



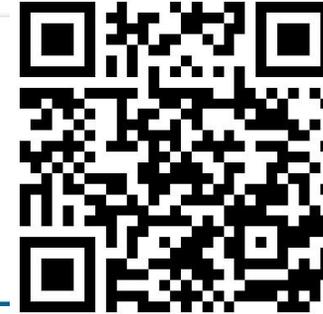
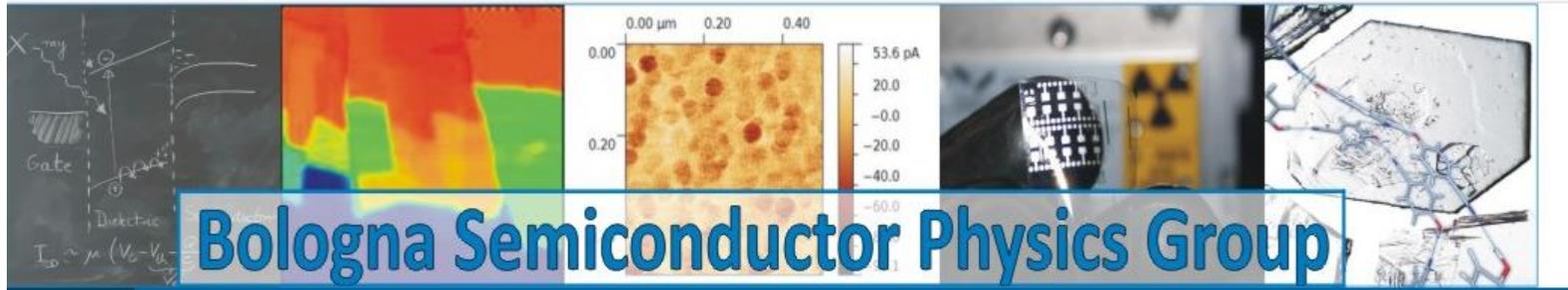
ALMA MATER STUDIORUM  
UNIVERSITÀ DI BOLOGNA

# X-ray detectors with ultrahigh sensitivity based on high performance printed Organic Field Effect Transistors

**Laura Basiricò**<sup>1,2</sup>, Ilaria Fratelli<sup>1,2</sup>, Andrea Ciavatti<sup>1,2</sup>, Adrian Tamayo<sup>3</sup>, Carme Martínez-Domingo<sup>3</sup>, Paolo Branchini<sup>4</sup>, Elisabetta Colantoni<sup>4</sup>, Stefania De Rosa<sup>4</sup>, Luca Tortora<sup>4</sup>, Adriano Contillo<sup>5</sup>, Raul Santiago<sup>6</sup>, Stefan T. Bromley<sup>6,7</sup>, John E. Anthony<sup>8</sup>, Marta Mas-Torrent<sup>3</sup>, Ioannis Kymissis<sup>9</sup>, and Beatrice Fraboni<sup>1,2</sup>

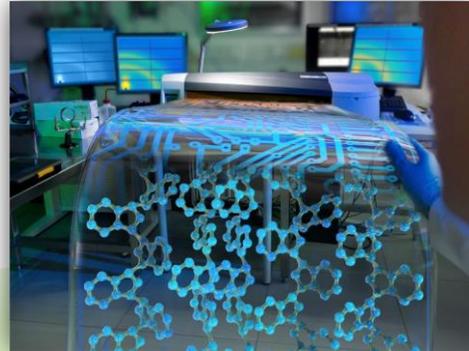
1. Physics and Astronomy Department of the University of Bologna, Viale Berti Pichat 6/2, 40127, Bologna (BO), Italy
2. Institute of Nuclear Physics, INFN-BO, Viale Berti Pichat 6/2, 40127, Bologna (BO), Italy
3. Institut de Ciència de Materials de Barcelona and Networking Research Center on Bioengineering, Biomaterials and Nanomedicine, Campus de la Universitat Autònoma de Barcelona, Cerdanyola, E-08193 Barcelona, Spain
4. Surface Analysis Laboratory INFN Roma Tre, Via della Vasca Navale 84, 00146 Rome, Italy
5. Elettra-Sincrotrone Trieste, Trieste, Italy
6. Departament de Ciència de Materials i Química Física & Institut de Química Teòrica i Computacional, Universitat de Barcelona, Barcelona, Spain
7. Institució Catalana de Recerca i Estudis Avançats, Barcelona, Spain
8. Center for Applied Energy Research, University of Kentucky, United States
9. Department of Electrical Engineering, Columbia University, New York, NY 10027, United States

# SEMICONDUCTOR PHYSICS GROUP @ DEPARTMENT OF PHYSICS AND ASTRONOMY-UNIVERSITY OF BOLOGNA



# ORGANIC-HYBRID SEMICONDUCTORS FOR IONIZING RADIATION DETECTION - ADVANTAGES

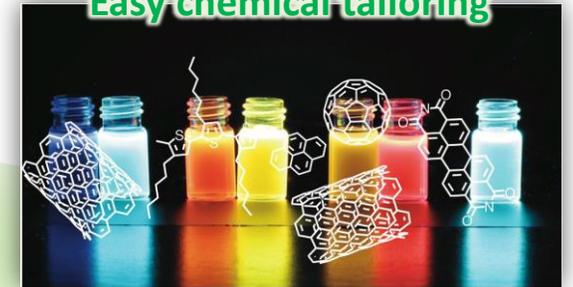
Low-cost large-area printing techniques



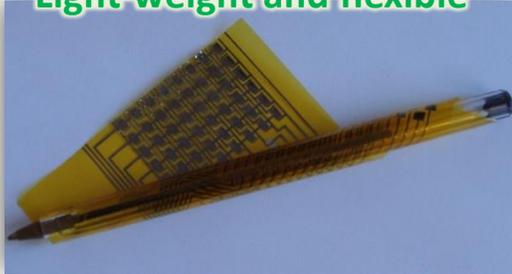
Wearable



Easy chemical tailoring



Light-weight and flexible



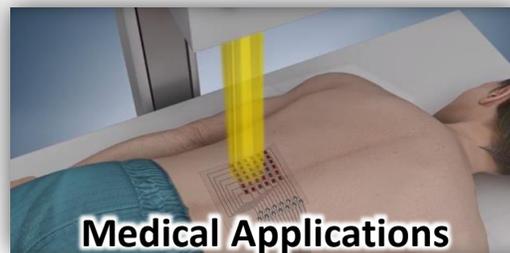
New generation of  
**low cost, low power supply and mechanical flexible**  
**Thin and conformable** sensor panels and patches



Space Missions



Airport Security



Medical Applications



Nuclear wastes  
management



Cultural Heritage

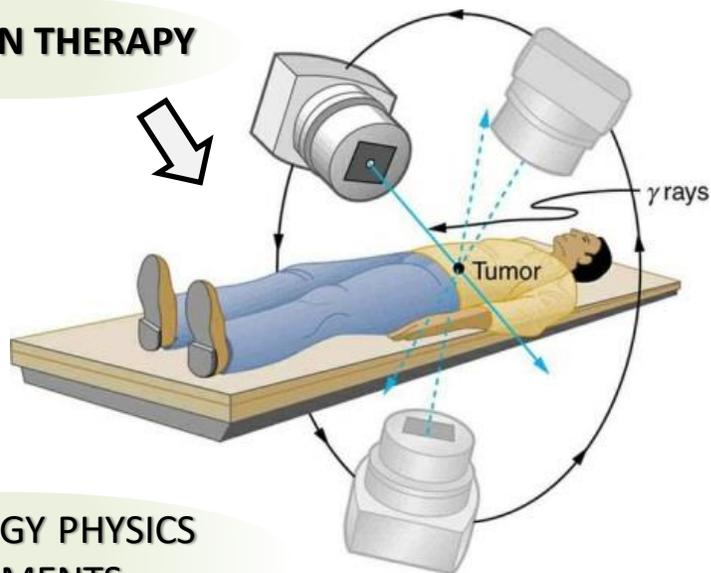


# THIN FILM AND LARGE AREA: WHERE?

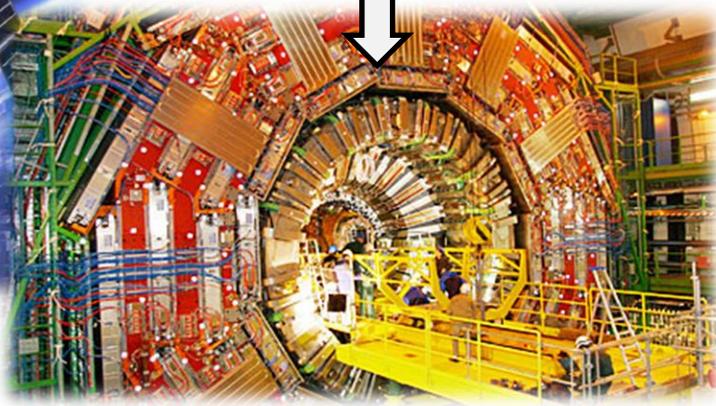
SPACE MISSIONS



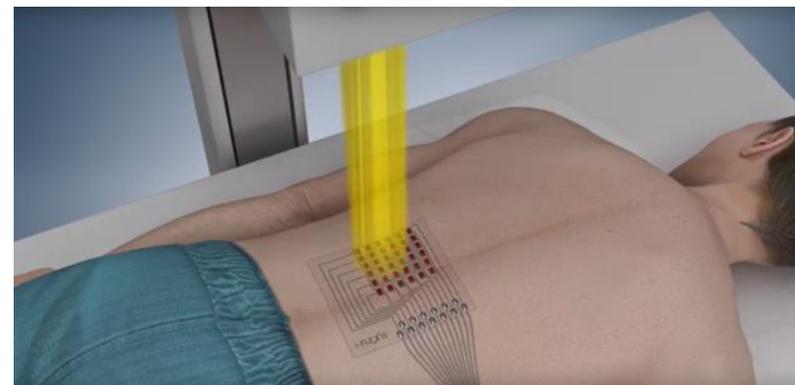
RADIATION THERAPY



HIGH ENERGY PHYSICS EXPERIMENTS



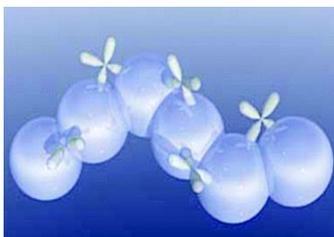
- **Light-weight** for limited amount of materials
- Possibility to **cover large surfaces** at low cost
- **Real-time** beam monitoring
- **Radiation hard** to strong fluxes due to weak radiation abortion
- **In-situ** dose evaluation thank to conformability to human tissues



# FLEXIBLE LARGE AREA ELECTRONICS: MATERIAL PLATFORMS

## High Mobility Oxide Semiconductors

e.g.  $\text{Ga}_x\text{In}_y\text{Zn}_z\text{O}$

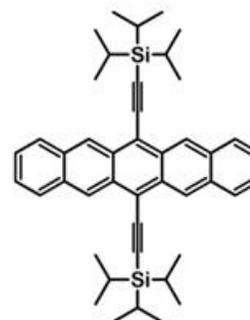
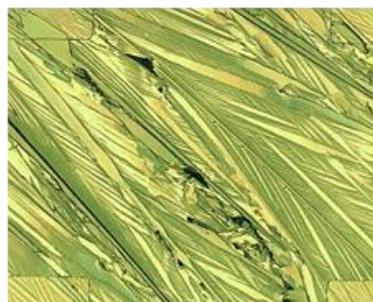


Physical/solution deposition  
 $\mu = 10 - 50 \text{ cm}^2/\text{Vs}$

*T. Cramer et al., Sc.Adv., 4, 63 (2018)*

## Organic Semiconductors

e. g. TIPS pentacene



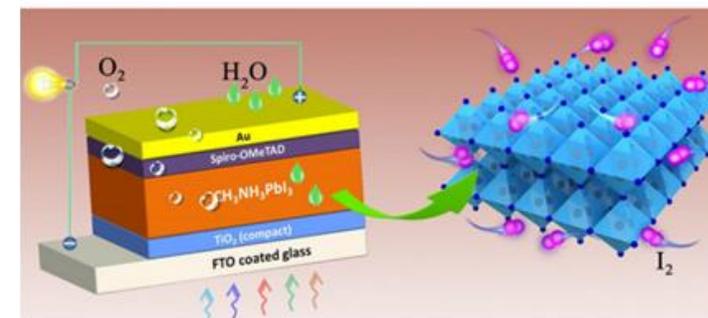
solution deposition  
 $\mu = 1 \text{ cm}^2/\text{Vs}$

