

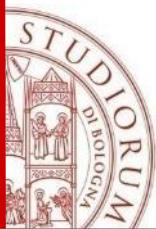
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# Innovative Large Area Flexible Ionizing Radiation Detectors

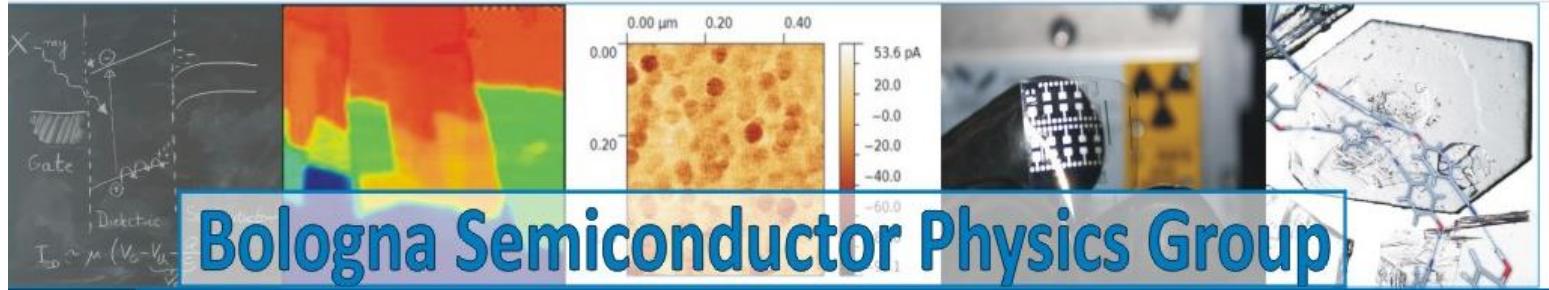
Laura Basiricò

*31<sup>st</sup> January 2020*

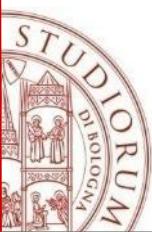
[laura.basirico2@unibo.it](mailto:laura.basirico2@unibo.it)



# Our Group @ DIFA

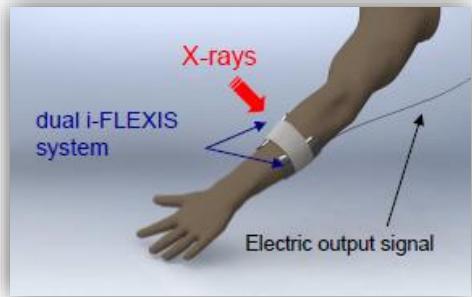


BOLOGNA



# Motivation: Flexible large area X-rays detectors

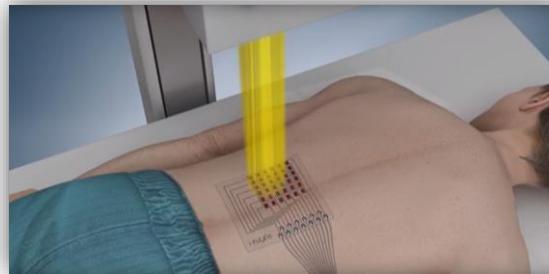
Wearable health  
diagnostic applications



Citizens security:  
“smart walls/pillars”



Personal dosimetry  
Medical application



Cultural Heritage



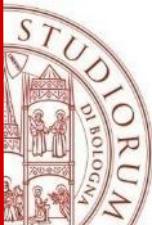
New generation of  
**low cost, low power supply**  
and **mechanical flexibility**  
**Thin and conformable**  
sensor panels and patches

Airport security



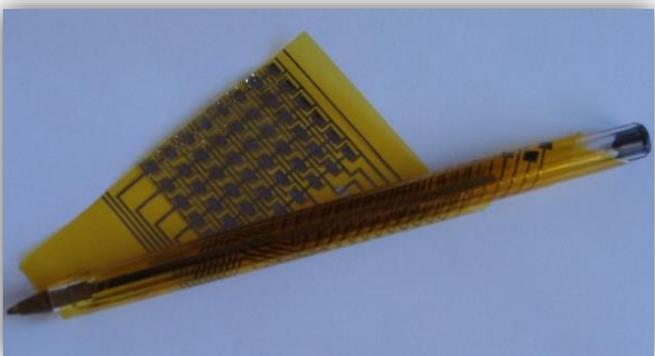
Defense





# Why Organic Materials?

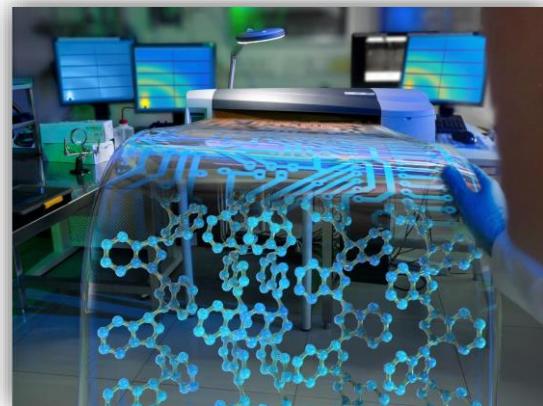
Light-weight and flexible



Wearable



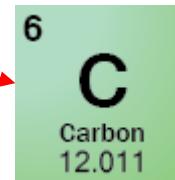
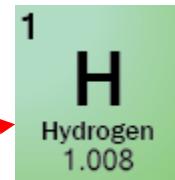
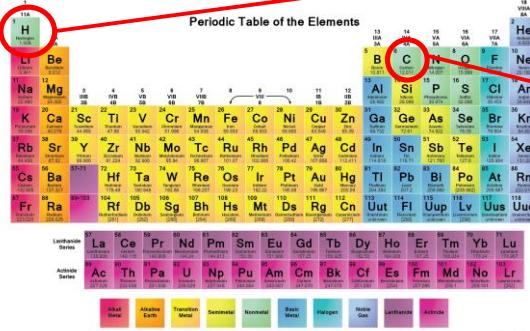
Low-cost printing techniques



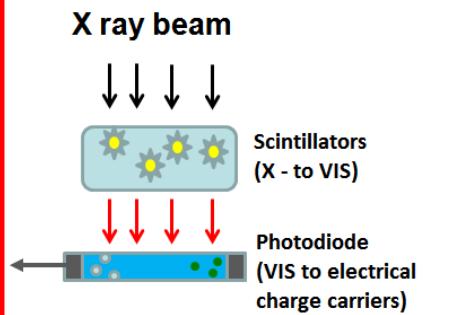
Easy chemical tailoring



Low-Z → tissue-like

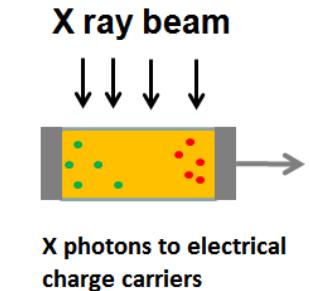


## INDIRECT DETECTION

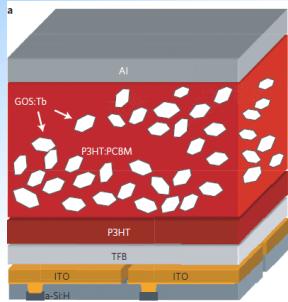


# Organic X-Ray detectors

## DIRECT DETECTION

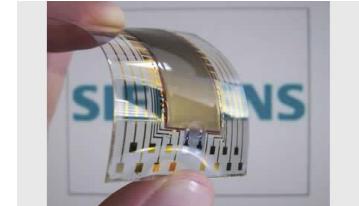


P. Buchele, *Nature Photonics*, 9 (2015)



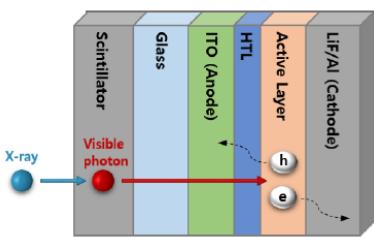
Sensitivity 10.000 nC/mGy/cm<sup>3</sup> @10V

GOS:Tb scintillator particles in organic polymer blend (P3HT:PCBM)



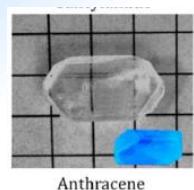
[http://www.siemens.com/innovation/en/news/2013/e\\_inno\\_1305\\_1.htm](http://www.siemens.com/innovation/en/news/2013/e_inno_1305_1.htm)

P3HT:ICBA  
CsI(Tl) scintillator film



Seon et al. DOI  
10.1109/TNS.2016.2645228,  
*IEEE Trans. Nuc. Sci.* (2017)

Hull et al., *IEEE Trans. on Nucl. Sci.* 56, 3, (2009)

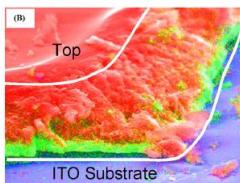


## Organic single crystals

B. Fraboni et al., *Adv. Mater.*, 24, 17, 2289-2293 (2012)



A. Ciavatti et al. *Adv. Mater. Adv. Mater.* 2015, 27, 7213–7220



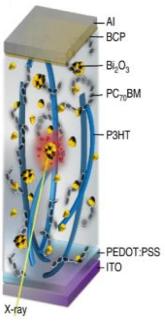
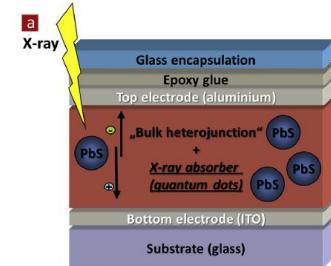
## PTAA/Bi<sub>2</sub>O<sub>3</sub> NPs

Intaniwet, et al., *Journ. App. Phys.*, 106, (2009)  
Intaniwet et al., *Org. Electr.* 12 (2011)

Sensitivity up to 480 nC/mGy/cm<sup>3</sup>  
@50-100V

## P3HT:PCBM:PbS QDs BHJ

Ankah et al. *Organic Electronics* 33 (2016)

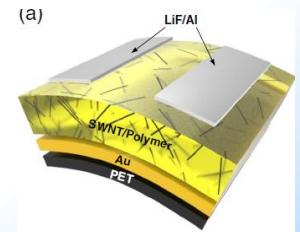


## P3HT:PCBM/Bi<sub>2</sub>O<sub>3</sub> NPs

Thirimanne, et al., *Nat. Comm.* 9, (2018)

## CNT

H. Han et al, *Nanoscale Research Letters*, 9 (2014)

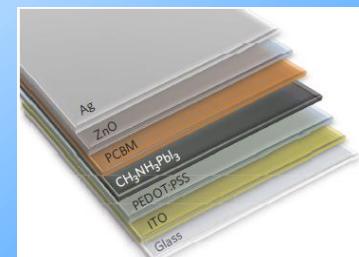


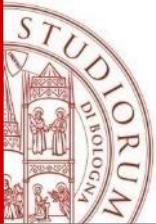
Sensitivity 10.000 nC/mGy/cm<sup>3</sup> @30V

## Solution-processed lead halide perovskites

S. Yakunin et al, *Nature Photonics*, 9 (2015)

Basiricò et.al. *Adv. Func. Mat.* 2019





# The FIRE Project (CSN5 call 2019)



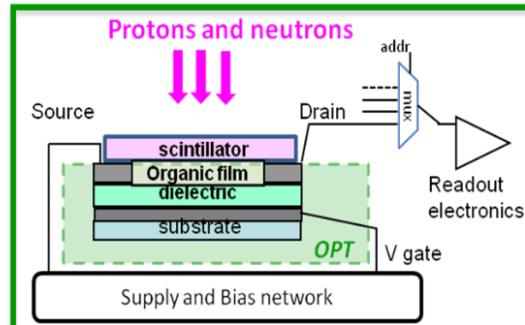
# FIRE

Flexible organic Ionizing  
Radiation dEtectors

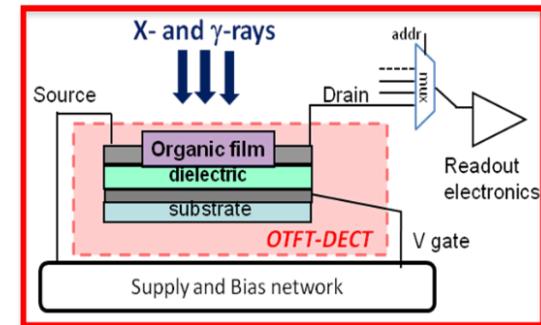
- **Coordination:** Bologna (B.Fraboni)
  - **5 Research Units:**
- TIFPA, LNL, Bologna, RomaTre, Napoli
- **Time schedule:** 2019-2021

