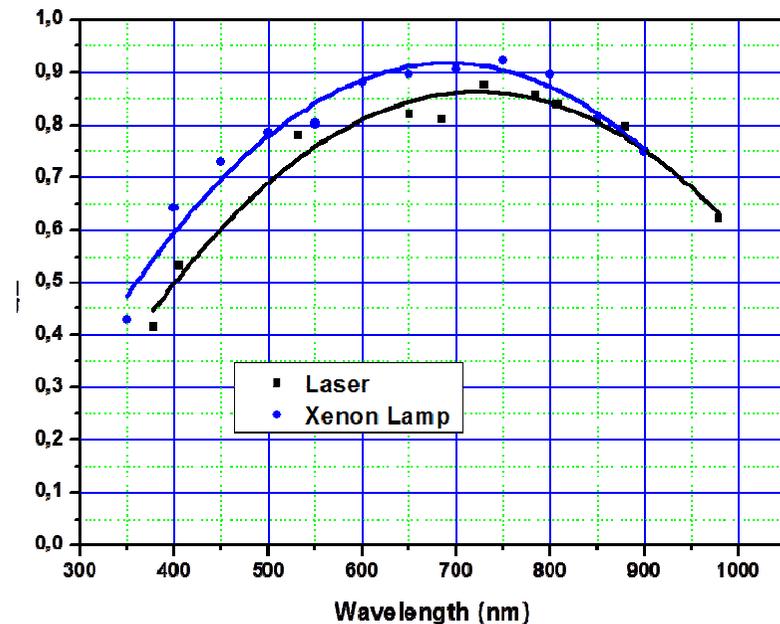
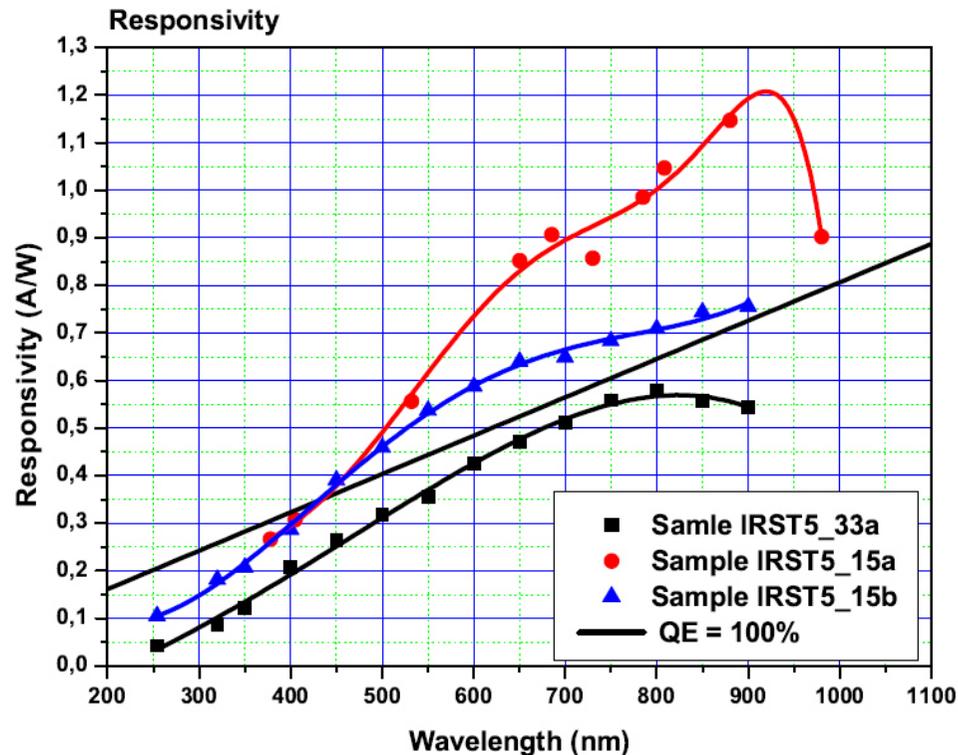
The signal generated everywhere in the sample can be collected to the metallic electrodes through the ITO layer, whose resistivity is very low.

Si-CNT photodetector responsivity



Responsivity comparison between Si-CNT prototype and commercial photodiode

Si-CNT photoresponsivity



A particular layered substrate without the schottky junction on the back shows very peculiar characteristics: high quantum efficiency in all the wide wavelength range, a hetero-junction threshold at 2.5 V and a well defined tunnel effect under this voltage. Work on this substrate is still in progress.