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

*I. Temino et al., Nature Comm. 11, 235 (2020)*

## Perovskites

e. g.  $\text{MAPbI}_3$



solution deposition  
 $\mu = 1-600 \text{ cm}^2/\text{Vs}$

*A. Ciavattiet al., Adv. Funct. Mater. 29, 1902346 (2019)*



# FLEXIBLE LARGE AREA ELECTRONICS: MATERIAL PLATFORMS

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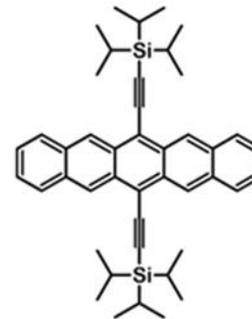
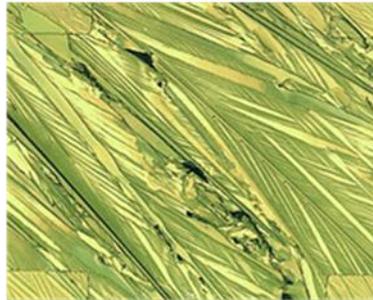


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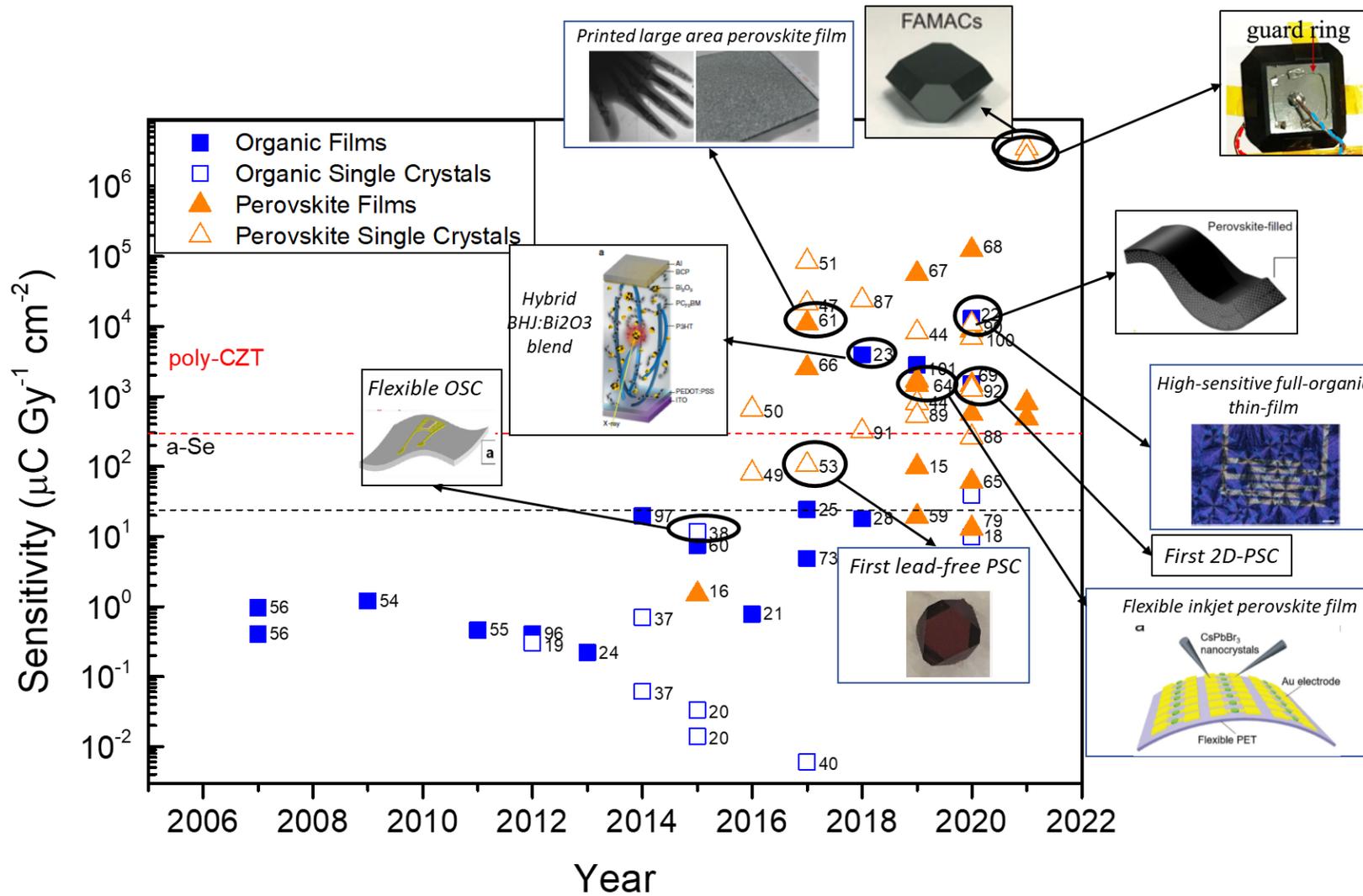


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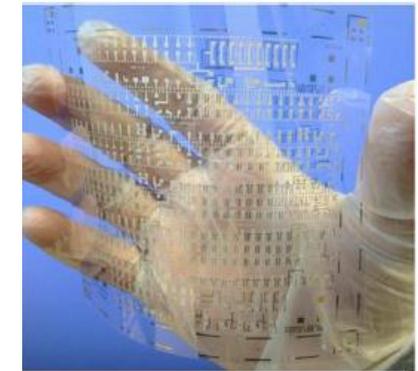


# ORGANIC/HYBRID MATERIALS FOR X-RAY RADIATION DETECTION



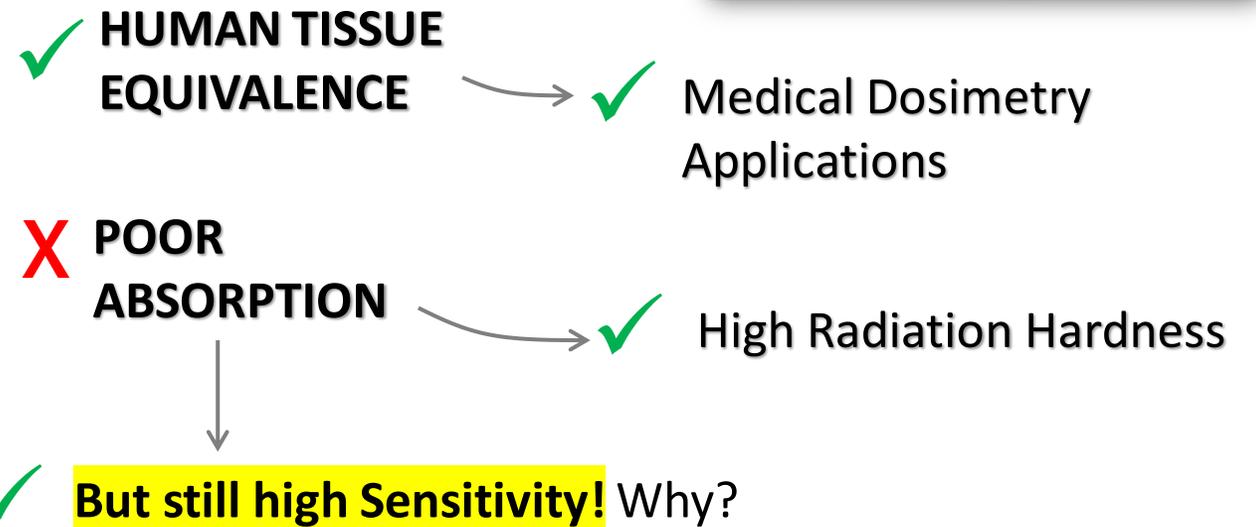
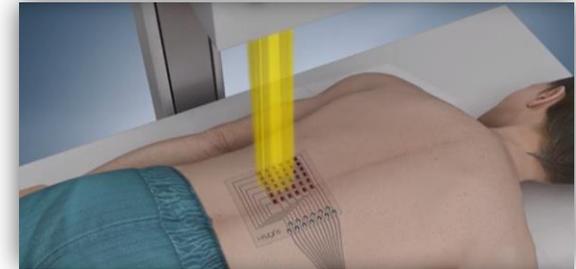
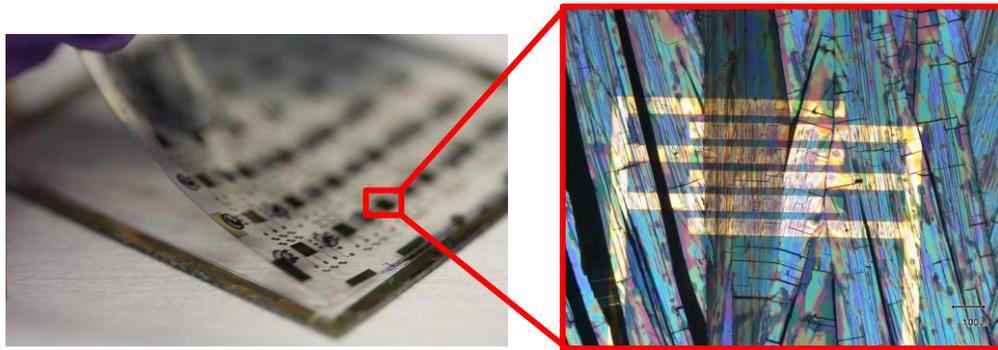
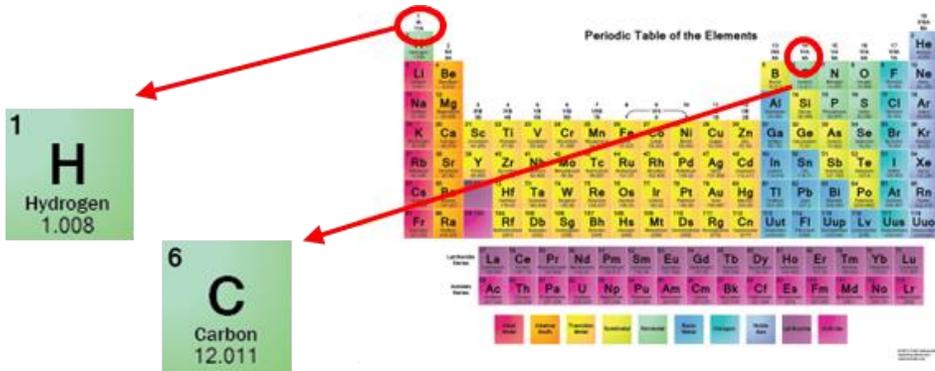
**Sensitivity:  $10^6 \mu\text{C/Gy cm}^2$  @ 0.2V @ RT**

**>> than polyCZT or a-Se**



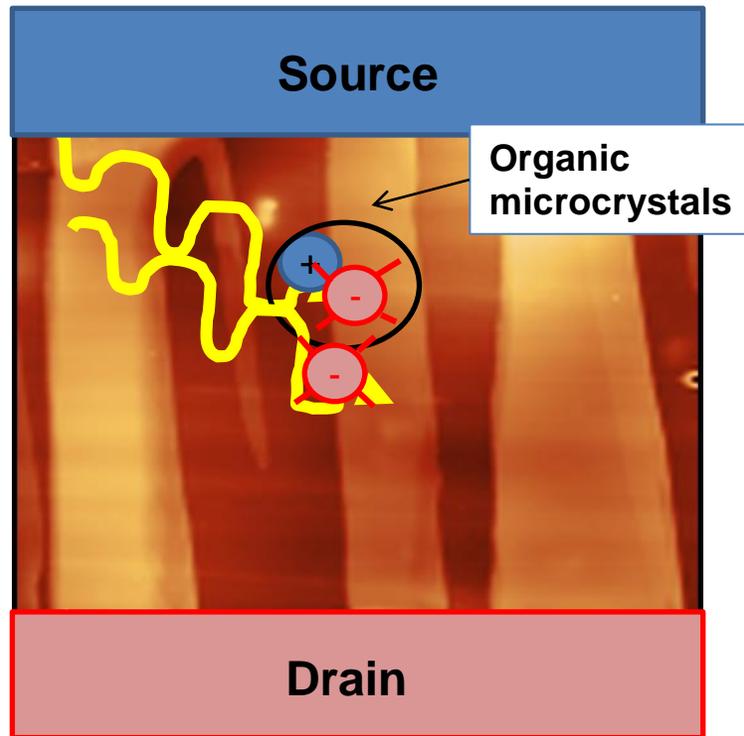
# WHY HIGH SENSITIVITY?