## FIRE at a glance

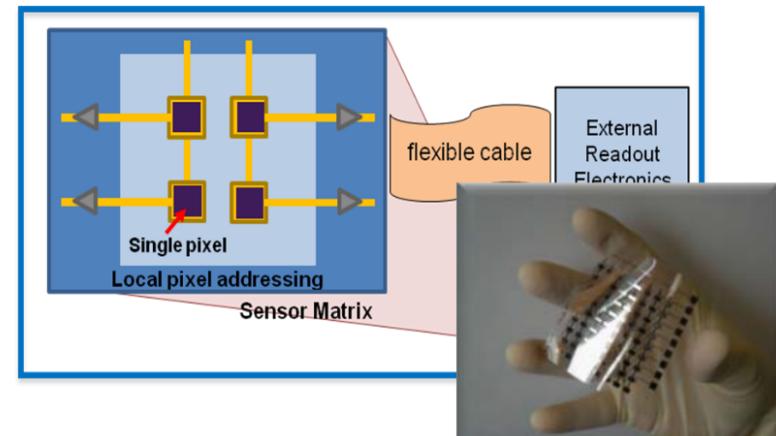
INDIRECT DETECTING SINGLE PIXEL (NEPRO)

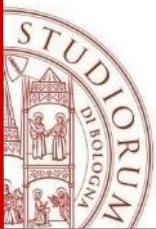


DIRECT DETECTING SINGLE PIXEL (PHOX)



FULLY INTEGRATED FLEXIBLE DETECTING SYSTEM

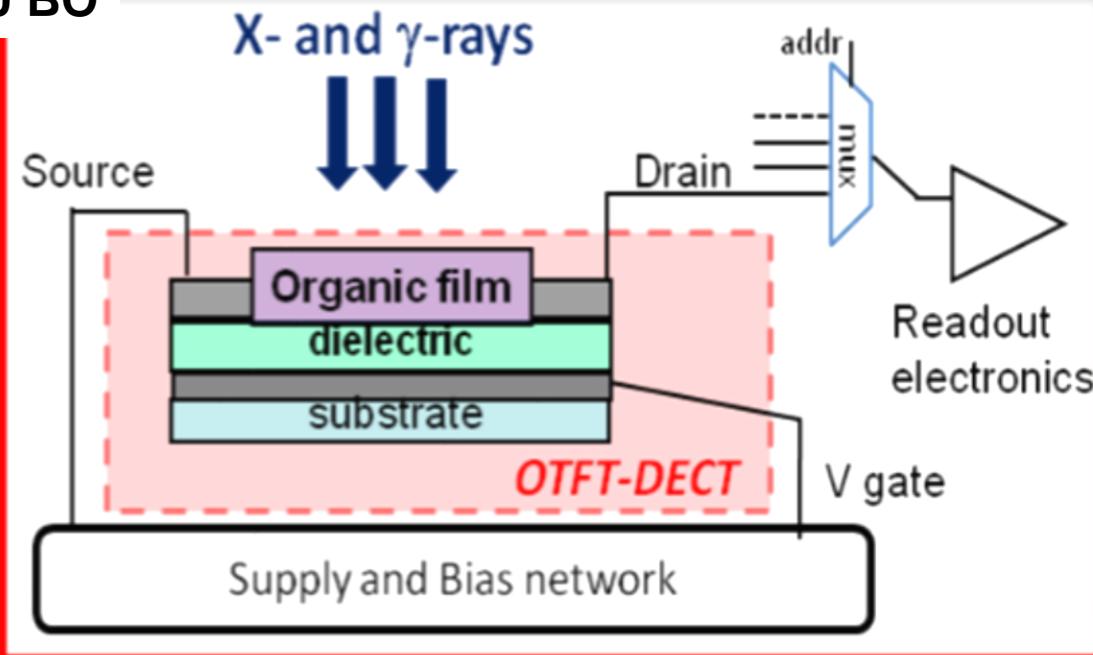




# FIRE at a glance - FINAL TARGETS

RU BO

## DIRECT DETECTING SINGLE PIXEL (PHOX)



DEMO2

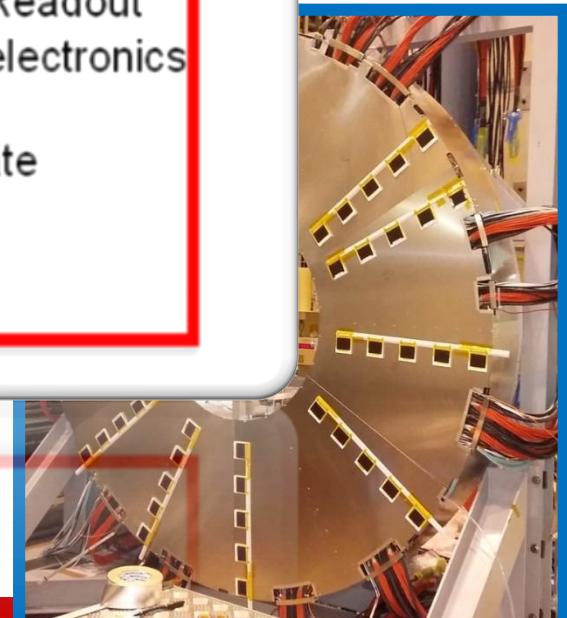
(Electron)

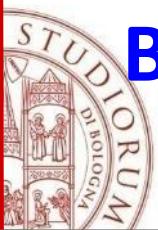
- HIGH  
low d

for proton

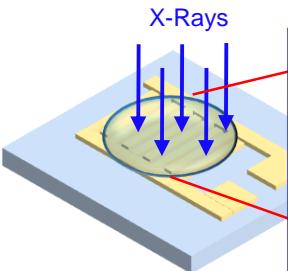
treatment and

ability, human

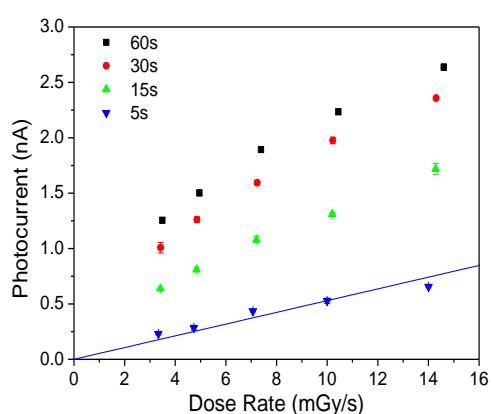
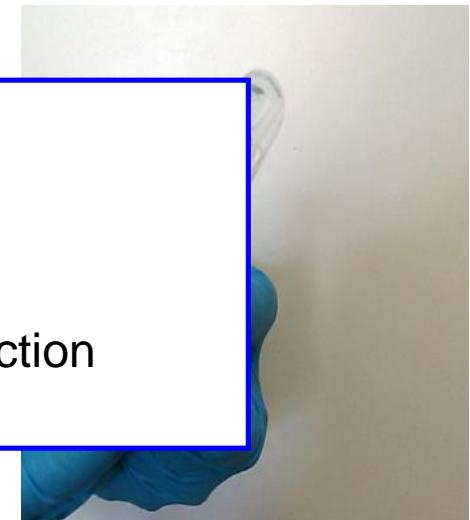




# Background: Flexible direct X-ray detectors based on Organic thin films



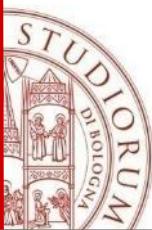
- ✓ Very high Sensitivity
- ✓ Flexible (100nm thin film)
- ✓ Very low voltage (0.2V)
- ✗ Extremely low Q.E. (0.0015% attenuated fraction  
150nm thick film)



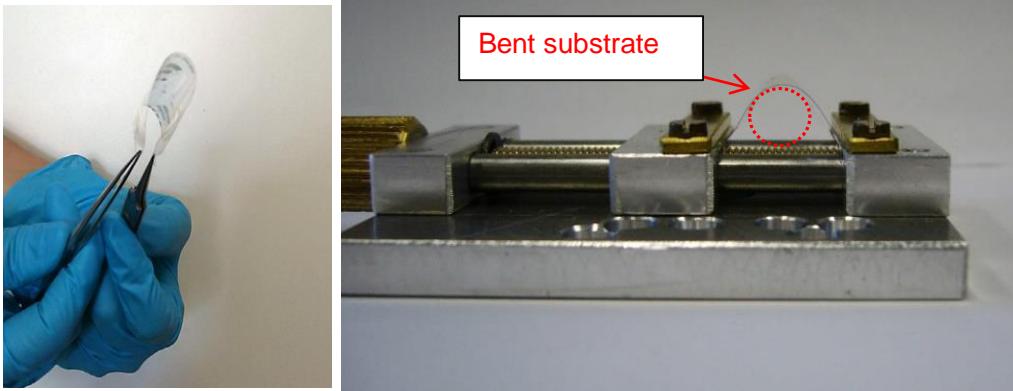
$$S = \frac{\partial I}{\partial D}$$

**Sensitivity: 180 nC/Gy (72000 nC/mGy cm<sup>3</sup>) @0.2V**

**Room temperature and real-time operation**



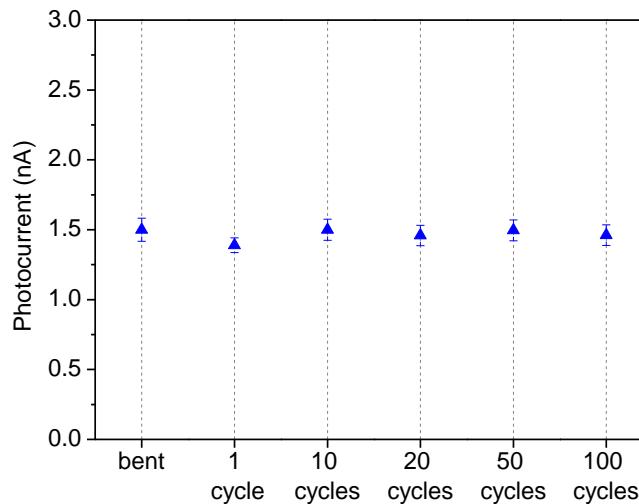
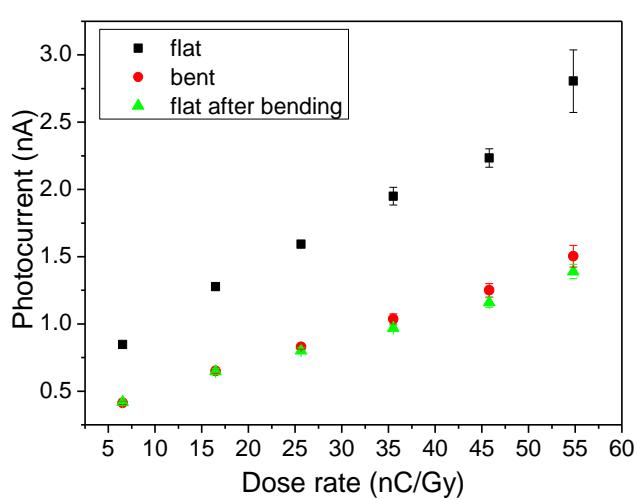
# Bendability and Mechanical flexibility



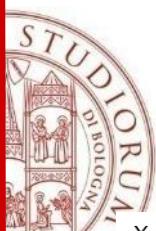
**Assessment of the mechanical reliability of the system.**

Experimental set-up used for the characterization of the detector during bending.

**bending radius of 0.3 cm**  
→ Conformable to human body



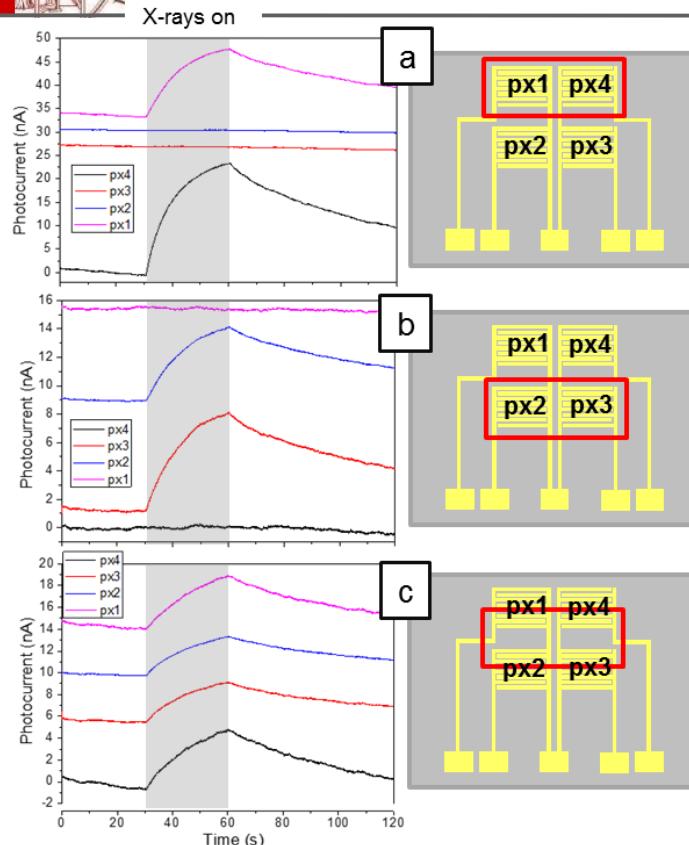
- Decrease of about 50% after first bending
- Stability over 100 bending cycles.



Pixel size 5 mm

# 2x2 pixel matrix

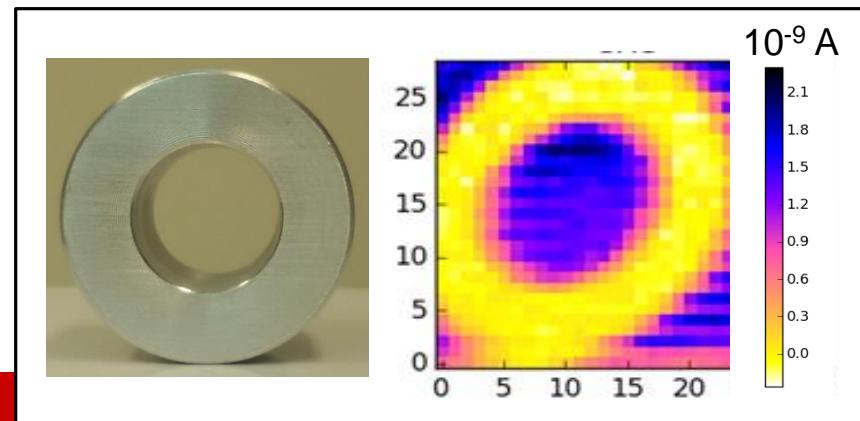
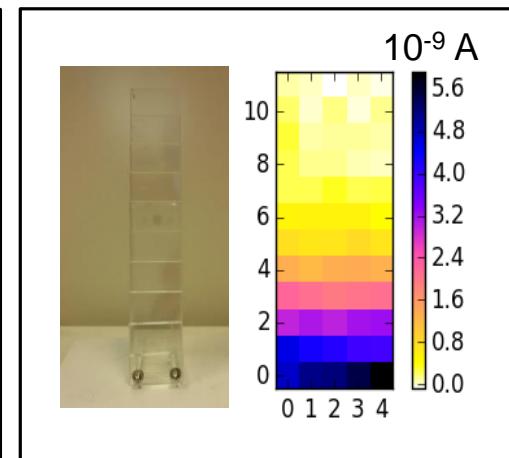
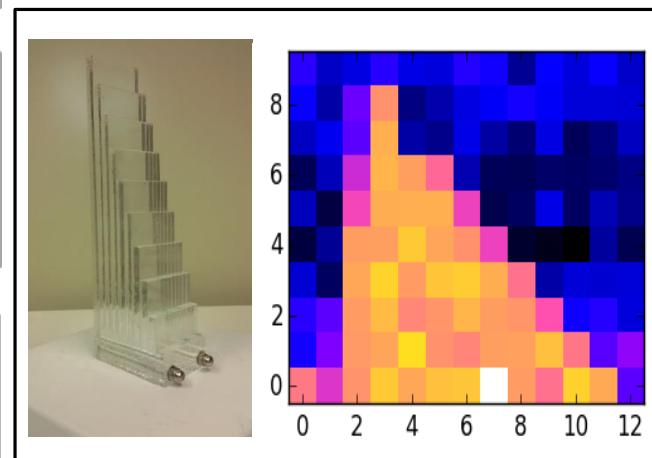
Ultra-low  
voltage: 0.2V

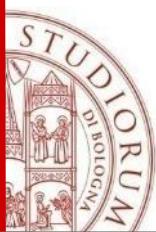


## pixel matrix organic detector

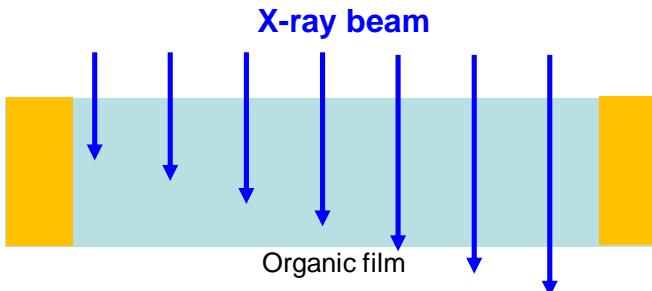
- a) only pixels 1 and 4 are irradiated
- b) only pixels 2 and 3 are irradiated
- c) all the pixels are irradiated.

Radiation source:  
monochromatic synchrotron  
X-ray beam at 17 keV with  
a dose rate of 28.5 mGy/s.





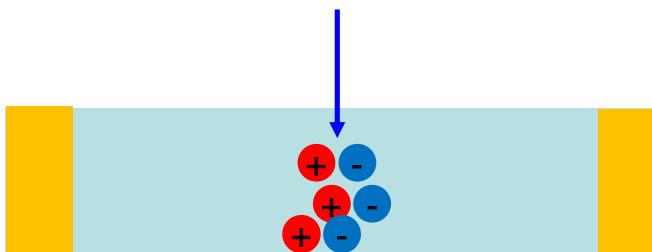
# Charge Collection Model



## X-ray photocurrent

$$I_{CC} = \Phi n q$$

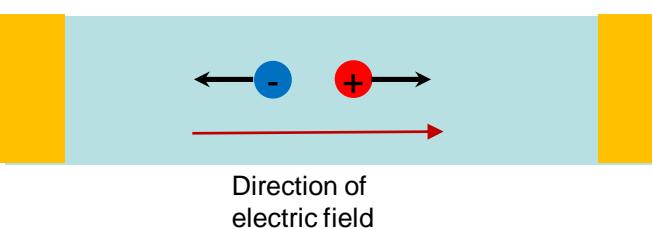
$q$  - is the elementary charge  
 $\Phi$  - is the photon absorption rate  
 $n$  - is the number of the generated electron-hole pairs per absorbed photon.



## Absorbed photon flux

$$\Phi = \Phi_0 [1 - \exp(-h/l)]$$

$E_{ph}$  is the photon energy  
 $E_g \approx 2 \text{ eV}$  the energy gap of TIPS-pentacene.  
 $A = 0.015 \text{ cm}^2$  - active area  
 $h = 100 \text{ nm}$  – thickness  
 $\lambda = 0.68 \text{ cm}$  - attenuation length  
absorption = 0.0015 %

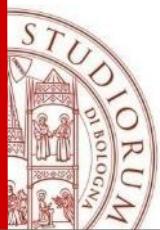


## # photogenerated carriers/photons

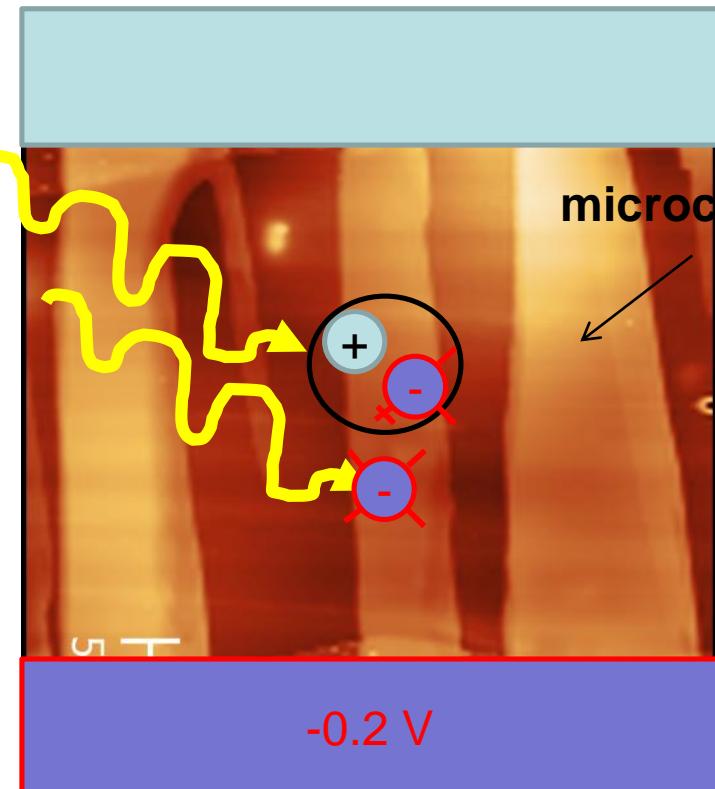
$$n = E_{ph} / E_g$$



$$I_{CC,max} < 2 \text{ pA}$$



# Why is it working? charge traps and photoconductive gain



- trapping of n-type carriers
- injecting contacts

L. Basiricò et al., *Nat. Commun.* 7, 2016.

## under X-ray irradiation:

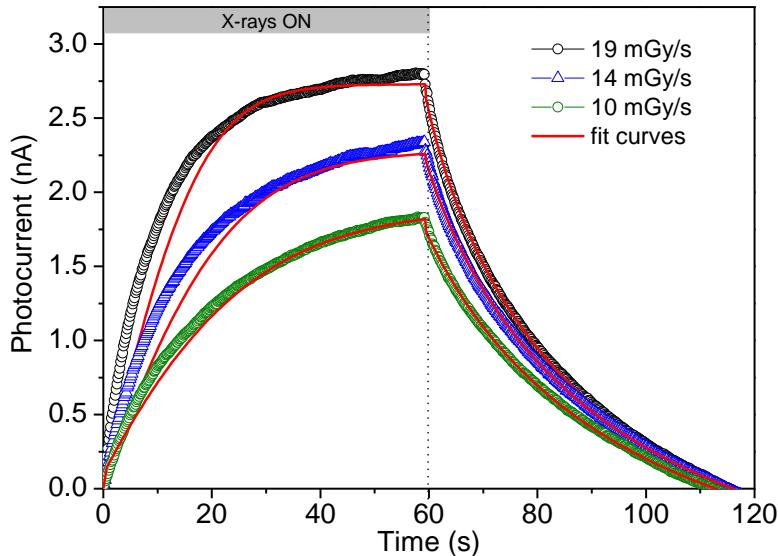
- 1) Additional electrons and holes are generated.
- 2) **Holes drift** along the electric field and reach the collecting electrode while **electrons remain trapped** in deep trap states and act as “doping centers”.
- 3) To guarantee charge neutrality, **holes are continuously emitted** from the injecting electrode. As a consequence, for each electron-hole pair created, more than one hole contributes to the photocurrent.
- 4) **Recombination process takes place**, counterbalancing the charge photogeneration in the steady-state.



$$\Delta I_{PG} = G I_{CC}$$

**G = photoconductive gain  $\approx 10^4$**

# Fitting with experimental data



**Dynamics of X-ray response and consequences on detector operation.** a) Experimental and simulated curves of the dynamic response of the detector for three different dose rates of the radiation. The experimental data refer to 60 s of exposure of the device ( $W = 48$  mm,  $L = 30$   $\mu\text{m}$ , bias 0.2 V) to a synchrotron 17 keV X-ray beam, with a bias of 0.2 V.

The model well reproduce the saw-tooth shape of three experimental set of data, using a single set of fitting parameters

$$G = \frac{\tau_r}{\tau_t} = \frac{29.4}{1.1 \times 10^{-3}} = 2.6 \times 10^4$$

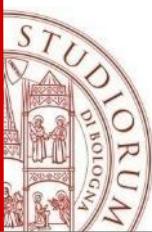
**Carriers lifetime**

$$\tau_r = \frac{\alpha}{\gamma} \left[ \alpha \ln \left( \frac{\rho_0}{\rho_x} \right) \right]^{\frac{1-\gamma}{\gamma}} = 29.4 \text{ s}$$

**Transit time**

$$\tau_t = \frac{L^2}{V\mu} = 1.1 \text{ ms}$$

*L. Basiricò et al.*  
Nature Commun. 7, 13063 (2016).