Chemical composition formed  
by  
**LOW-Z ELEMENTS**



# WHY HIGH SENSITIVITY? CHARGE TRAPS AND PHOTOCOCONDUCTIVE GAIN

$$V_{DS}=0.2V$$



- trapping of n-type carriers
- injecting contacts

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.
- 4) **Recombination process takes place**

→  $\Delta I_{PG} = G I_{CC}$

**G = photoconductive gain  $\approx 10^6$**

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

# PHOTOCONDUCTIVE GAIN MECHANISM

$$\textit{Photocurrent} = G \cdot I_{CC} = \frac{\tau_r}{\tau_t} \cdot I_{CC} = \frac{\alpha}{\gamma} \cdot \left[ \alpha \cdot \ln \left( \frac{\rho_0}{\rho_X} \right) \right]^{\frac{1-\gamma}{\gamma}} \cdot \frac{V \cdot \mu}{L^2} \cdot I_{CC}$$

## PHOTOCONDUCTIVE GAIN EFFECT

Study of the electron trap states

→ MORPHOLOGY

→ INTERFACES

## TRANSPORT PROPERTIES

Boosting the electrical mobility

*I. Temiño, L. Basiricò et al., Nat. Commun., vol. 11, 2136, 2020*  
*Tamayo, A., Fratelli, I., et al, Adv. Electron. Mater., 2200293 (2022).*

## IONIZING RADIATION ABSORPTION

Small molecules  
 CHEMICAL TAILORING

*A. Ciavatti et al., Adv. Funct. Mater. 29, 1–8 (2019).*

## DEVICE ARCHITECTURE

OFET structure

*S.Lai et al, Adv.Electr.Mat. 8, 1600409 (2017)*



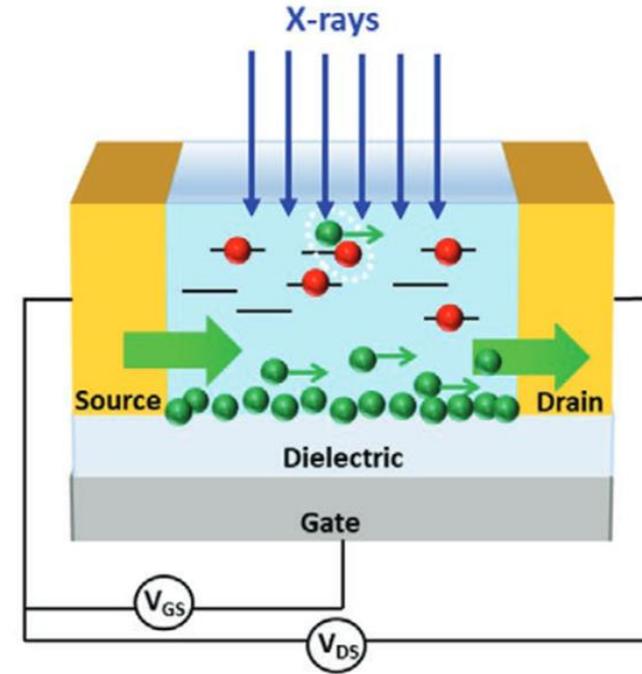
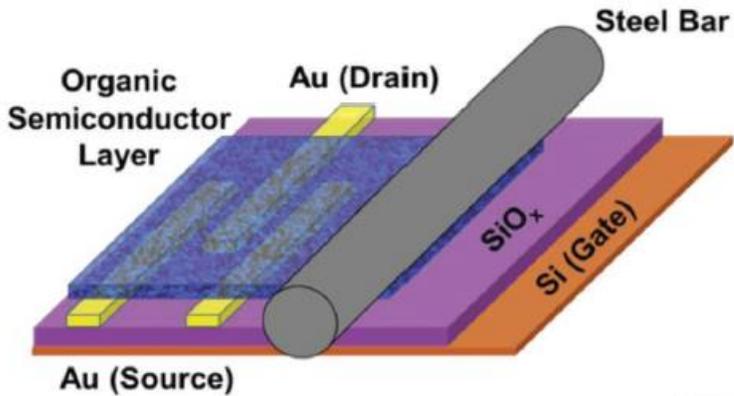
# DEVICE ARCHITECTURE/TRANSPORT PROPERTIES

## Organic Field Effect Transistors as X-Rays detectors

$$\text{Photocurrent} = G \cdot I_{CC} = \frac{\tau_r}{\tau_t} \cdot I_{CC} = \frac{\alpha}{\gamma} \cdot \left[ \alpha \cdot \ln \left( \frac{\rho_0}{\rho_x} \right) \right]^{\frac{1-\gamma}{\gamma}} \cdot \frac{V \cdot \mu}{L^2} \cdot I_{CC}$$

**BAMS:** Bar Assisted Meniscous Shearing Technique

**BOTTOM GATE – BOTTOM CONTACTS OFETs**



$$\rho_0 \nearrow \quad G \nearrow$$



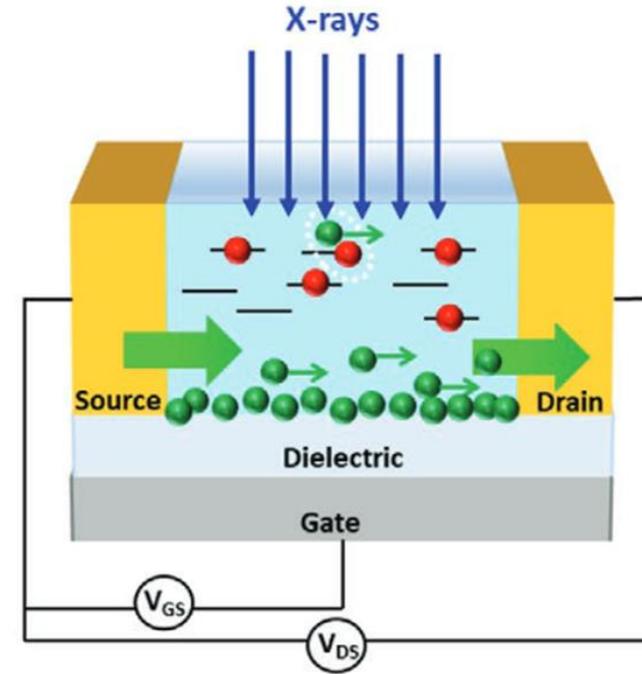
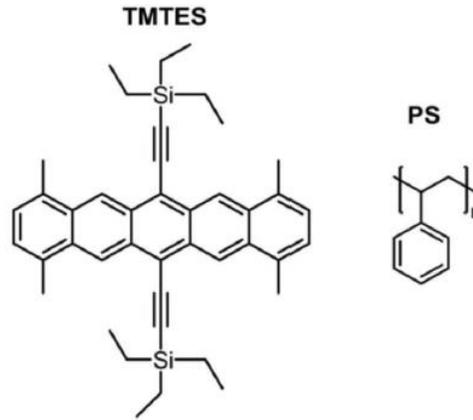
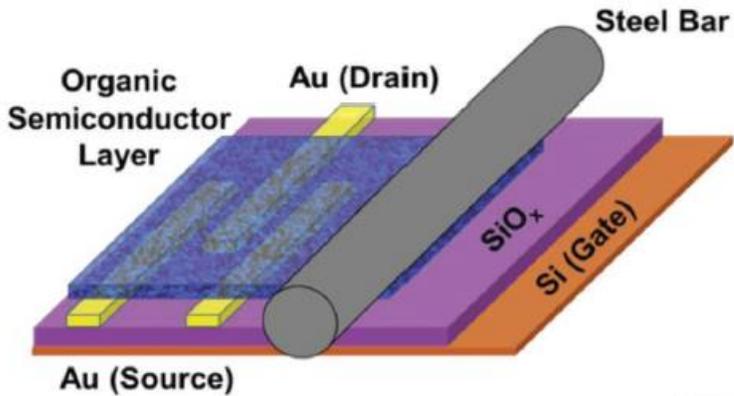
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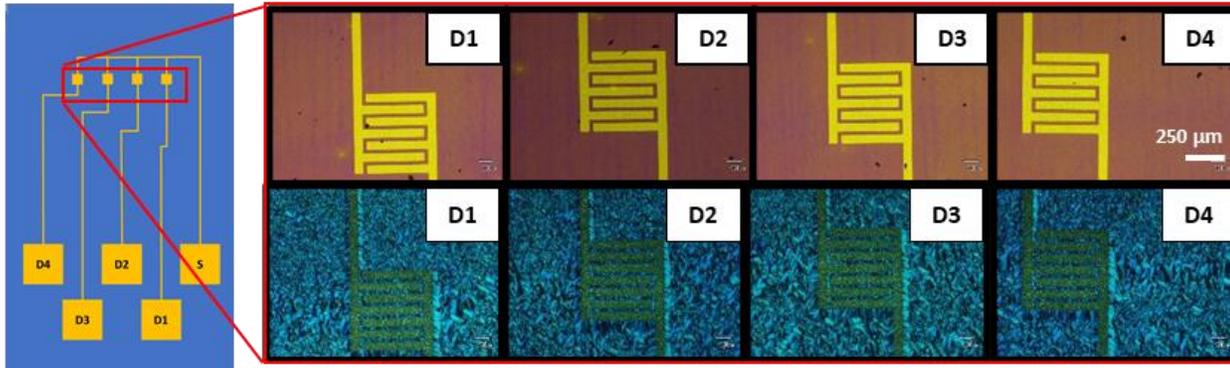
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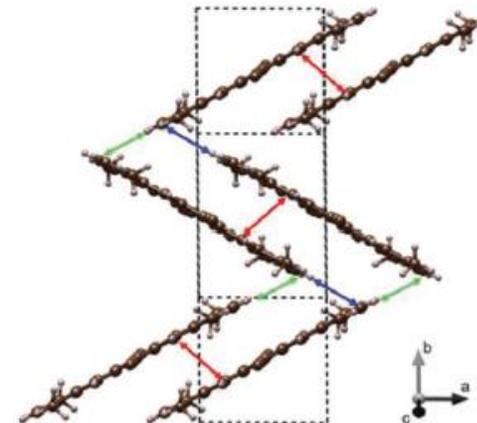
**BOTTOM GATE – BOTTOM CONTACTS OFETs**



$\rho_0 \nearrow$   $G \nearrow$



TMTES - PII



**Crystallization in herringbone motif**  
 → 2D electronic isotropy, more desirable for charge transport



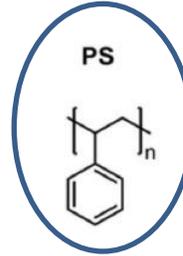
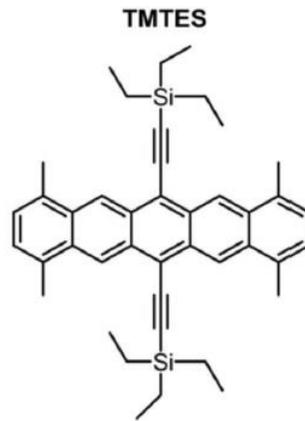
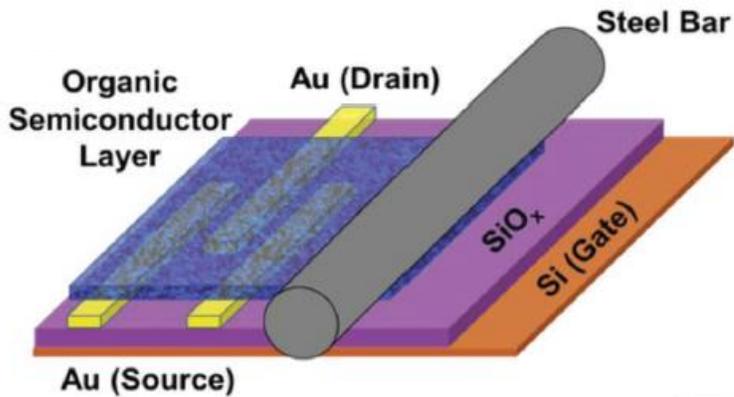
# DEVICE ARCHITECTURE/TRANSPORT PROPERTIES

## Organic Field Effect Transistors as X-Rays detectors

ToF-SIMS: Time-of-Flight Secondary Ion Mass Spectrometry

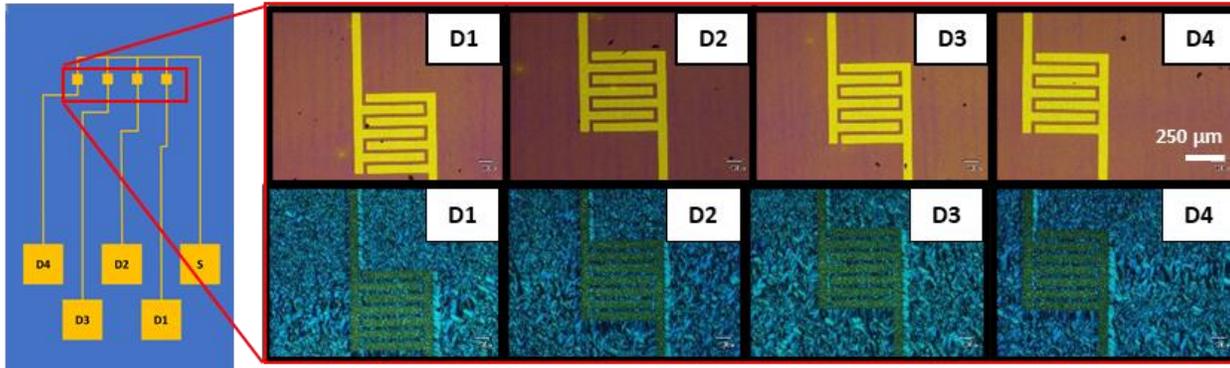
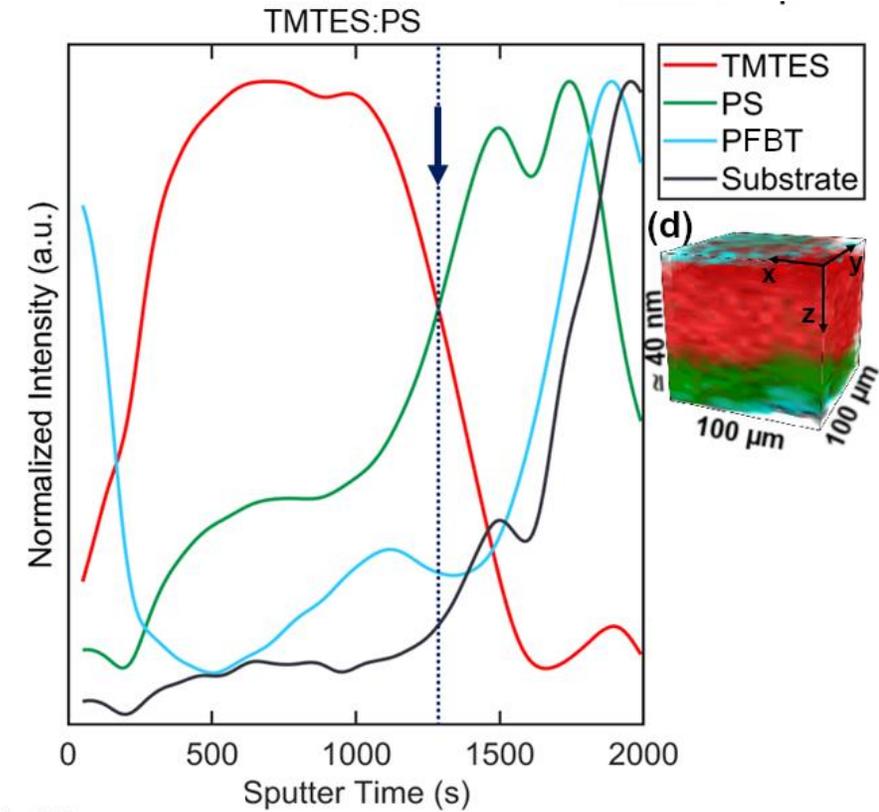
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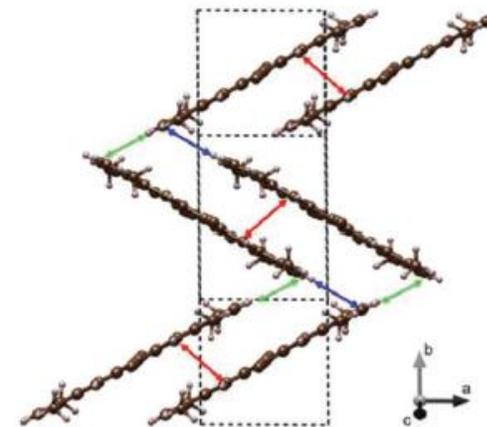


TMTES:PS 2:1

Majority Trap passivation at SiOx/OSC interface



TMTES - PII



**Crystallization in herringbone motif**

→ 2D electronic isotropy, more desirable for charge transport



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# DEVICE ARCHITECTURE/TRANSPORT PROPERTIES

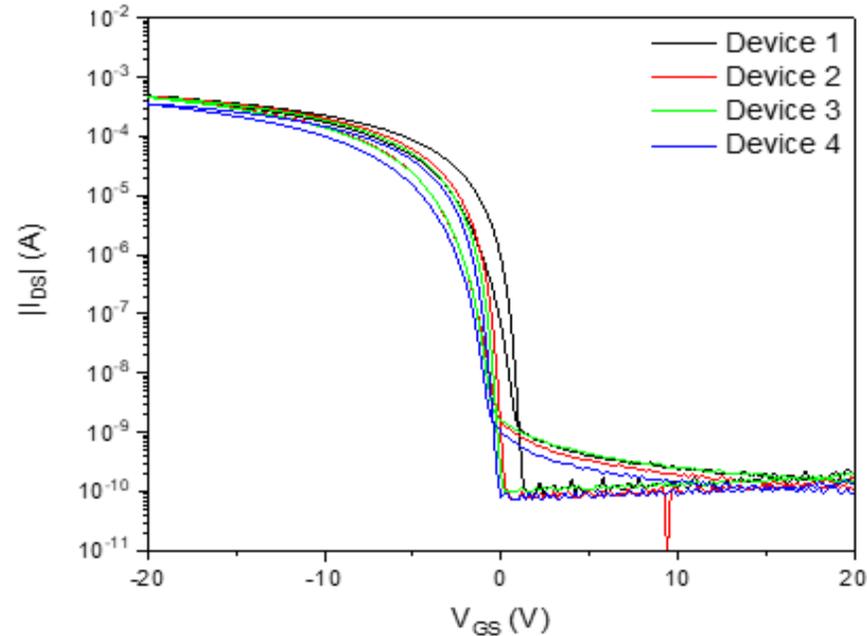
## ROLE OF TRAP STATES: Dielectric/SC interface

High electronic performance and reproducibility

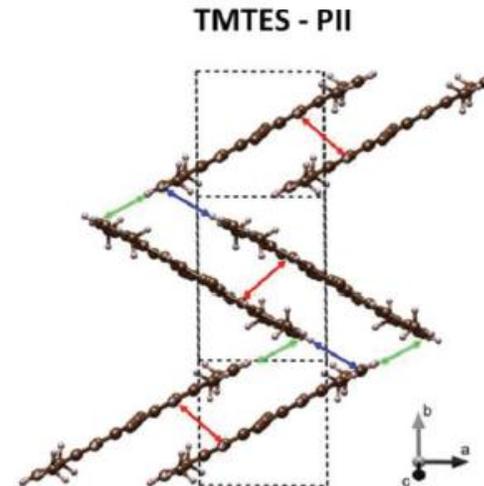
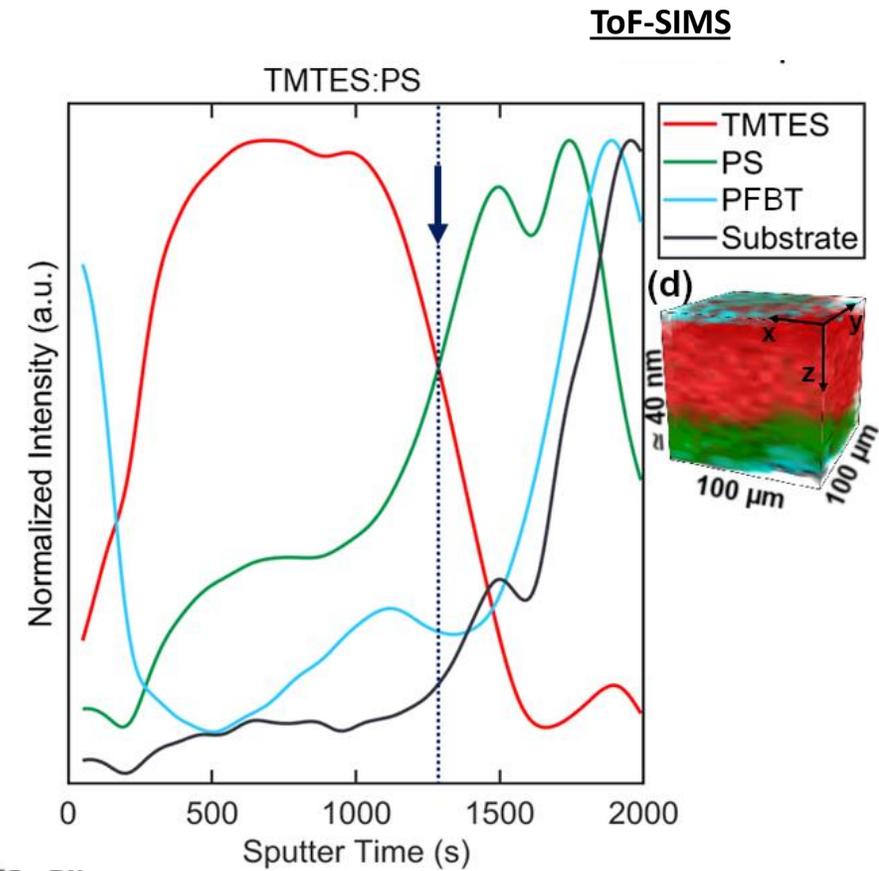
Lower trap density for holes

$(3.9 \pm 0.9) \times 10^{11} \text{ eV}^{-1} \text{ cm}^{-2}$  with PS  
 $(6.2 \pm 1.1) \times 10^{12} \text{ eV}^{-1} \text{ cm}^{-2}$  w/o PS

$\mu_{\text{holes-FET}} \approx 2.6 \pm 0.6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$



Majority Trap passivation at SiO<sub>x</sub>/OSC interface



**Crystallization in herringbone motif**  
 → 2D electronic isotropy, more desirable for charge transport



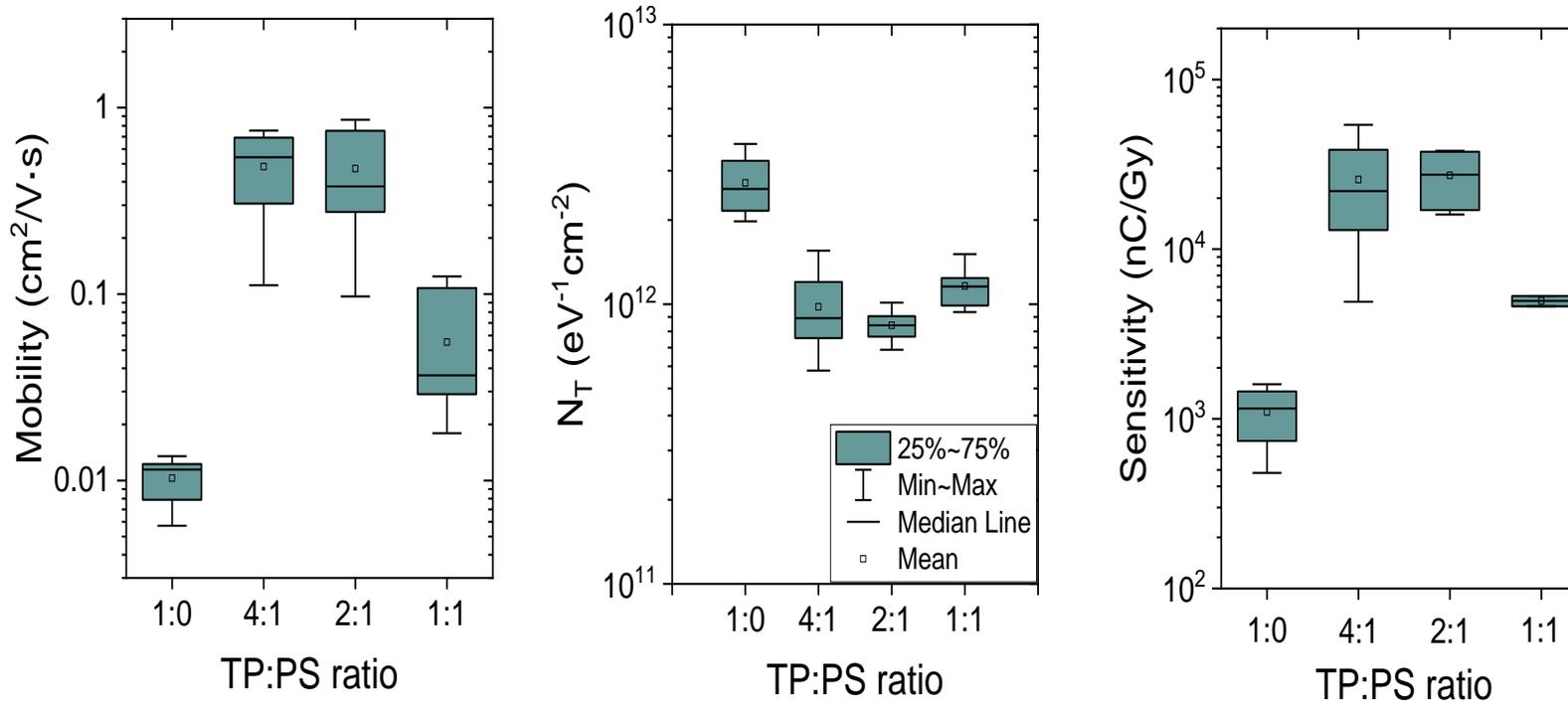
# DEVICE ARCHITECTURE/TRANSPORT PROPERTIES