# Improving sensitivity

$$\Delta I_{PG} = G I_{CC}$$

Photoconductive gain

$$G = \frac{\tau_r (\rho_x)}{\tau_t}$$

→ Carrier lifetime

→ Transit time

$$\tau_t = \frac{L^2}{V\mu}$$



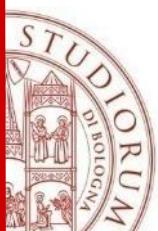
Increasing **charge carriers density (OTFTs)**



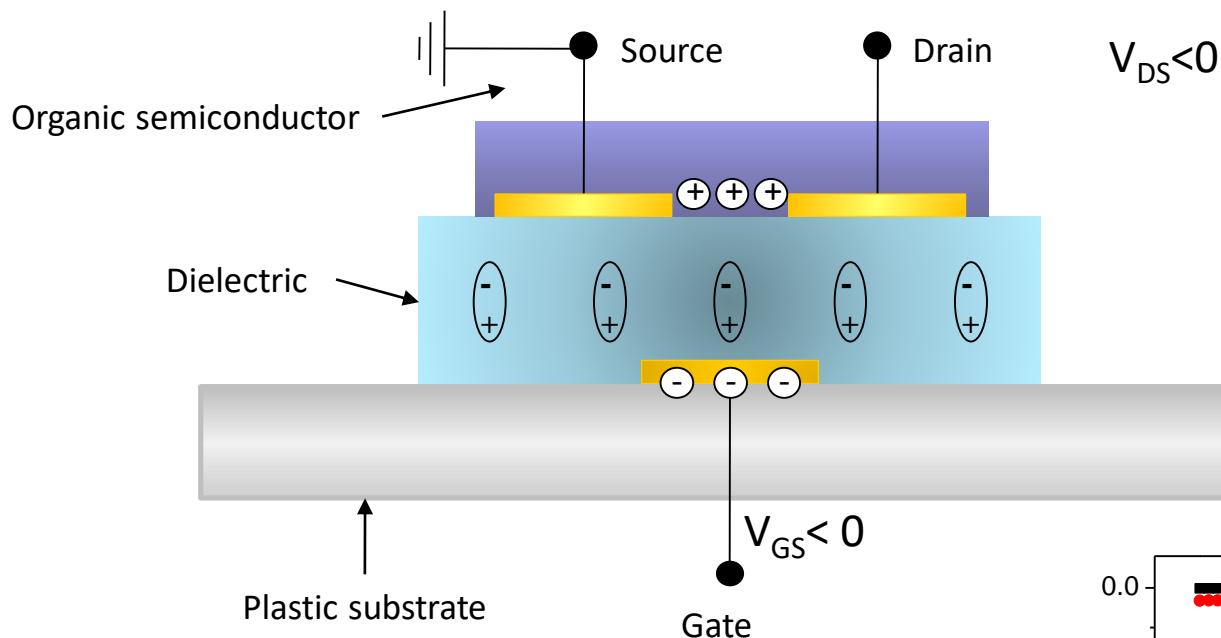
Increasing **attenuated fraction**



Increasing **mobility**

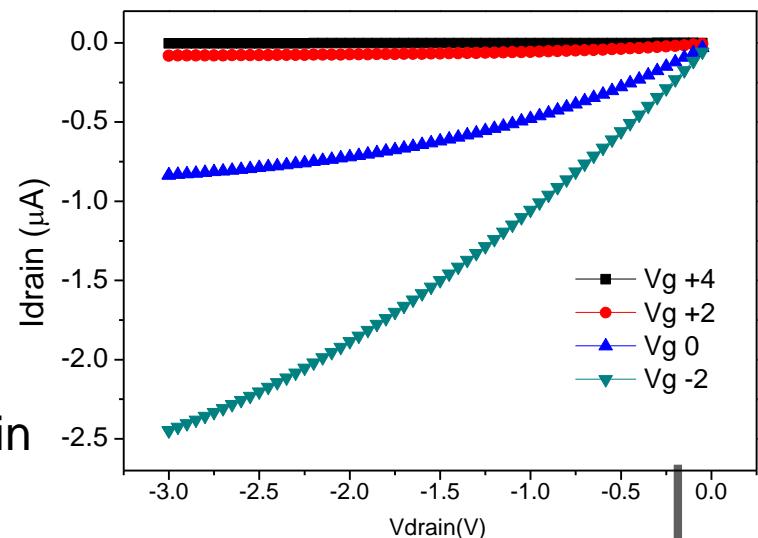


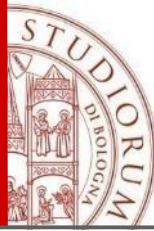
# Low Voltage Organic Field Effect Transistors as X-ray detectors



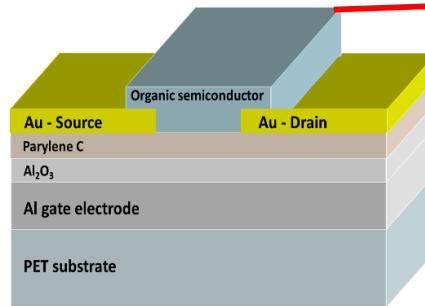
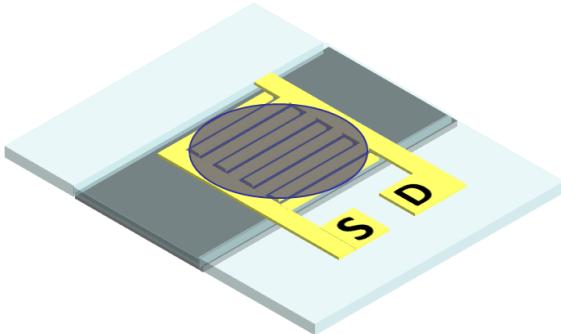
OTFTs  
working  
principle

- it works in **accumulation regime** “p-type” semiconductors
- $V_{GS} < 0$ : hole accumulation on the interface
- $V_{DS} < 0$ : current flows between source and drain

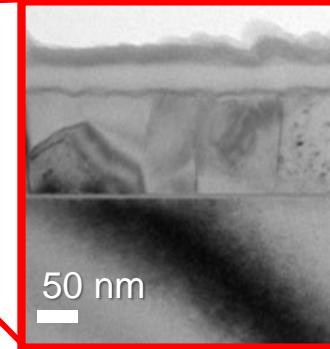




# Towards Scaling-up the system: Low Voltage Organic Field Effect Transistors as X-ray detectors

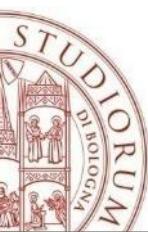


P.Cosseddu et al., AppPhysLett, 100 (2012)

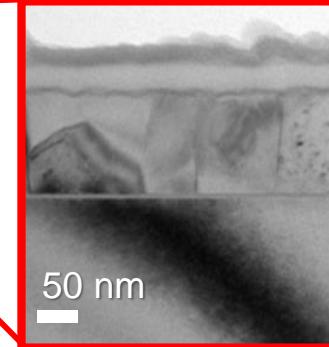
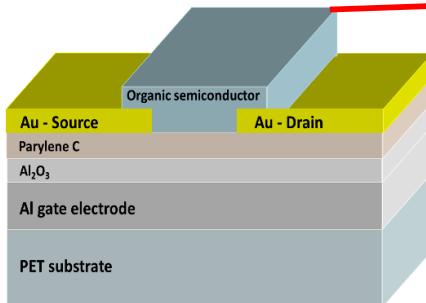
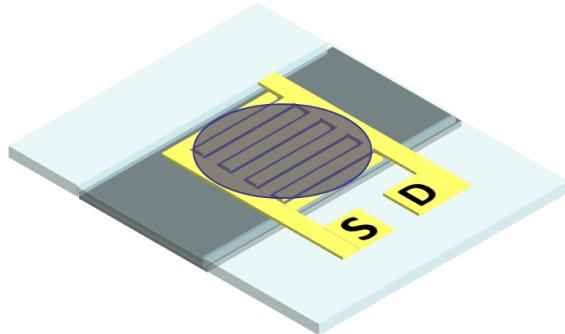


Parylene C  
 $\text{Al}_2\text{O}_3$   
Al

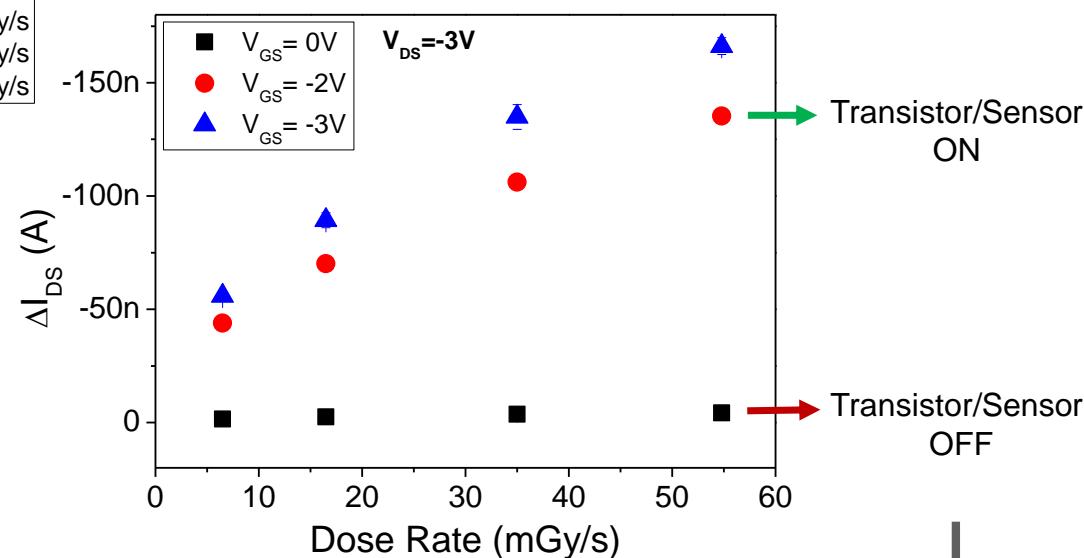
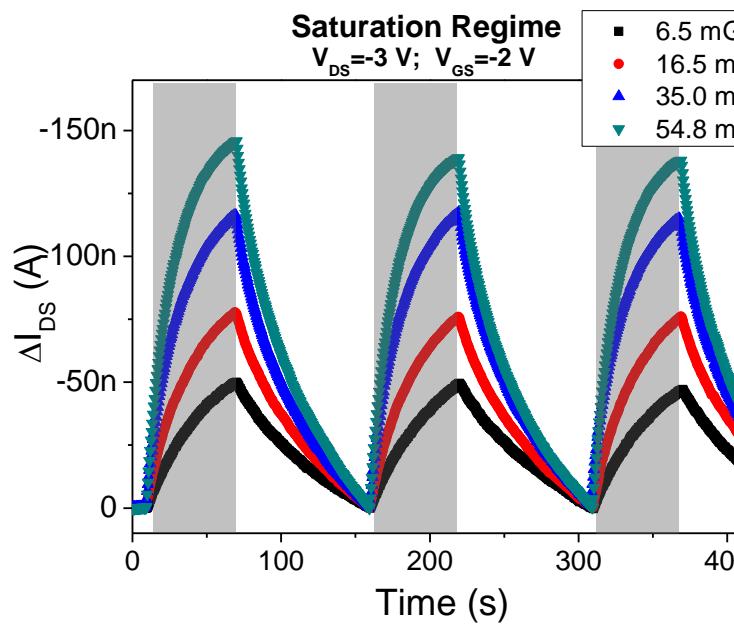
# Towards Scaling-up the system: Low Voltage Organic Field Effect Transistors as X-ray detectors



P.Cosseddu et al., AppPhysLett, 100 (2012)



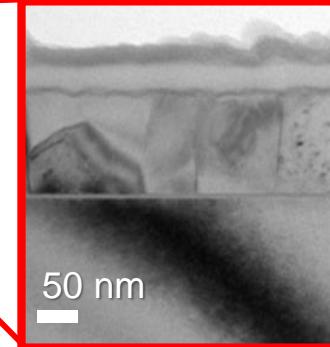
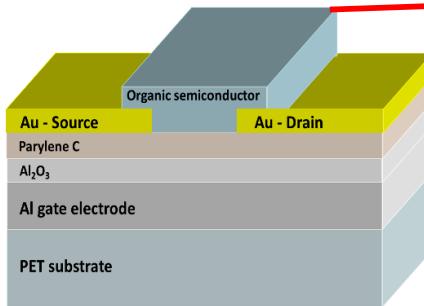
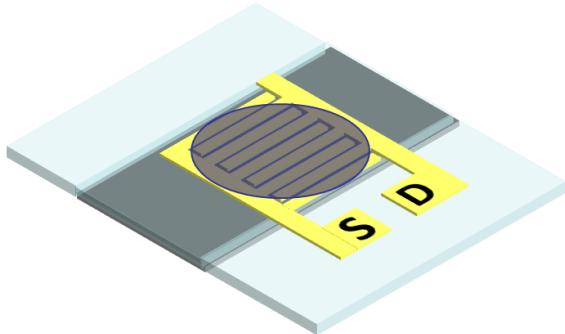
Parylene C  
 $\text{Al}_2\text{O}_3$   
Al