## ROLE OF TRAP STATES: Dielectric/SC interface

$$\text{Photocurrent} = G \cdot I_{CC} = \frac{\tau_r}{\tau_t} \cdot I_{CC} = \frac{\alpha}{\gamma} \cdot \left[ \alpha \cdot \ln \left( \frac{\rho_0}{\rho_X} \right) \right]^{\frac{1-\gamma}{\gamma}} \cdot \frac{V \cdot \mu}{L^2} \cdot I_{CC}$$

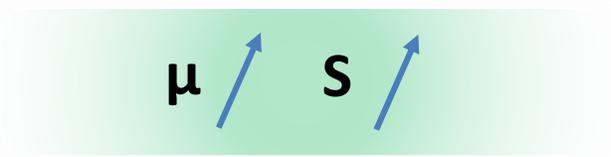
Mo-target X-ray tube 35 kV  
dose rates in the range 5–55 mGy s<sup>-1</sup>

**BLENDS OSC:Polystyrene** passivates the interface state  
with the dielectric → >> *hole mobility*



$$S = \frac{\Delta I}{dr}$$

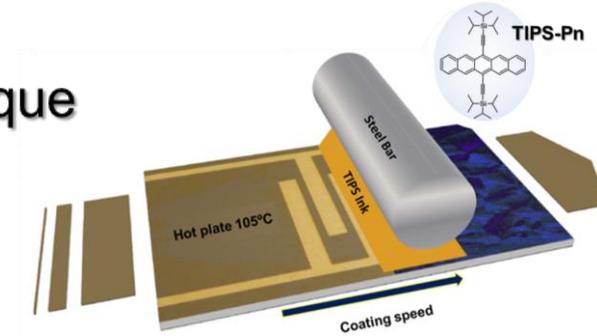
$$S \propto G$$



# FILM MORPHOLOGY: ROLE of GRAIN BOUNDARIES

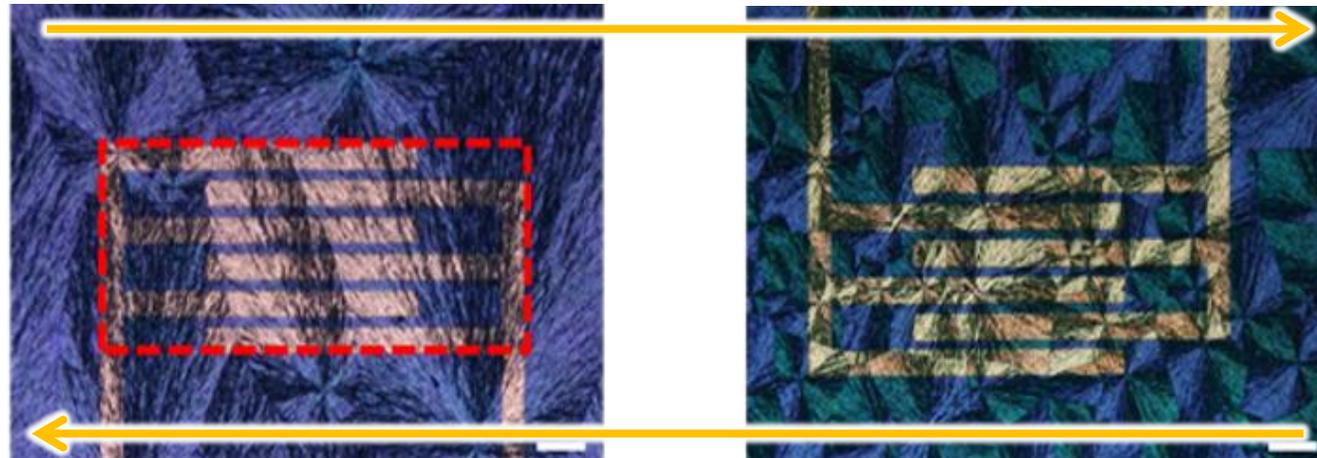
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**BAMS:** Bar Assisted Meniscus Shearing deposition technique



Tuning deposition parameters  $\Rightarrow$  Tuning morphology

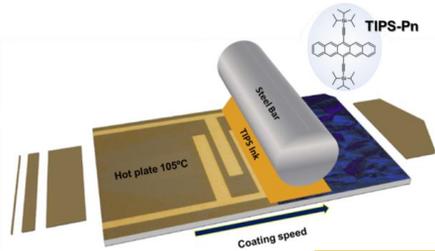
INCREASING THE DEPOSITION SPEED



INCREASING THE GRAIN DIMENSIONS

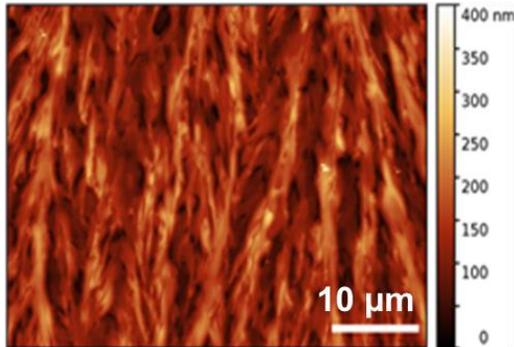
I. Temiño, et al *Nat. Commun.* **11**, 1–10 (2020).

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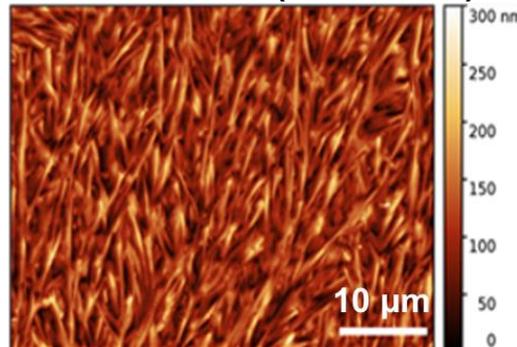


INCREASING THE DEPOSITION SPEED

LOW SPEED (4 mm s<sup>-1</sup>)



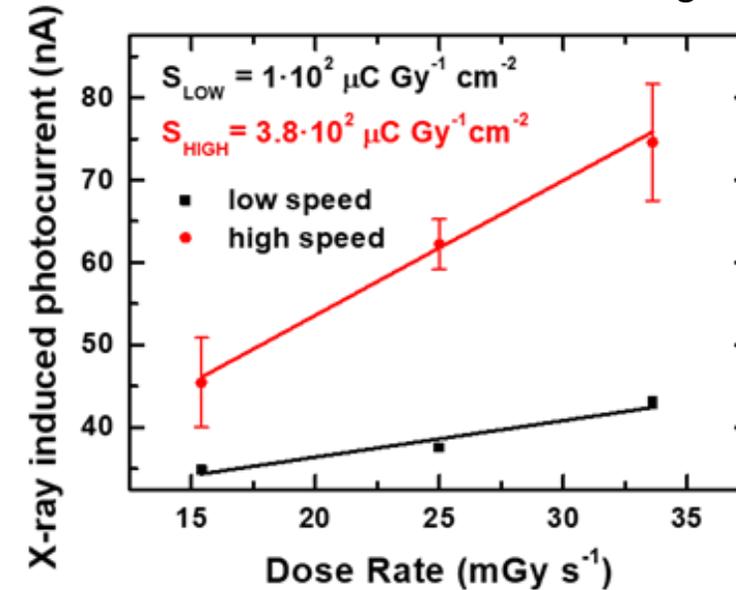
HIGH SPEED (28 mm s<sup>-1</sup>)



Deposition Speed (mm s <sup>-1</sup> )	Grain Size (μm <sup>2</sup> )	Thickness (nm)	Mobility (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	N <sub>T</sub> (10 <sup>12</sup> eV <sup>-1</sup> cm <sup>-2</sup> )	Sensitivity (μC Gy <sup>-1</sup> cm <sup>-2</sup> )
Low (4)	17 ± 3	70 ± 20	(2.5 ± 0.7) · 10 <sup>-2</sup>	1.7 ± 0.4	(1.0 ± 0.2) · 10 <sup>2</sup>
Standard (10)	6 ± 2	80 ± 20	(1.7 ± 0.5) · 10 <sup>-2</sup>	1.8 ± 0.5	(3.8 ± 0.1) · 10 <sup>2</sup>
High (28)	6 ± 2	120 ± 50	(2.4 ± 0.6) · 10 <sup>-2</sup>	1.6 ± 0.4	(3.8 ± 1.2) · 10 <sup>2</sup>

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Mo-target X-ray tube 35 kV  
dose rates in the range 5–55 mGy s<sup>-1</sup>



$$S = \frac{\Delta I}{dr}$$

Grain size ↓ S ↑

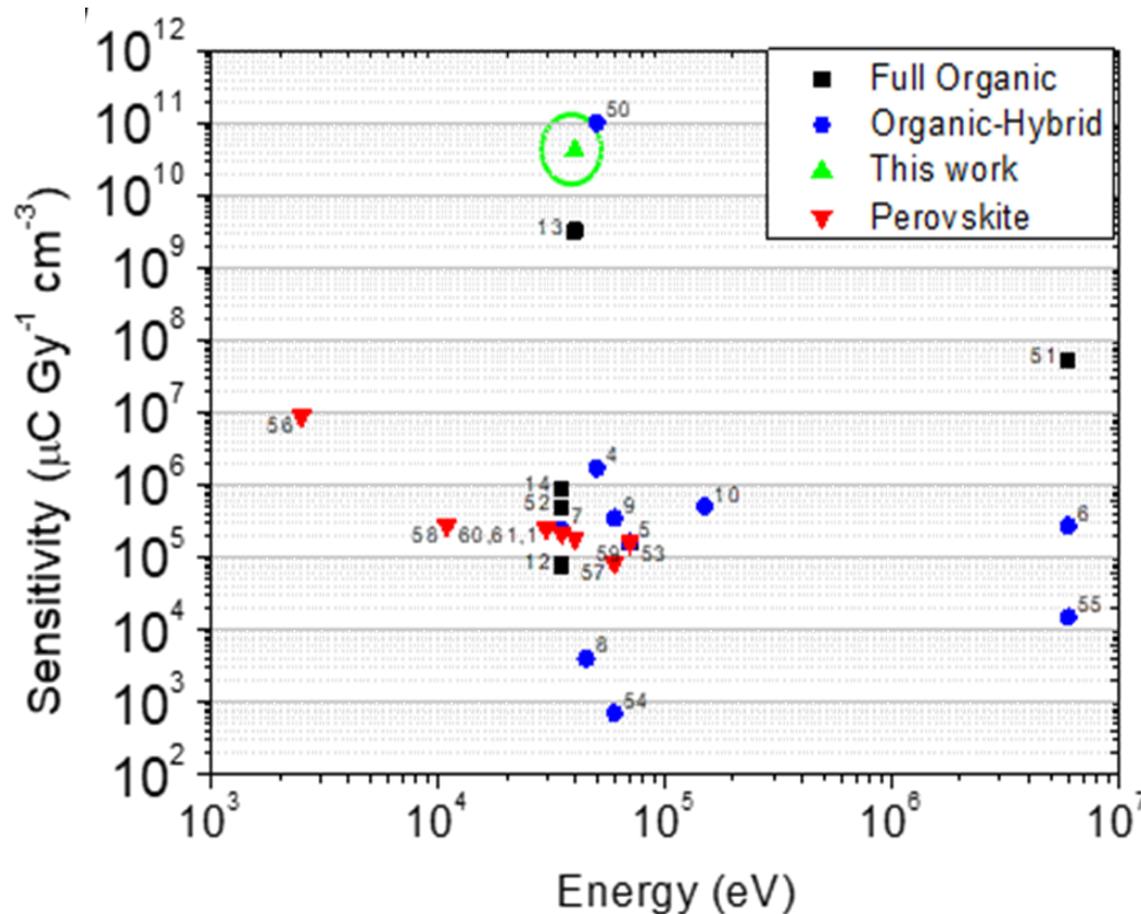
I. Temiño, et al *Nat. Commun.* **11**, 1–10 (2020).



# CHEMICAL TAILORING/TUNING TRANSPORT PROPERTIES

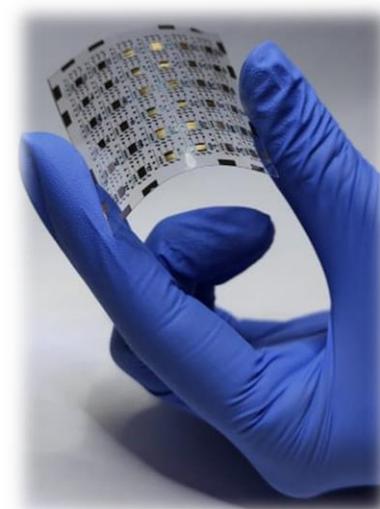
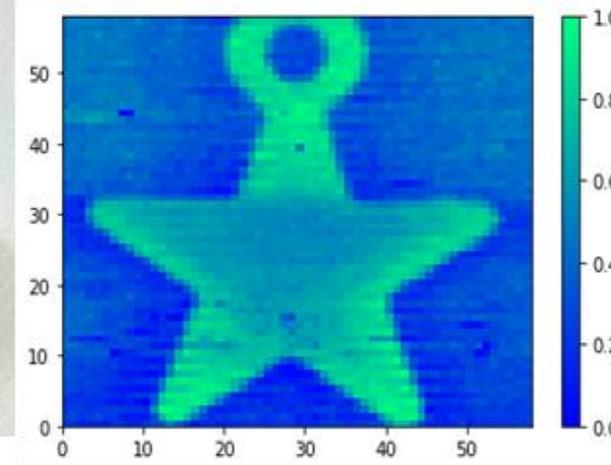
Tamayo, et al, *Adv. Electron. Mater.*, 2200293 (2022).