# Towards Scaling-up the system: Low Voltage Organic Field Effect Transistors as X-ray detectors



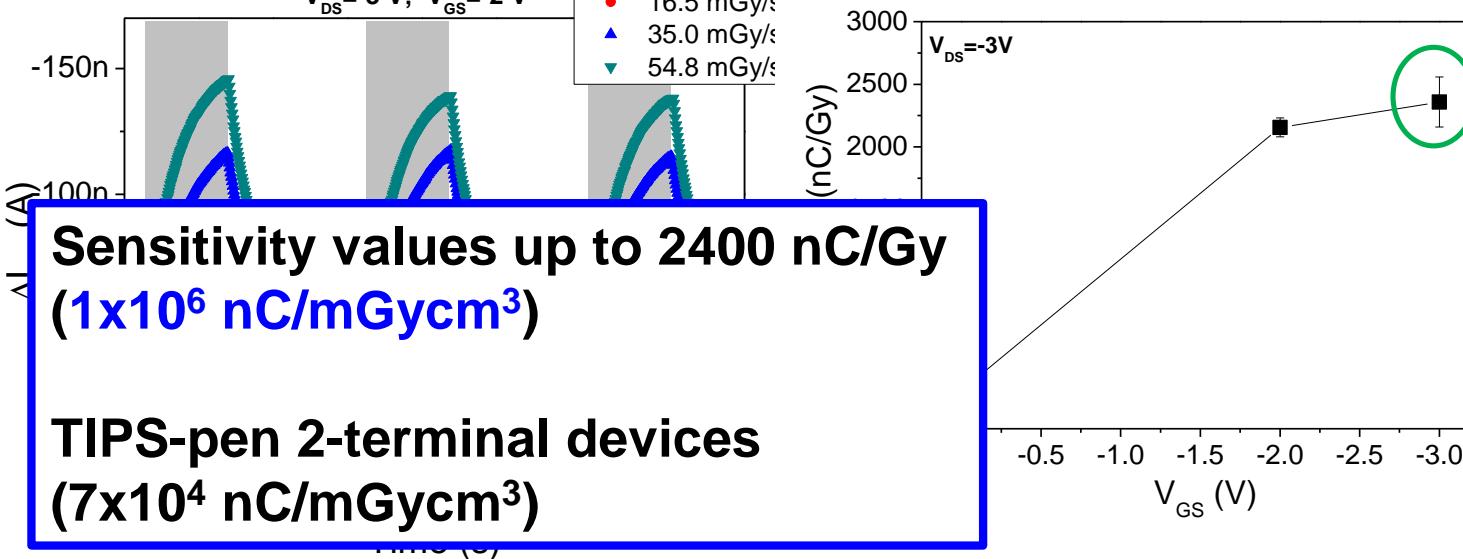
P.Cosseddu et al., AppPhysLett, 100 (2012)



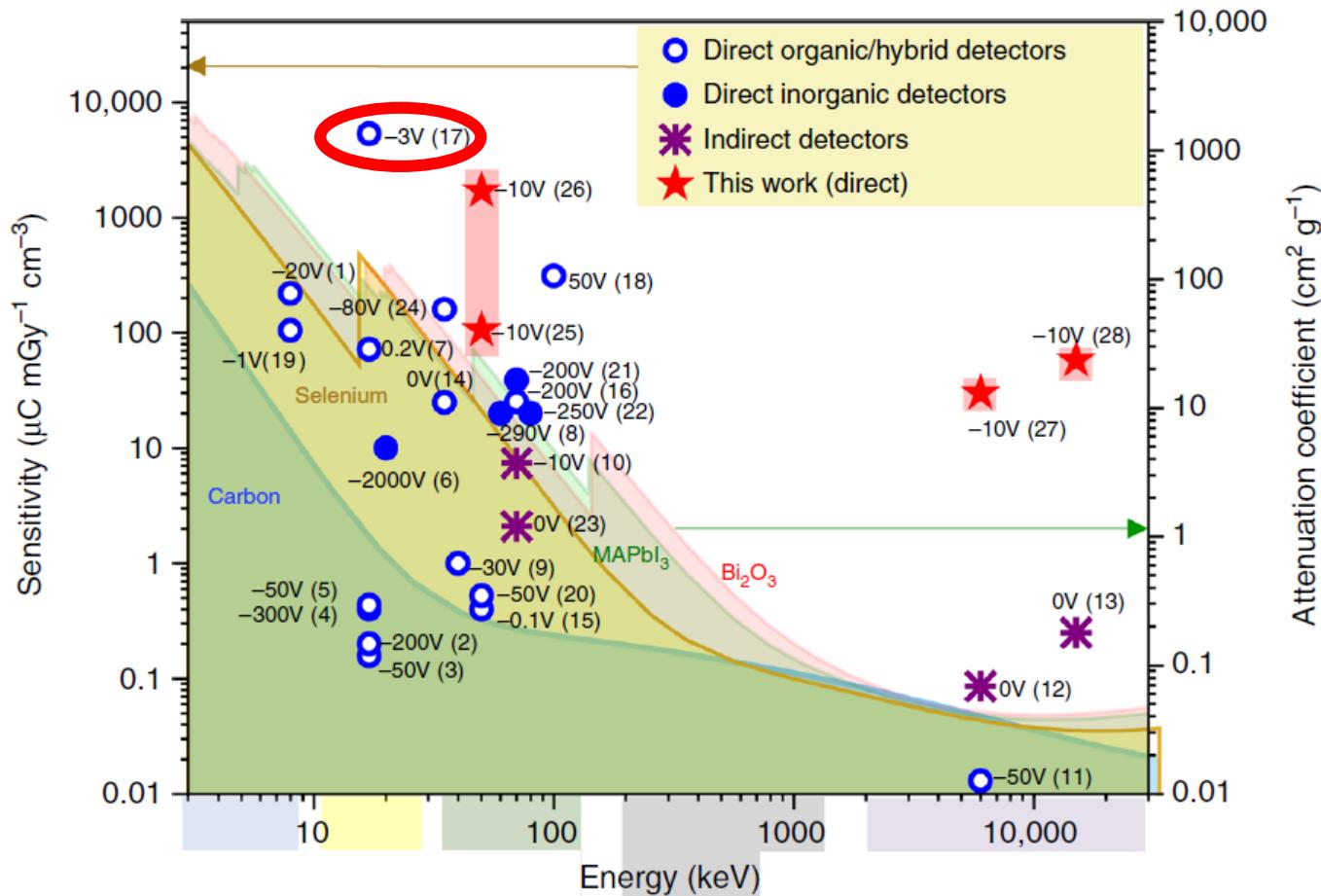
Parylene C  
 $\text{Al}_2\text{O}_3$   
AI

Saturation Regime  
 $V_{DS} = -3 \text{ V}$ ;  $V_{GS} = -2 \text{ V}$

- 6.5 mGy/s
- 16.5 mGy/s
- ▲ 35.0 mGy/s
- ▼ 54.8 mGy/s



# SoA



*Thirimanne et al, Nat Comm 9,2926 (2018)*



# Improving sensitivity

$$\Delta I_{PG} = G I_{CC}$$

Photoconductive gain

$$G = \frac{\tau_r(\rho_x)}{\tau_t}$$

→ Carrier lifetime

→ Transit time

$$\tau_t = \frac{L^2}{V\mu}$$



Increasing **charge carriers density (OTFTs)**

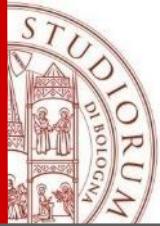


Increasing **attenuated fraction**

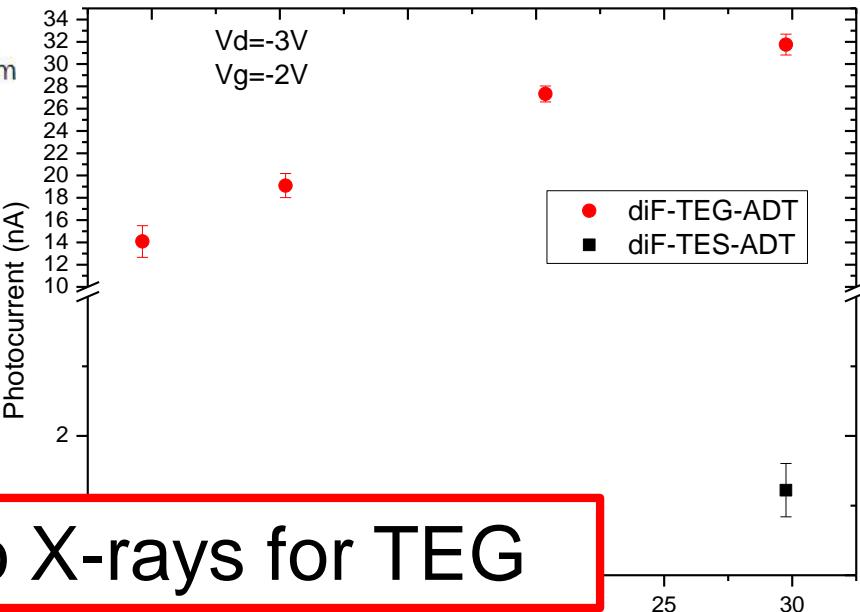
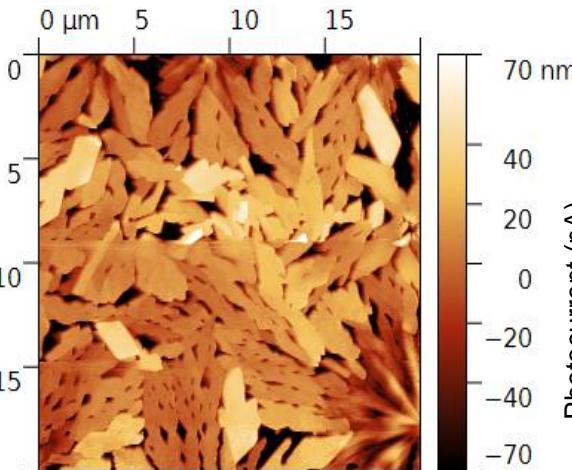
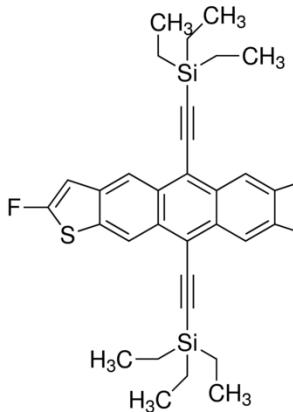


Increasing **mobility**

# Increasing attenuated fraction: higher Z – TES & TEG

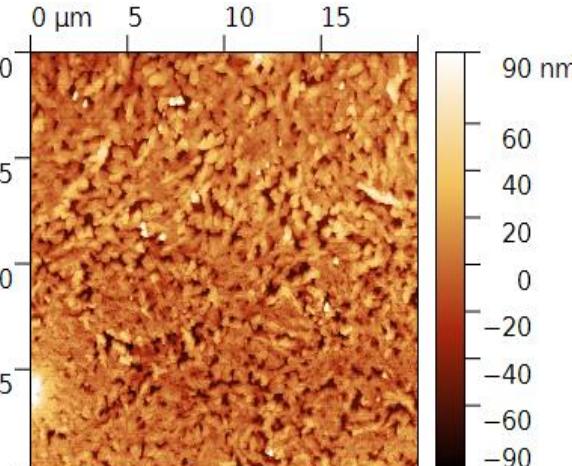
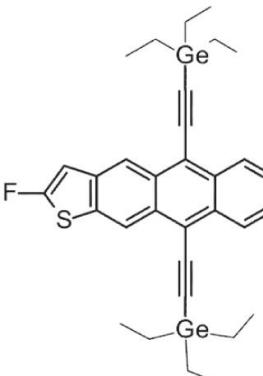


diF-TE $\textcolor{blue}{S}$ -ADT



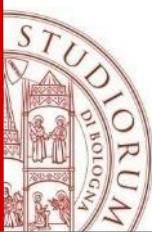
Higher sensitivity to X-rays for TEG

diF-TE $\textcolor{magenta}{G}$ -ADT



2,8-Difluoro-5,11-bis(trimethylgermyl)anthradithiophene

Molecule	Mobility [ $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ ]	Operative voltage	$S_V$ [ $\mu\text{C Gy}^{-1}\text{cm}^{-3}$ ]
diF-TE $\textcolor{blue}{S}$ -ADT	$(5.2 \pm 0.3) \cdot 10^{-2}$	$V_{DS} = -3\text{V}$ $V_{GS} = -2\text{V}$	$80 \pm 10$
diF-TE $\textcolor{magenta}{G}$ -ADT	$(6.9 \pm 0.4) \cdot 10^{-2}$		$3400 \pm 400$



# Improving sensitivity

$$\Delta I_{PG} = G I_{CC}$$

Photoconductive gain

$$G = \frac{\tau_r(\rho_x)}{\tau_t}$$

→ Carrier lifetime

→ Transit time

$$\tau_t = \frac{L^2}{V\mu}$$



Increasing **charge carriers density (OTFTs)**



Increasing **attenuated fraction**



Increasing **mobility (BUT low dark current)**



# Increasing attenuated fraction & mobility:

## TIPGe

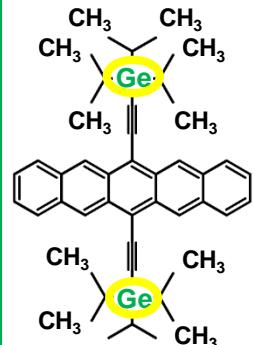
NOVEL MOLECULE!



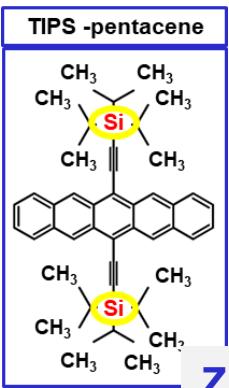
University of  
Kentucky

Prof. John Anthony

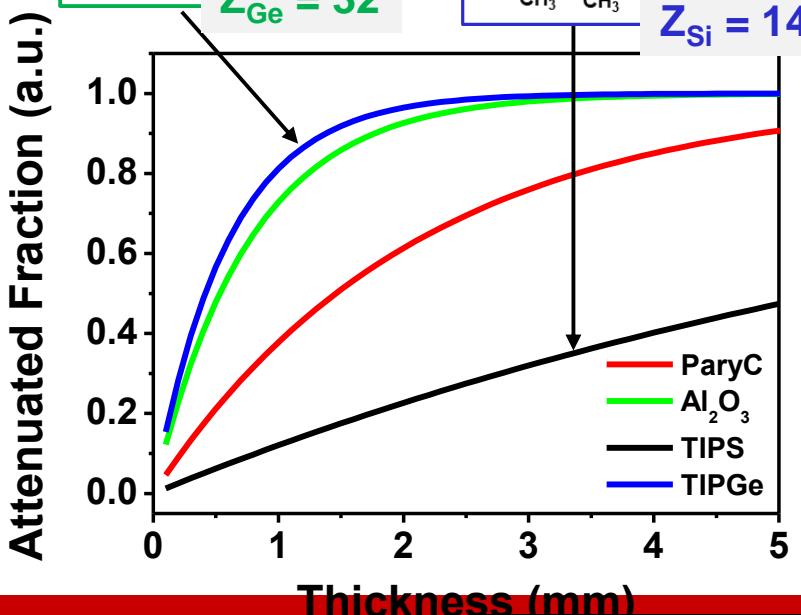
TIPGe -pentacene



$$Z_{\text{Ge}} = 32$$



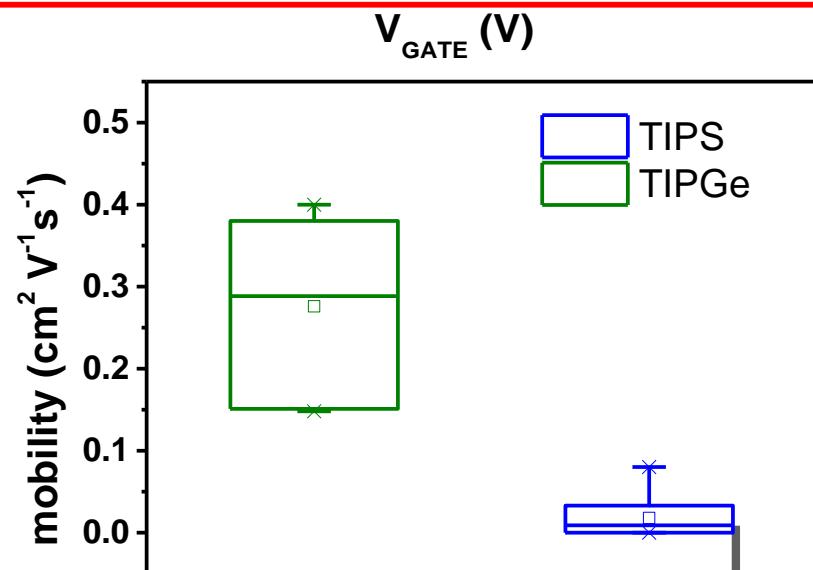
$$Z_{\text{Si}} = 14$$

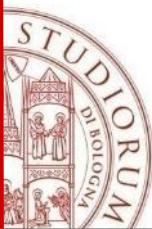


→ TIPGe attenuated fraction 0.03% @ 200nm (x10 times than TIPS film)

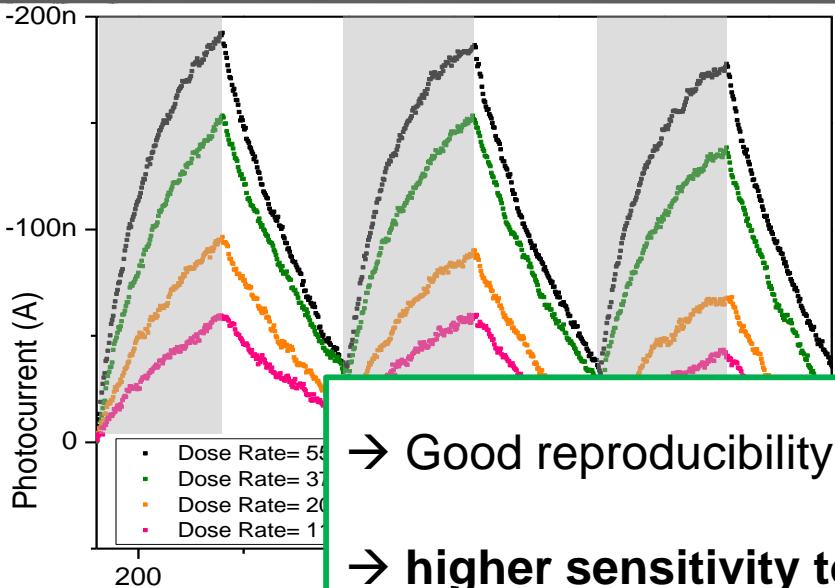
→ TIPGe mobility 0.27 cm<sup>2</sup>/Vs (x10 times than TIPS film)

Higher sensitivity than TIPS is expected



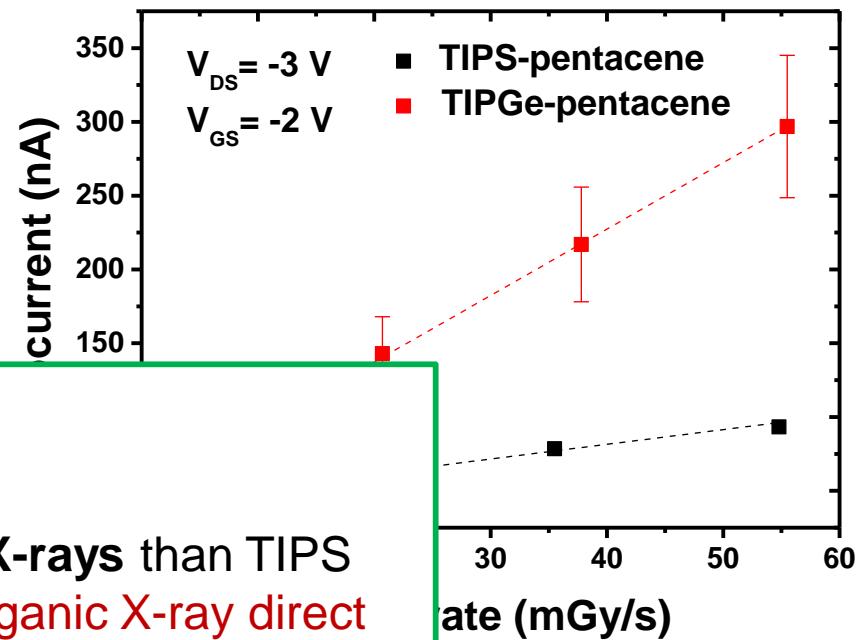


# Increasing attenuated fraction & mobility: TIPGe

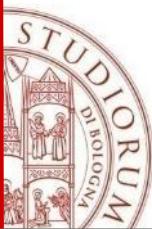


→ Good reproducibility

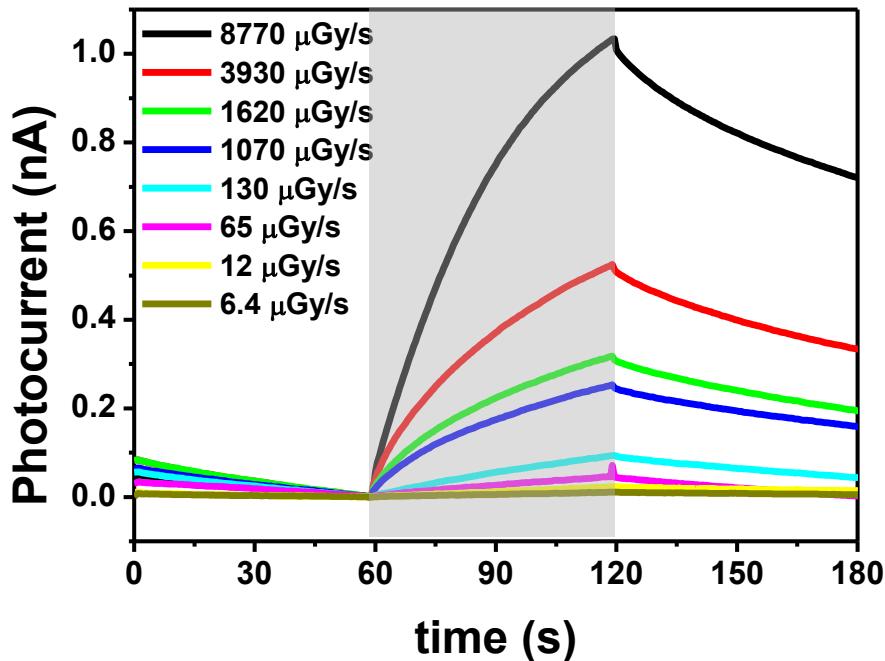
→ higher sensitivity to X-rays than TIPS  
Record value for full-organic X-ray direct  
detectors:  $9.0 \times 10^5 \mu\text{C Gy}^{-1} \text{cm}^{-3}$



Molecule	Solution	Mobility [ $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ ]	Operative voltage	Film thickness [nm]	Average Sensitivity [ $\text{nC Gy}^{-1}$ ]	$S_\gamma$ [ $\mu\text{C Gy}^{-1} \text{cm}^{-3}$ ]
TIPS	0.5wt.% in toluene dropcast	$0.02 \pm 0.01$	$V_{DS} = -3\text{V}$ $V_{GS} = -2\text{V}$	150	$960 \pm 70$	$(2.6 \pm 0.2) \cdot 10^5$
TIPGe		$0.27 \pm 0.09$		200	$3080 \pm 20$	$(6.2 \pm 0.1) \cdot 10^5$



# Increasing attenuated fraction & mobility: TIPGe



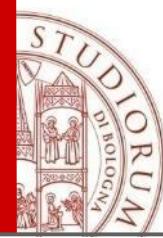
## LIMIT OF DETECTABILITY

Lowest detectable  
dose rate  
**1.66  $\mu\text{Gy s}^{-1}$**

Lowest detectable  
total dose  
**12  $\mu\text{Gy}$**



Molecule	Solution	Mobility [ $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ ]	Operative voltage	Film thickness [nm]	Average Sensitivity [ $\text{nC Gy}^{-1}$ ]	$S_\gamma$ [ $\mu\text{C Gy}^{-1} \text{cm}^{-3}$ ]
TIPS	0.5wt.% in toluene dropcast	$0.02 \pm 0.01$	$V_{DS} = -3\text{V}$ $V_{GS} = -2\text{V}$	150	$960 \pm 70$	$(2.6 \pm 0.2) \cdot 10^5$
TIPGe		$0.27 \pm 0.09$		200	$3080 \pm 20$	$(6.2 \pm 0.1) \cdot 10^5$