**Sensitivity =  $4 \times 10^{10} \mu\text{C Gy}^{-1} \text{cm}^{-3}$**



Elettra Sincrotrone Trieste

SYRMEP beamline @ ELETTRA synchrotron



**Transfer on flexible large area substrates**

*Fratelli et al., Adv. Mater. Technol.* **2023**, 2200769

**Beam monitoring for medical application**



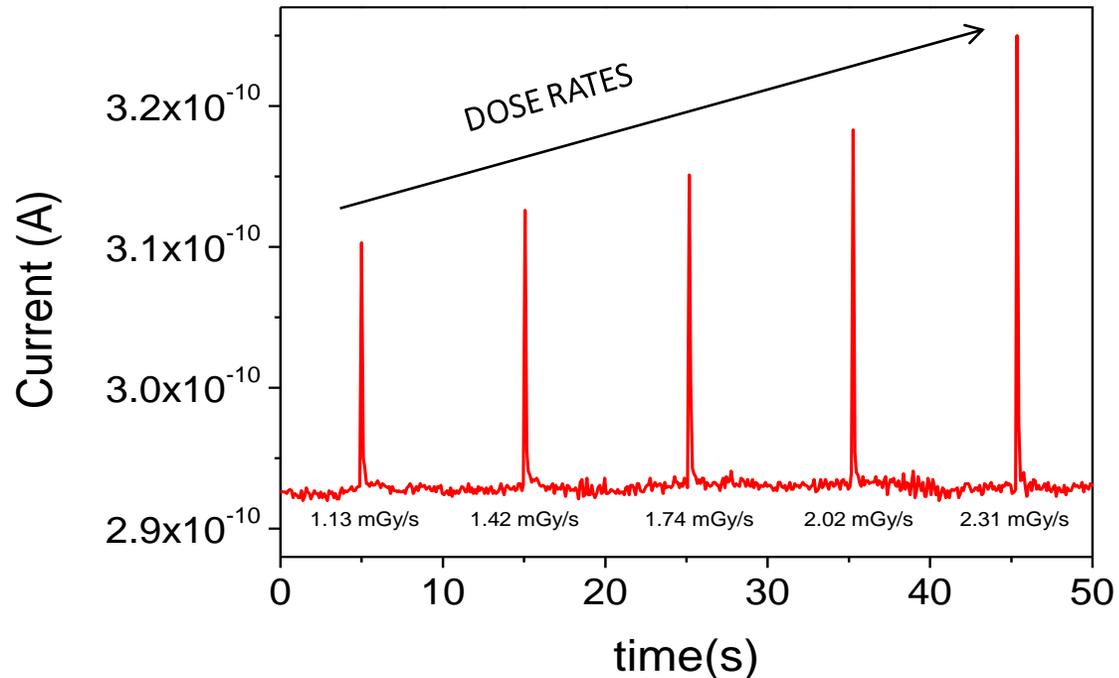
ALMA MATER STUDIORUM  
UNIVERSITÀ DI BOLOGNA

# DIRECT RADIATION DETECTION BY ORGANIC THIN FILMS: VALIDATION IN REAL-LIFE MEDICAL APPLICATION

## DENTAL RADIOGRAPHY



Intraskan DC (dental radiography system, Skaneray Europe srl.)



**Limit Of Detection = 8  $\mu$ Gy**

typical **dose delivered** during dental diagnostic

**100 ms**

Typical **frame rates** of a commercial dental radiographic apparatus

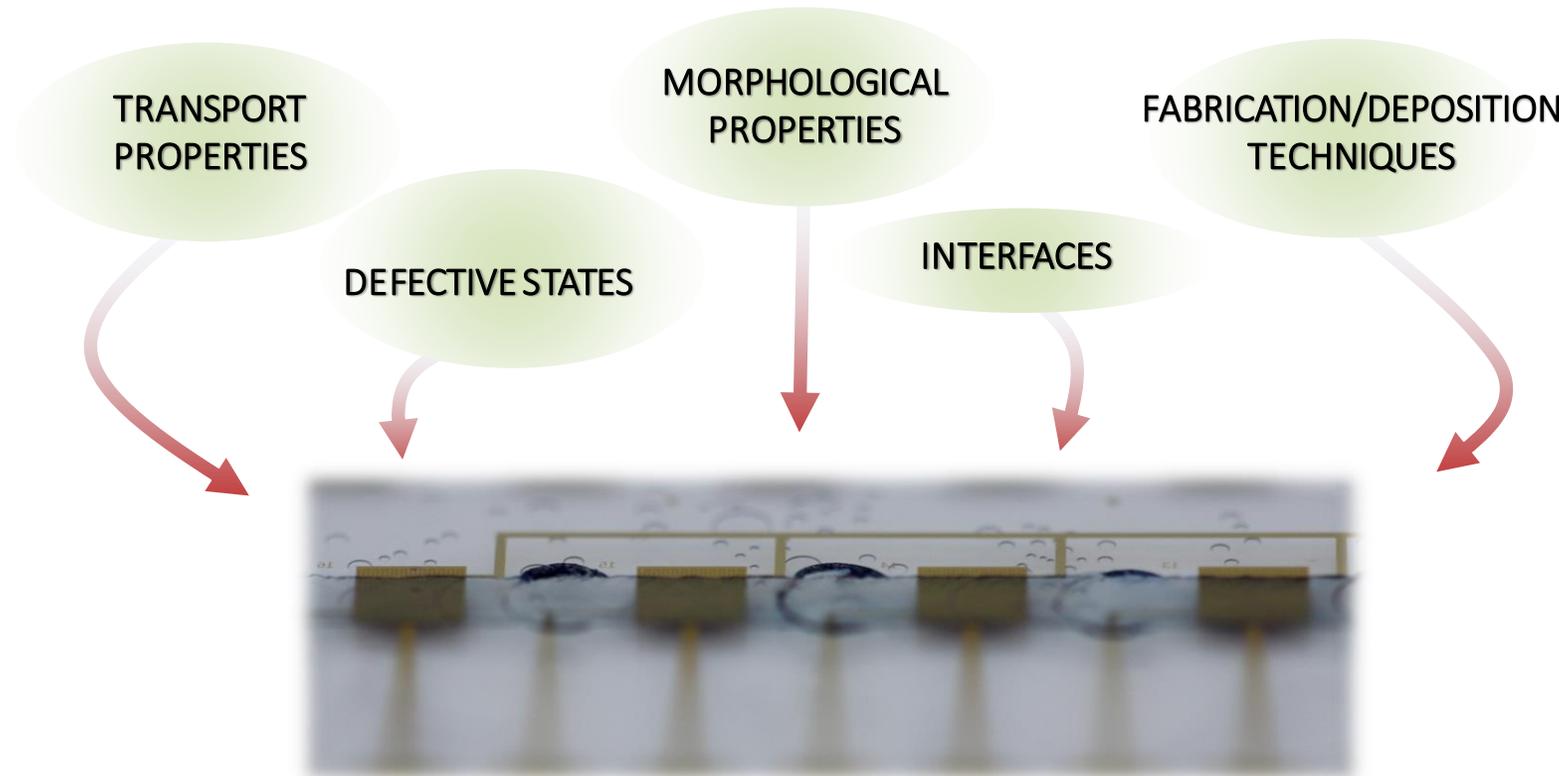
L. Basiricò, et. al. *Frontiers in Physics*. **8**, 11 (2020).



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UNIVERSITÀ DI BOLOGNA

# CONCLUSIONS

Fully organic, lightweight, printed radiation detectors based on **high performance printed Organic Field Effect Transistors**, can effectively and **directly detect ionizing radiation with ultrahigh sensitivity**



# ACKNOWLEDGMENTS



People of Fraboni's Group working on this research  
@ Department of Physics and Astronomy – DIFA UNIBO

<https://site.unibo.it/semiconductor-physics/en>

Prof. Beatrice Fraboni  
Dr. Andrea Ciavatti  
Dr. Ilaria Fratelli



Columbia  
University

CLUE LAB and  
Prof. Ioannis Kymissis



University of  
Kentucky

Prof. John Anthony



Elettra Sincrotrone Trieste

ELETTRA Sincrotrone, Trieste

Dr. Giuliana Tromba  
Dr. Diego Dreossi



EXCELENCIA  
SEVERO  
OCHOA

ICMAB-CSIC

Dr. Marta Mas-Torrent



*Flexible organic Ionizing Radiation  
dEtectors*

INFN (Italian Institute for  
Nuclear Physics)  
(2019-2022)



*RivELatOri innovativi per cure ADroterapiche*

cdp  
(2024-2025)

Fondazione  
CARIPLO  
TUTE SERVARE MUNIFICE DONARE • 1816



# BACKUP SLIDES



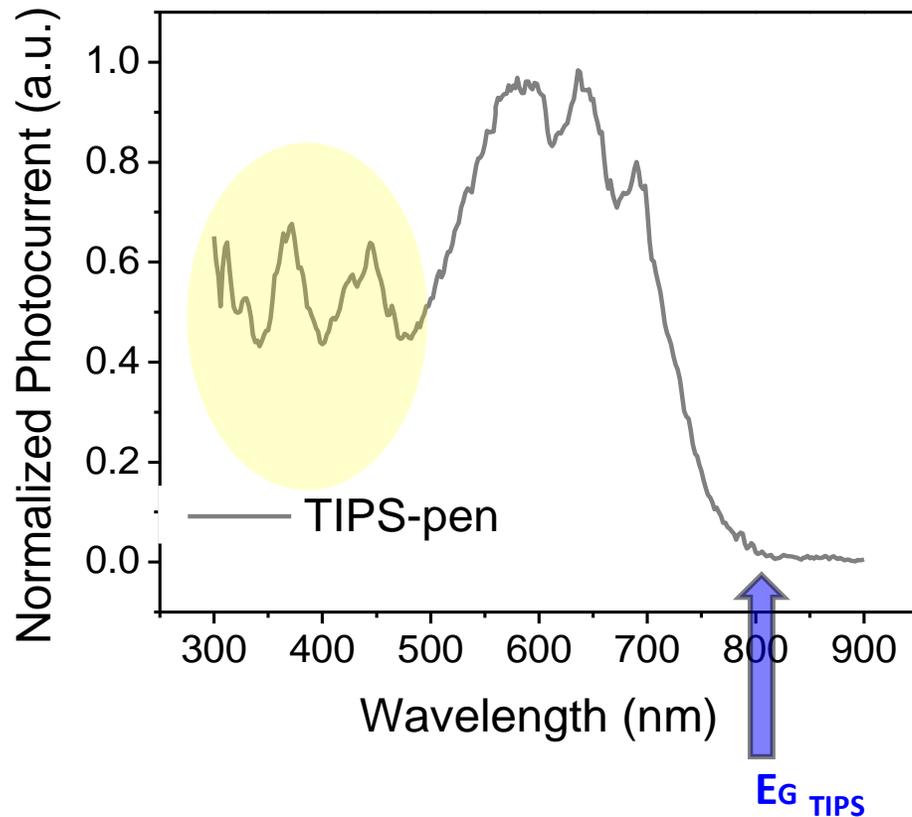
# ROLE OF PHOTOACTIVE TRAPS STATES:

## Photocurrent Spectroscopy Optical Quenching

→ experimentally assess and identify the trap states which activate the photoconductive gain effect in organic thin film based devices

*Fratelli et al., Adv. Mater. Technol. 2023, 2200769*

→ Simultaneous irradiation with **X-rays** (W-target X-150 kVp) and **visible photons**



*I. Kymissis et al., IEEE Trans. Electron Devices 57, 380–384 (2010).*

→ **electron traps** in organic transistors, enhance the photoconductivity for photons in the range [350 – 480] nm