# Response to commercial dental radiography system – Low doses

L. Basiricò et al., 2020 *Frontiers in Physics*, in press

## Commercial W-target dental panoramic X-ray tube

70 kVp - 4 mA

exposure time window [0.5 -2] s

Dose range [15.5 – 64.6]  $\mu$ Gy

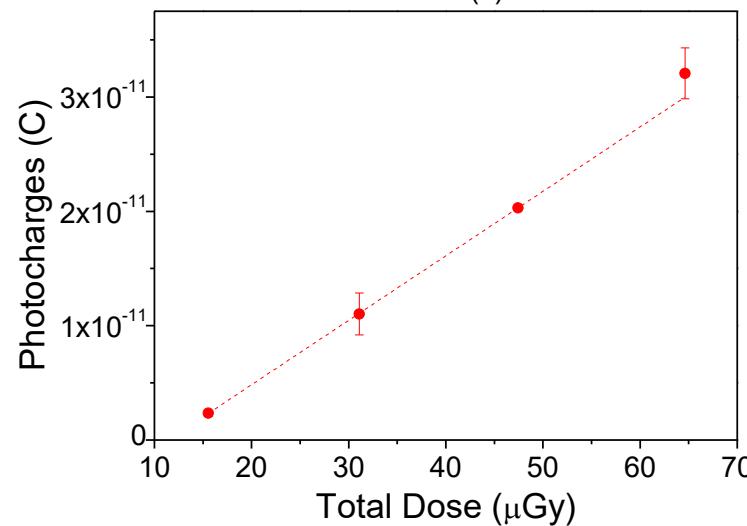
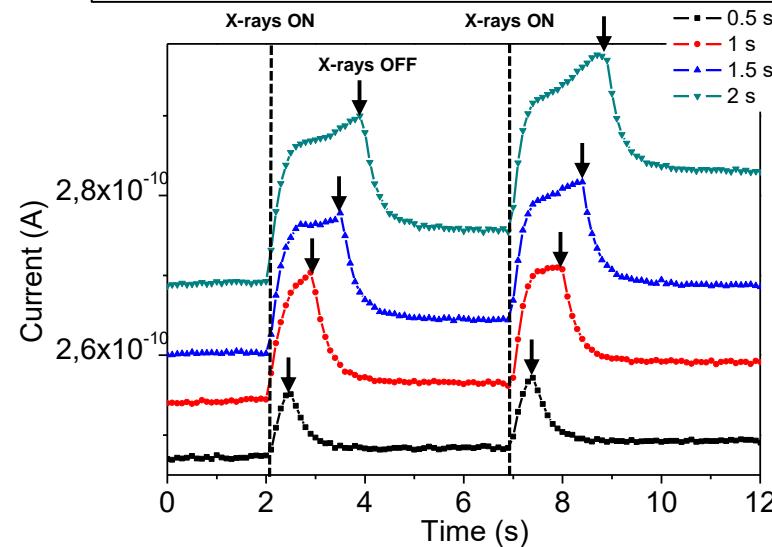


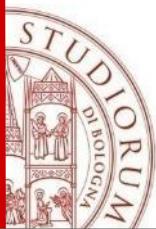
COMPAGNIA ELETTRONICA ITALIANA

OPX/105



by Skan-X Radiology Devices S.p.A





# Response to commercial dental radiography system – Shorter pulses

L. Basiricò et al., 2020 *Frontiers in Physics, in press*

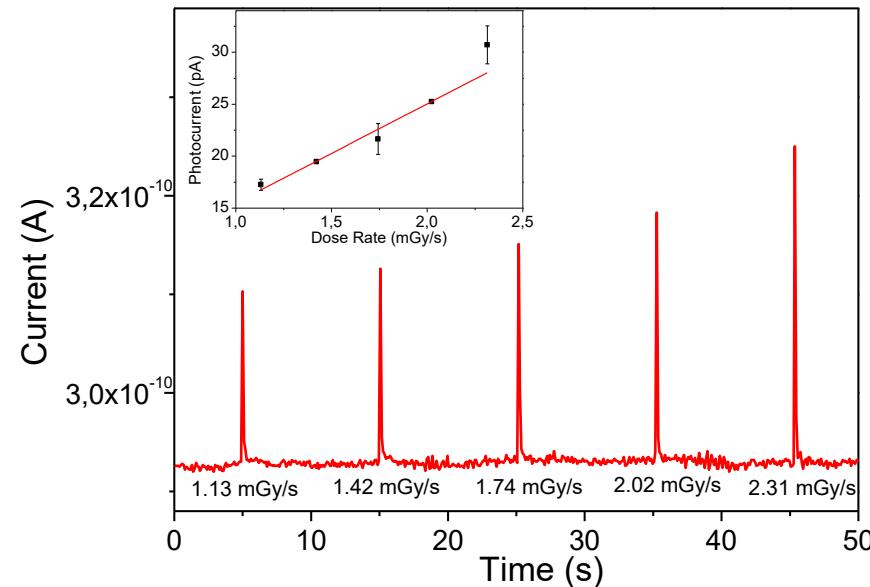
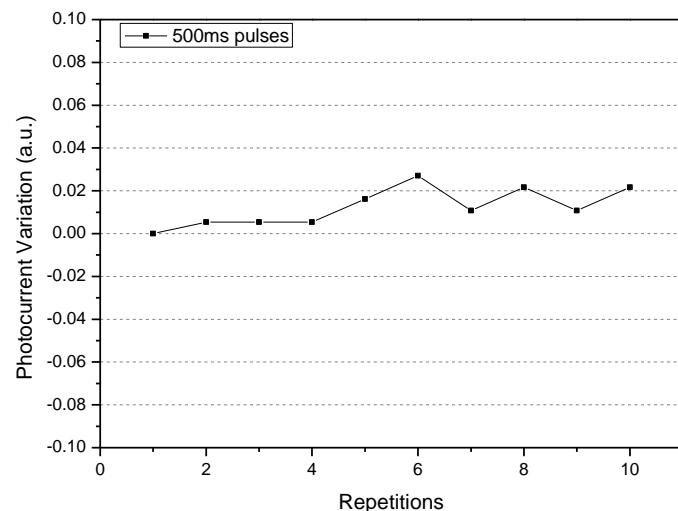
## Commercial dental radiography system

Intraskan DC by Skanray Europe srl



dose rates  
[1.13-2.31] mGy s<sup>-1</sup>

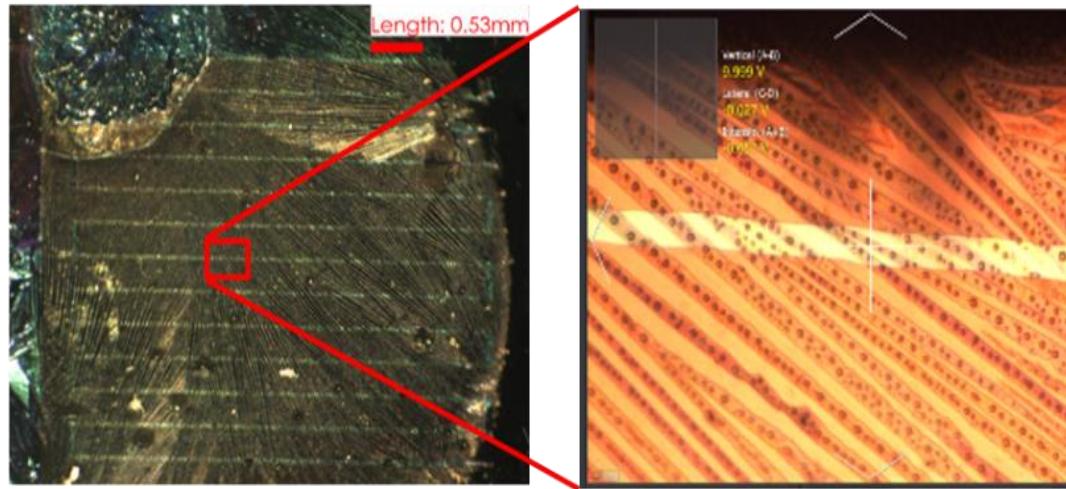
100ms pulses  
70keV



Linear response following **typical frame rates** of a commercial dental radiographic apparatus

Deviation of 2.7% for 10 subsequent repetitions of 500 ms X-rays exposure (Lag Effect reduction)

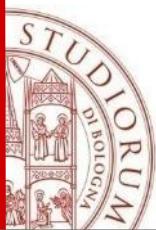
# Further Improving Sensitivity



**Limited area coverage (about 43%)**

potential sensitivity for full area coverage:

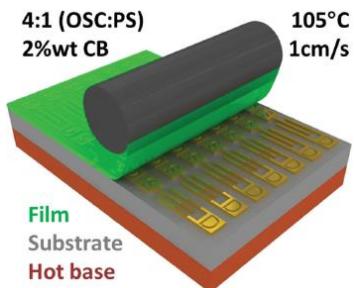
$$21 \cdot 10^5 \mu\text{C Gy}^{-1}\text{cm}^{-3}$$



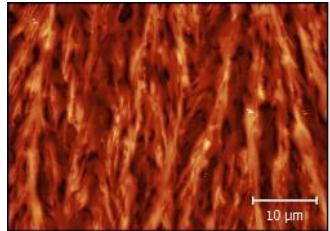
# Large Area controlled deposition

## Improve area coverage and electrical transport

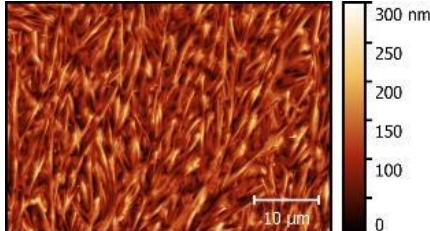
BAMS: bar-assisted meniscus shearing



**LOW SPEED**



**HIGH SPEED**



I.Temino et al.  
Under consideration

**Grain size  
( $\mu\text{m}^2$ )**

**Mobility  
( $\text{cm}^2/\text{Vs}$ )**

**Sensitivity  
( $\mu\text{C}/\text{Gy cm}^3$ )**

**Photoconducti ve GAIN**

**LOW SPEED**

17.3

0.023

$(7 \pm 1) \cdot 10^2$

$7 \cdot 10^5$

**HIGH SPEED**

5.7

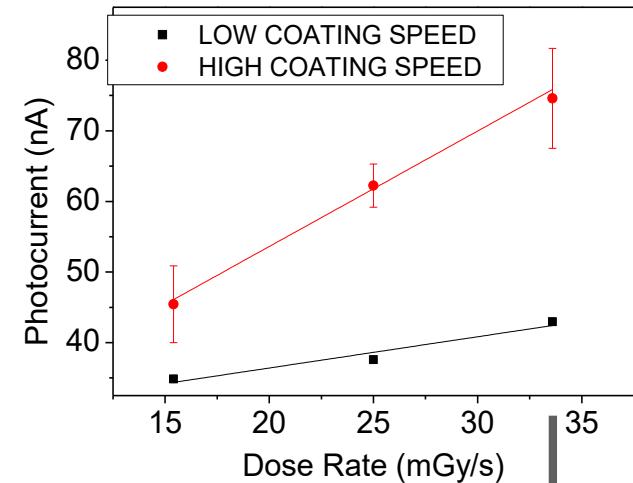
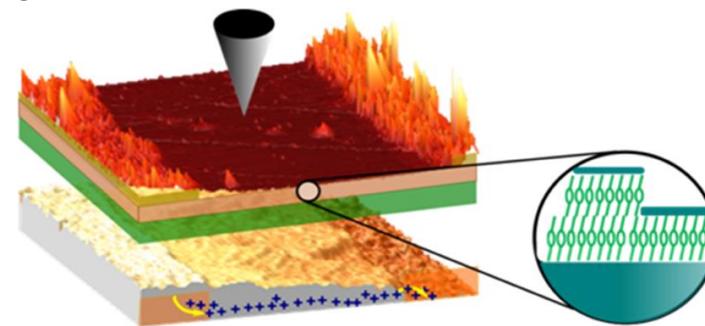
0.020