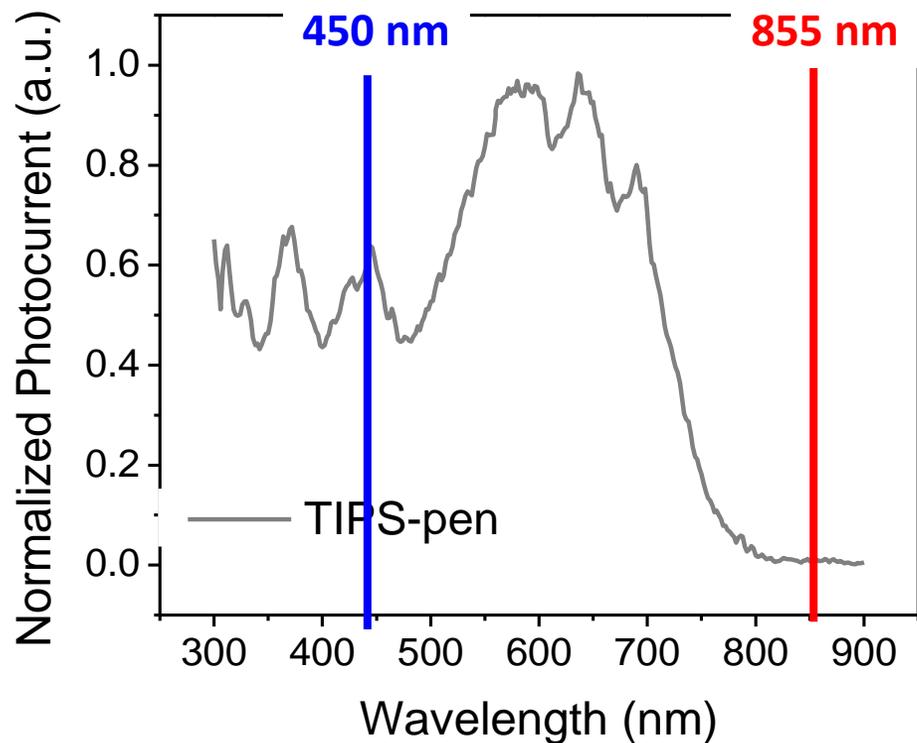
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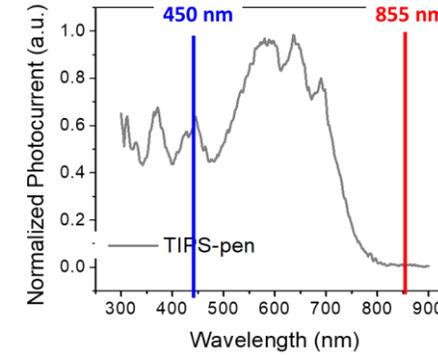
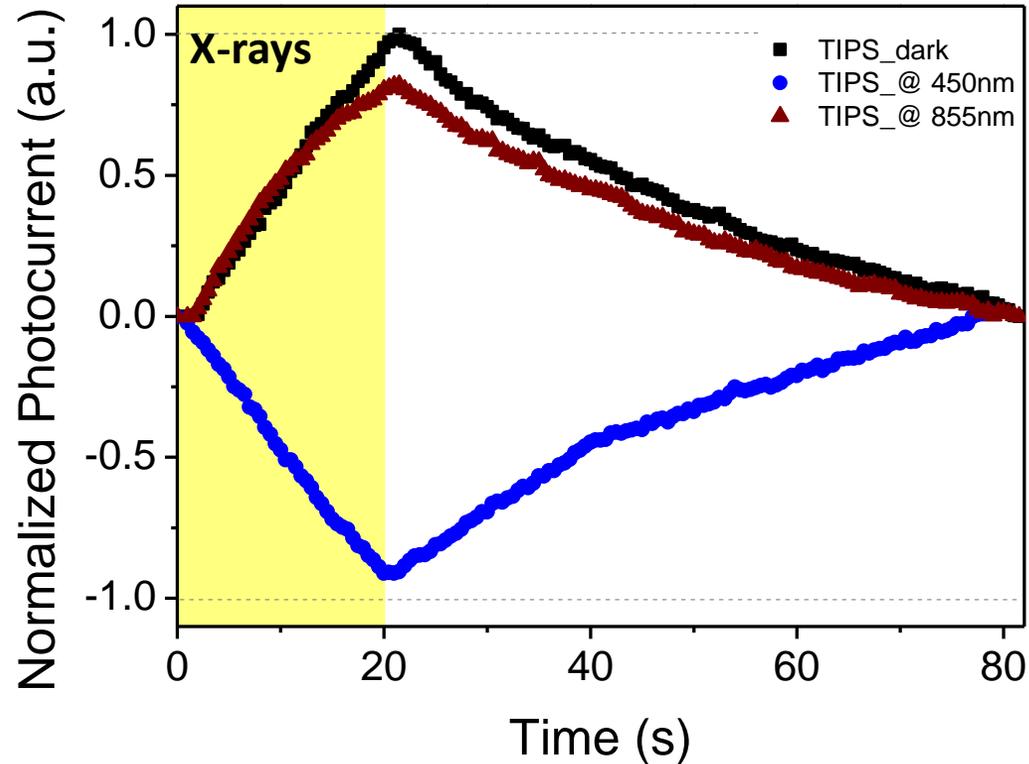
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LED (450 nm and 855 nm) have been selected because they correspond to two different and crucial position in the Photocurrent spectrum.



# ROLE OF PHOTOACTIVE TRAPS STATES:

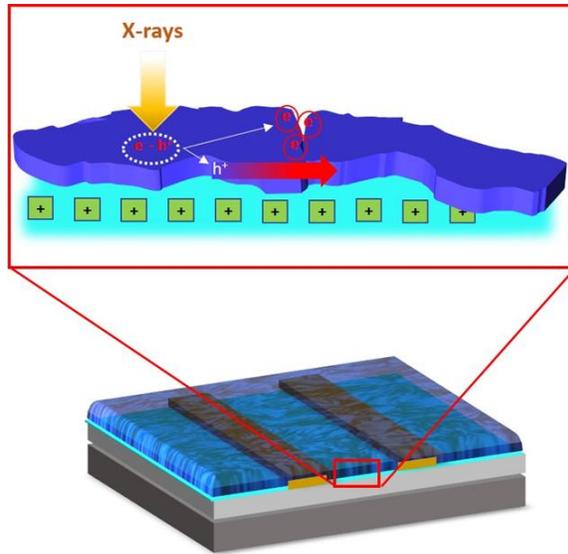
## Photocurrent Spectroscopy Optical Quenching



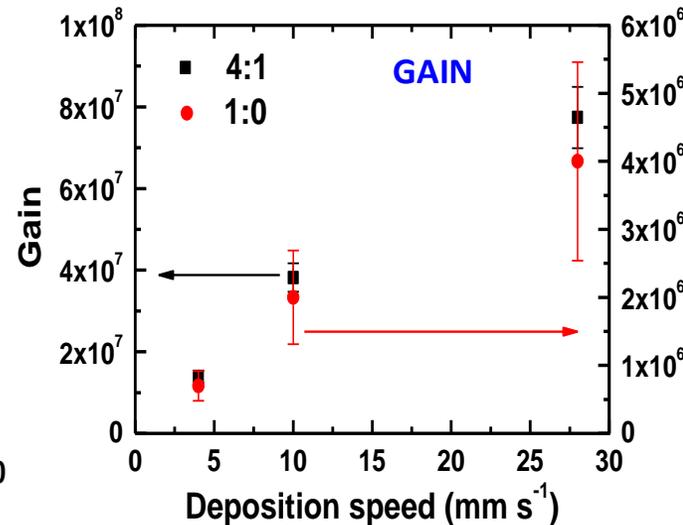
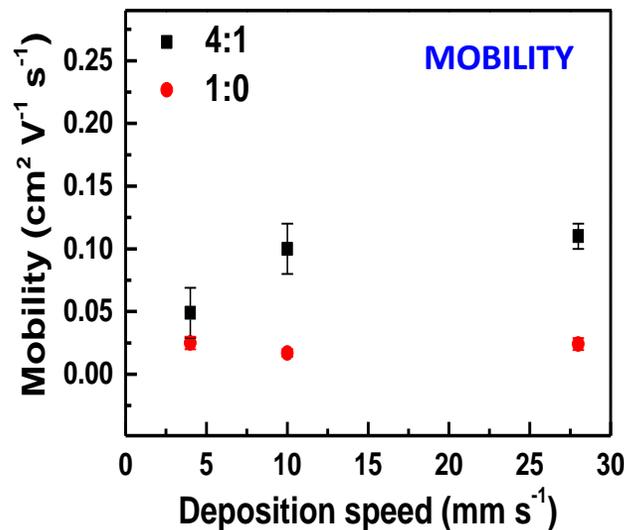
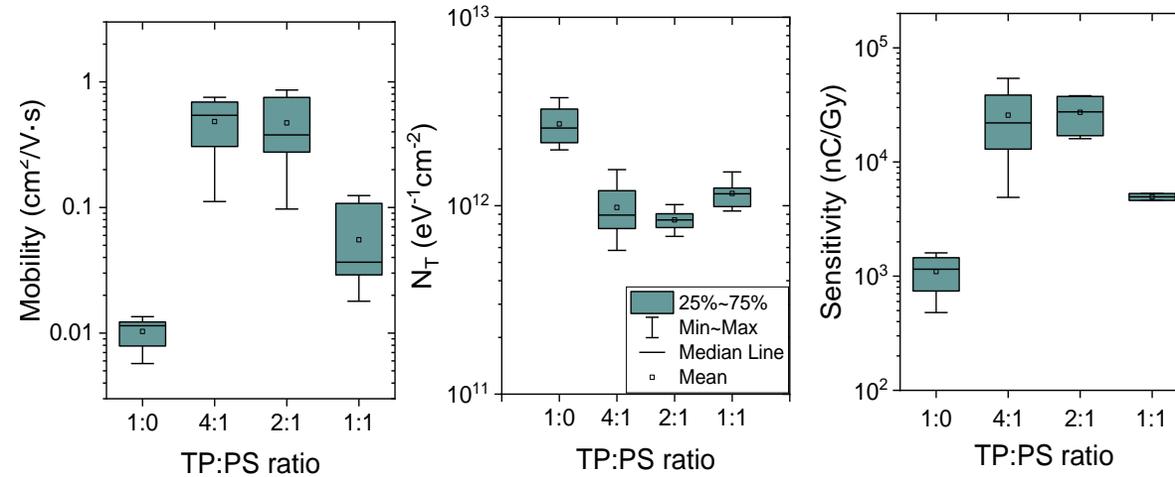
**855 nm** light (below bandgap) have no effect on X-ray response.

**450 nm** light fills and saturates e- traps that become inactive → completely **quench the PC gain** X-ray induced signal → decrease of current → X-rays facilitate a recombination between the electron already trapped and the hole already present/generated.

# CONTROL OF ELECTRICALLY ACTIVE DEFECTS: INTERFACE STATES



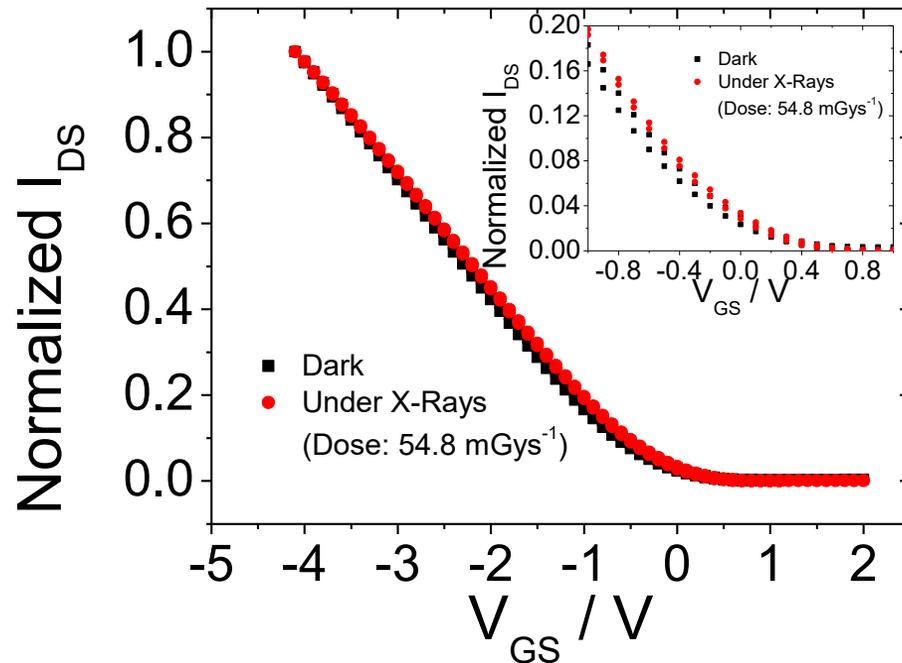
**BLENDS TIPS:Polystyrene** passivates the interface state with the dielectric  $\rightarrow \gg$  *hole mobility*



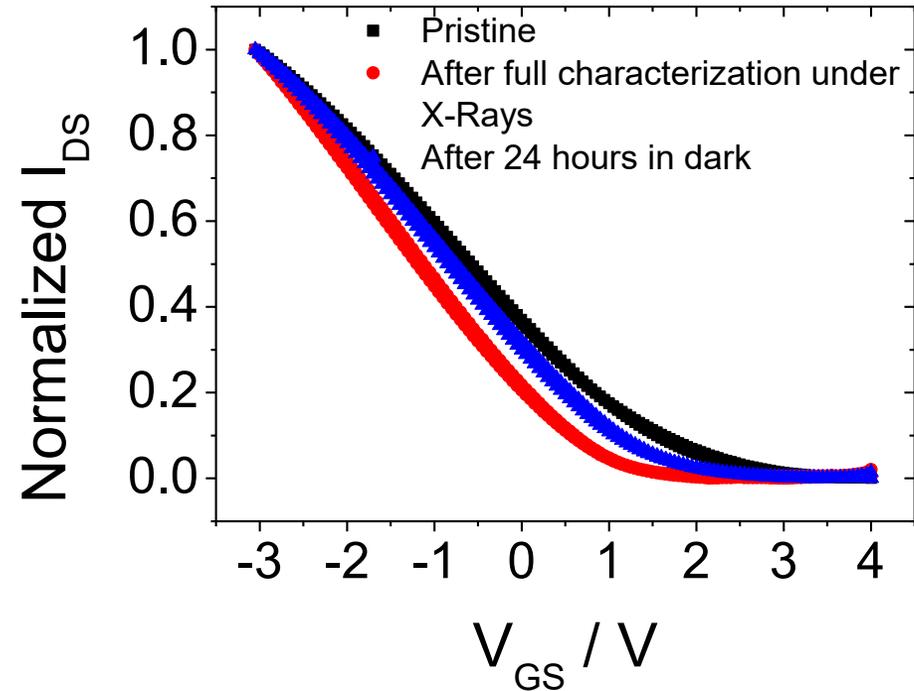
similar mobility values, **BUT** the photocurrent gain follows the grain size variation with deposition speed (e.g. *electron/interface traps*).