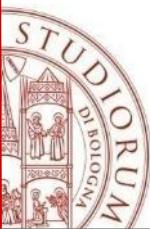
$(2.6 \pm 0.8) \cdot 10^3$

$4 \cdot 10^6$

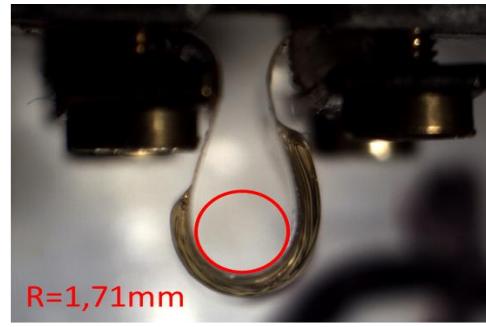
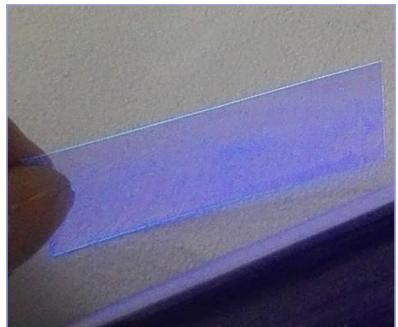
## Study the ROLE OF DEFECTS

Blend semiconductor:PS to passivate the interface state with the electrodes and to control grain size



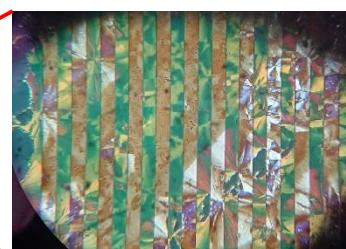
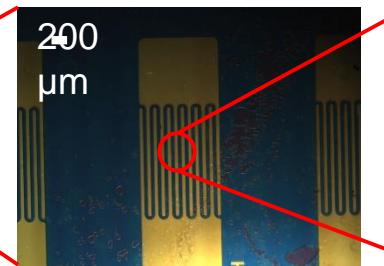
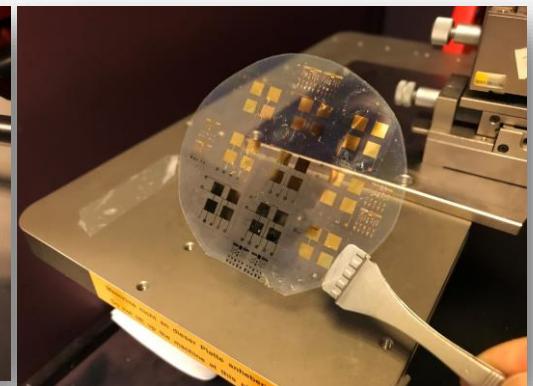
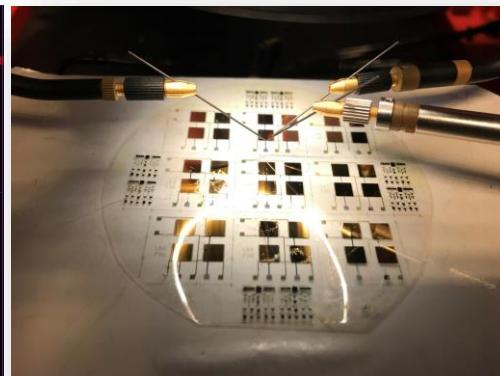
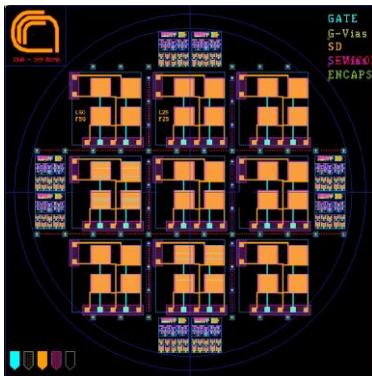


# Ongoing research on FIRE

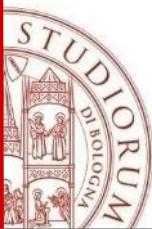


***Study of the best flexible scintillator/substrate coupling***

***Devices' design and fabrication process development***

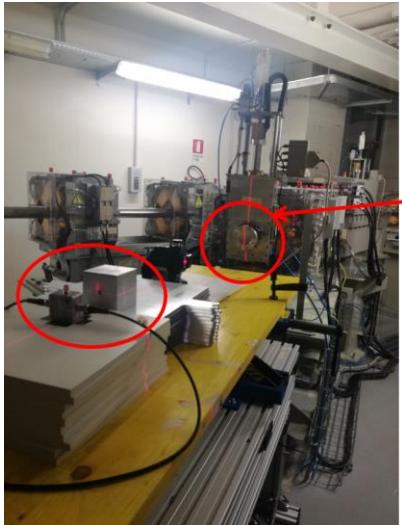


***Scalability by means of high throughput solution coating techniques***

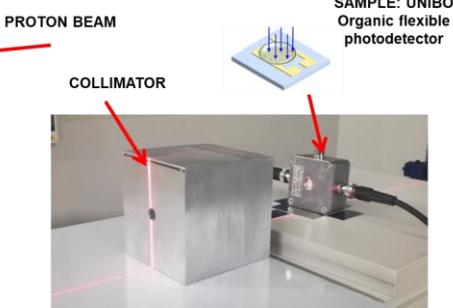


# Ongoing research on FIRE

## TIFPA

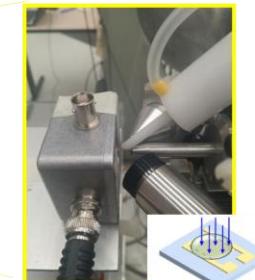
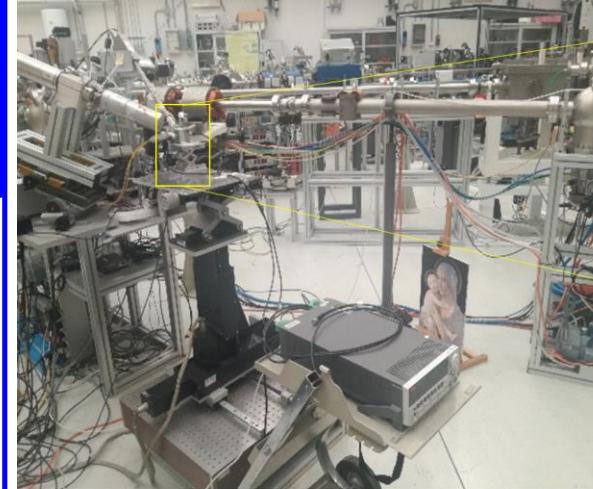


EXPOSURE CONDITION:  
ENERGY: 70 MeV ; TIME 60 s  
CURRENTS: [2.5; 5; 10; 15; 20] nA



**Tests under proton beam  
@TIFPA and LABEC**

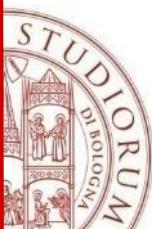
## LABEC



SAMPLE: UNIBO  
Organic flexible  
photodetector

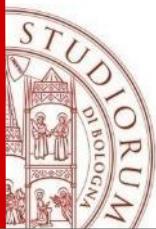
**...outcome in the next few  
months...!**

EXPOSURE CONDITION  
Energy: 5 MeV  
Intensity: 1-150 pA  
Time : 10 s



# Related Publication

- L. Basiricò, A. Ciavatti et al. *Medical Applications of Tissue-Equivalent, Organic-based Flexible Direct X-ray Detectors*, Frontiers in Physics, in press.
- L. Basiricò, S.P. Senanayak, A. Ciavatti, M. Abdi-Jalebi, B. Fraboni, H. Sirringhaus, *High sensitivity direct X-ray detectors based on solution-processed cesium-containing mixed triple cation perovskite thin films*, *Adv. Funct. Mater.* 29, 1902346 (2019).
- S. Lai, G. Casula, P. Cosseddu, L. Basiricò, A. Ciavatti, F. D'Annunzio, C. Loussert, V. Fischer, B. Fraboni, M. Barbaro, A. Bonfiglio, *A plastic electronic circuit based on low voltage, organic thin-film transistors for monitoring the X-Ray checking history of luggage in airports*, *Organic Electronics*, 58, 263-269, 2018.
- A. Ciavatti, L. Basiricò, I. Fratelli, S. Lai, P. Cosseddu, A. Bonfiglio, J.E. Anthony, B. Fraboni, *Boosting Direct X-Ray Detection in Organic Thin Films by Small Molecules Tailoring*, *Adv. Func. Mater.*, 1806119, (2018).
- G. Pipan, M. Bogar, A. Ciavatti, L. Basiricò, T. Cramer, B. Fraboni, A. Fraleoni-Morgera, *Direct Inkjet Printing of TIPS-Pentacene Single Crystals onto Interdigitated Electrodes by Chemical Confinement*, *Adv. Mater. Interfaces*, 1700925, 2017.
- S. Lai, P. Cosseddu, L. Basiricò, A. Ciavatti, B. Fraboni, A. Bonfiglio, *A highly sensitive, direct X-Rays detector based on a low-voltage Organic Field-Effect Transistor*, *Adv. El. Mat*, 3, 8, 1600409, 2017.
- L. Basiricò, A. F. Basile, P. Cosseddu, S. Gerardin, T. Cramer, M. Bagatin, A. Ciavatti, A. Paccagnella, A. Bonfiglio, B. Fraboni, Space environment effects on flexible, low-voltage organic thin film transistors, *ACS Appl. Mater. Interfaces*, 9 (40), pp 35150–35158, 2017.
- A. Ciavatti, T. Cramer, M. Carroli, L. Basiricò, R. Fuhrer, D. DeLeew, B. Fraboni, Dynamics of Direct X-ray Detection Processes high-Z Bi<sub>2</sub>O<sub>3</sub> nanoparticles-loaded PFO polymer-based diodes, *Appl. Phys. Lett.* 111, 183301, 2017.
- L. Basiricò, A. Ciavatti, T. Cramer, P. Cosseddu, A. Bonfiglio, B. Fraboni, *Direct X-ray Photoconversion in Flexible Organic Thin Film Devices Operated Below 1 V*, *Nat. Comm.*; 7: 13063, 2016.
- A. Ciavatti, P.J. Sellin, L. Basiricò et al, *Charged-particle spectroscopy in organic semiconducting single crystals*, *Applied Physics Letters* 108/15 (2016) 153301.
- L. Basiricò, A. Ciavatti, M. Sibilia, A. Fraleoni-Morgera, S. Trabattoni, A. Sassella, B. Fraboni, Solid state Organic X-ray Detectors based on Rubrene Single Crystals, *Nuclear Science, IEEE Transactions on*, 62 (4), 1791-1797, 2015.
- B. Fraboni, A. Ciavatti, L. Basiricò and A. Fraleoni Morgera, Organic Semiconducting Single Crystal as solid-state sensors for ionizing radiation, *Faraday Discuss.*, 174, 219-234, 2014.
- Ciavatti, A. et al. Toward low-voltage and bendable X-ray direct detectors based on organic semiconducting single crystals. *Adv. Mater.* 27, 7213–7220 (2015).
- Fraboni, B. et al. Organic semiconducting single crystals as next generation of low-cost, room-temperature electrical X-ray detectors. *Adv. Mater.* 24, 2289–2293 (2012).



# Acknowledgements

## Flexible organic Ionizing Radiation dEectors

INFN (Italian Institute for  
Nuclear Physics)  
(2019-2021)



## *Flexible, Large-area patches for real-time detection of ionizing radiation in medical diagnostics* (2019-2021)

Fortre<sup>mu</sup>



## **POC- ROXFET** *Radiation detectors based on flexible high mobility oxide transistors* (2019-2020)

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**UNI Bologna**  
**Prof. Beatrice Fraboni**  
Dr. Andrea Ciavatti  
Dr. Tobias Cramer  
Ilaria Fratelli



**UNI Cagliari**  
Prof. Annalisa Bonfiglio  
Prof. Piero Cosseddu  
Dr. Stefano Lai



**UNI Kentucky**  
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**ICMAB-CSIC**  
Dr. Marta Mas-Torrent  
Dr. Inés Temiño  
Adrian Tamayo

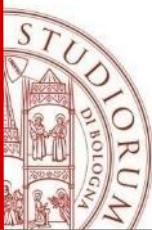


Elettra Sincrotrone Trieste

**ELETTRA Sincrotron, Trieste**  
Dr. Giuliana Tromba  
Dr. Diego Dreossi



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# Thank you for your attention