# Radiation Hardness (TIPS-pentacene)



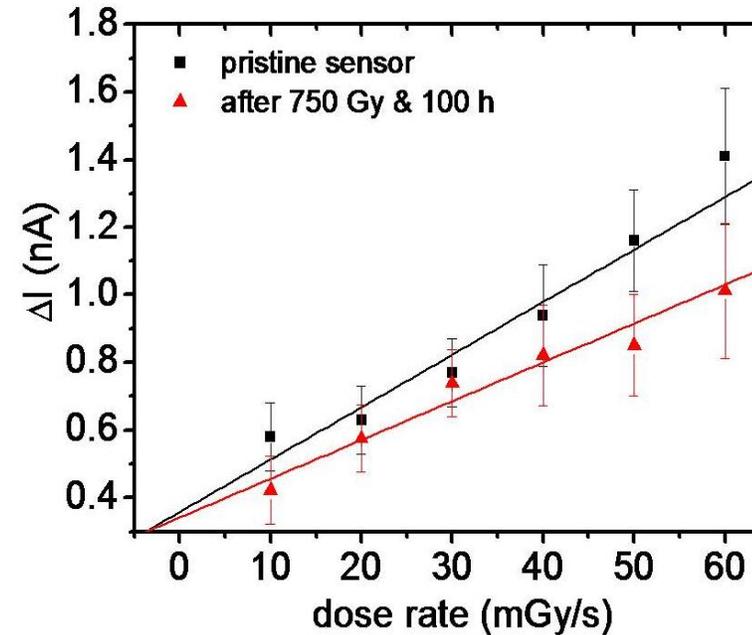
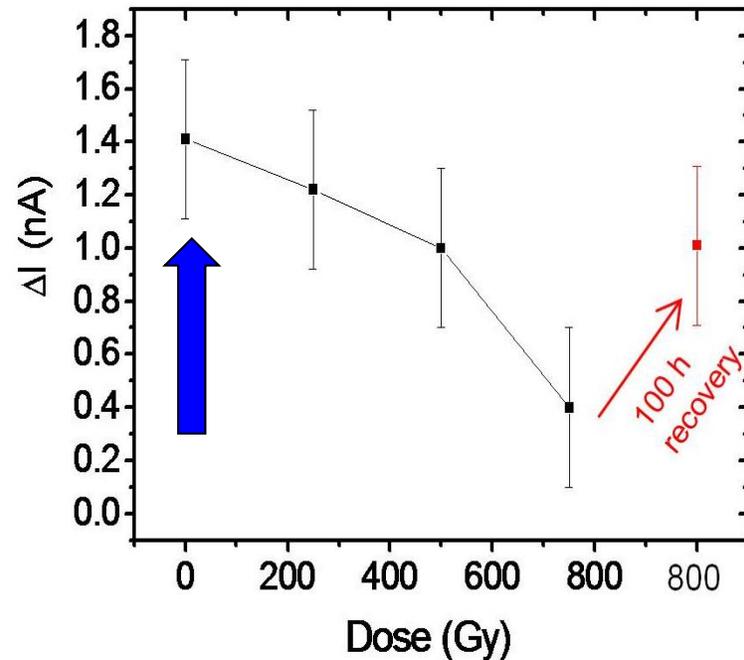
transfer characteristic curves :  
before (black squares) and during (red circles) X-ray exposure at a dose rate of 54.8 mGy s<sup>-1</sup>



transfer characteristic in pristine state (black squares), after X-ray exposure with a total dose of 160 Gy (red dots) and after 24 h kept in dark (blue stars).



# Radiation hardness under X-rays: organic thin film (TIPS- pentacene) - II

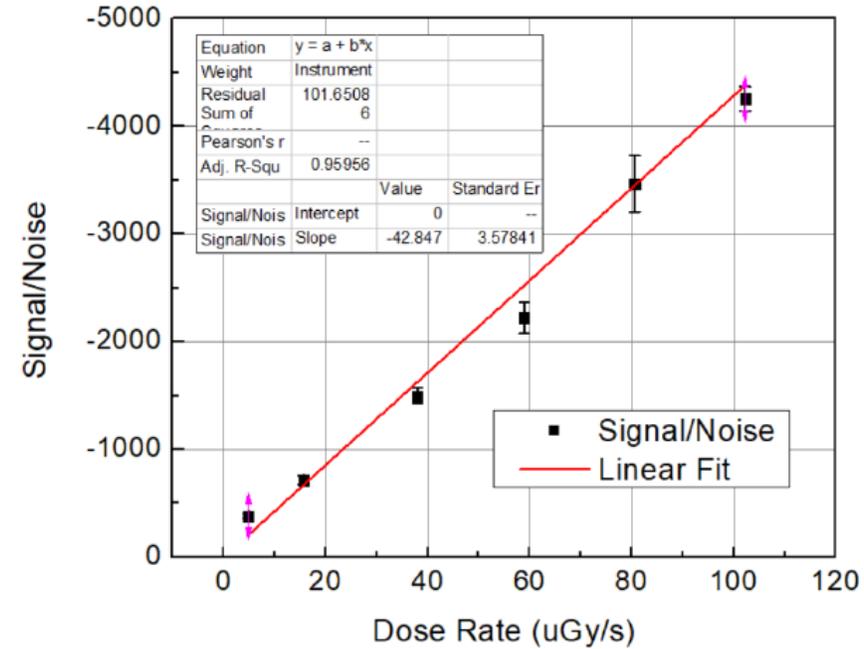
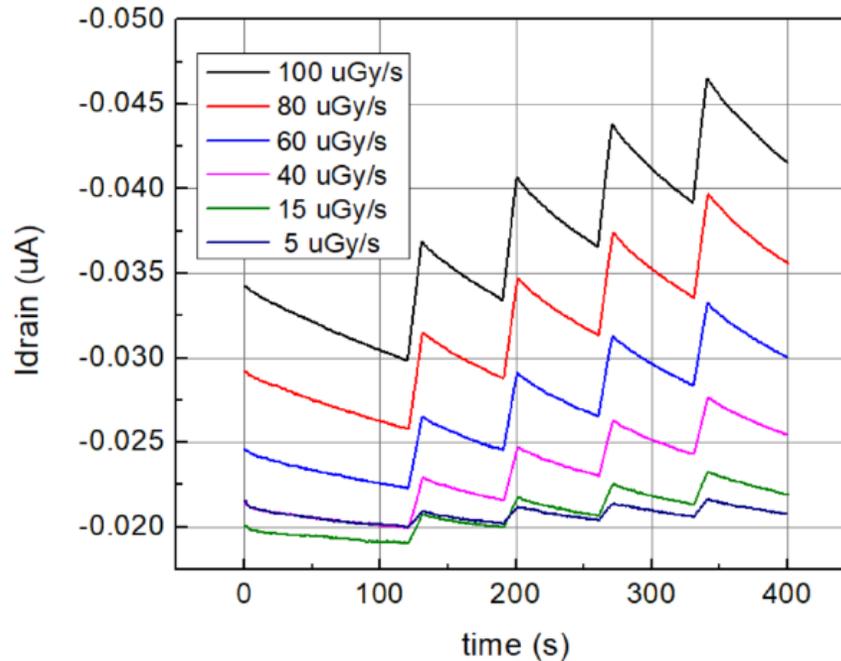


**Four steps of 200Gy X-ray** irradiation (35KV Mo tube). **Total dose 800Gy**  
Total irradiation dose for medical diagnostic detectors: 5-10 Gy/year

Recovery allowed only after the last step (100h in the dark)



# Limit of Detection



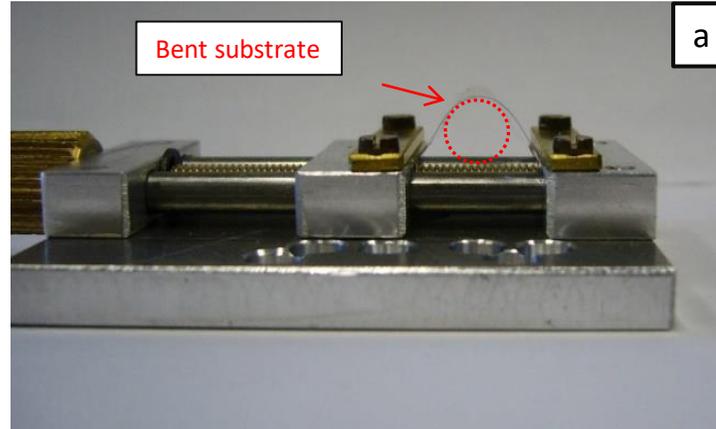
Measured LoD:  $5 \mu\text{Gy s}^{-1}$

Extracted:  $0.8 \mu\text{Gy s}^{-1}$

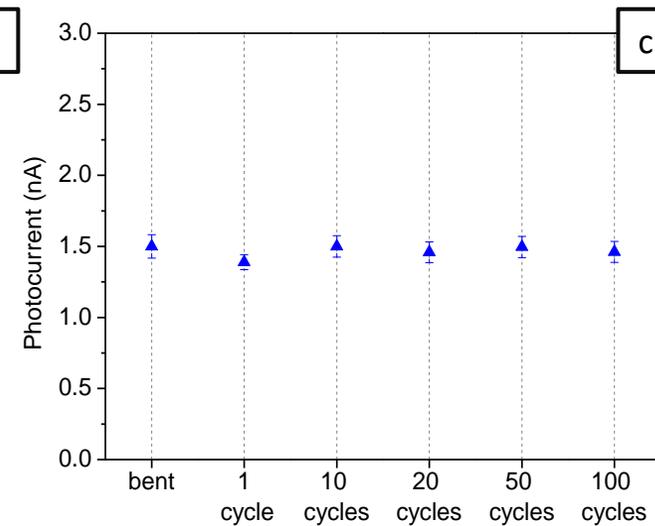
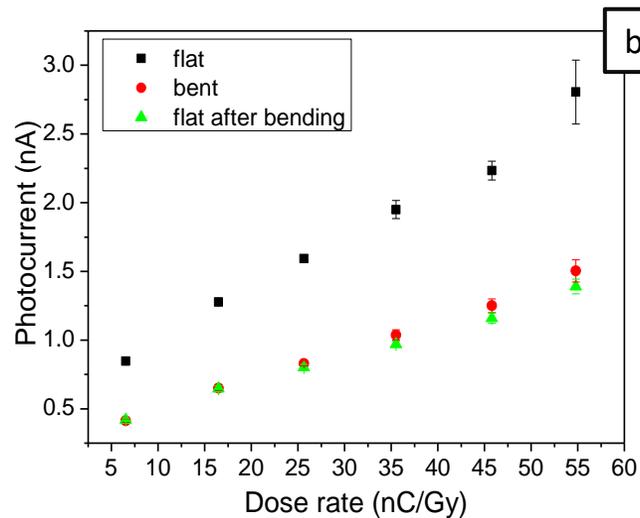
Typical dose rate values presently used in medical diagnostics:  $5.5 \mu\text{Gy s}^{-1}$

standard radiographic examinations have average effective total doses in the range: 0.005–10 mGy

# Mechanical flexibility



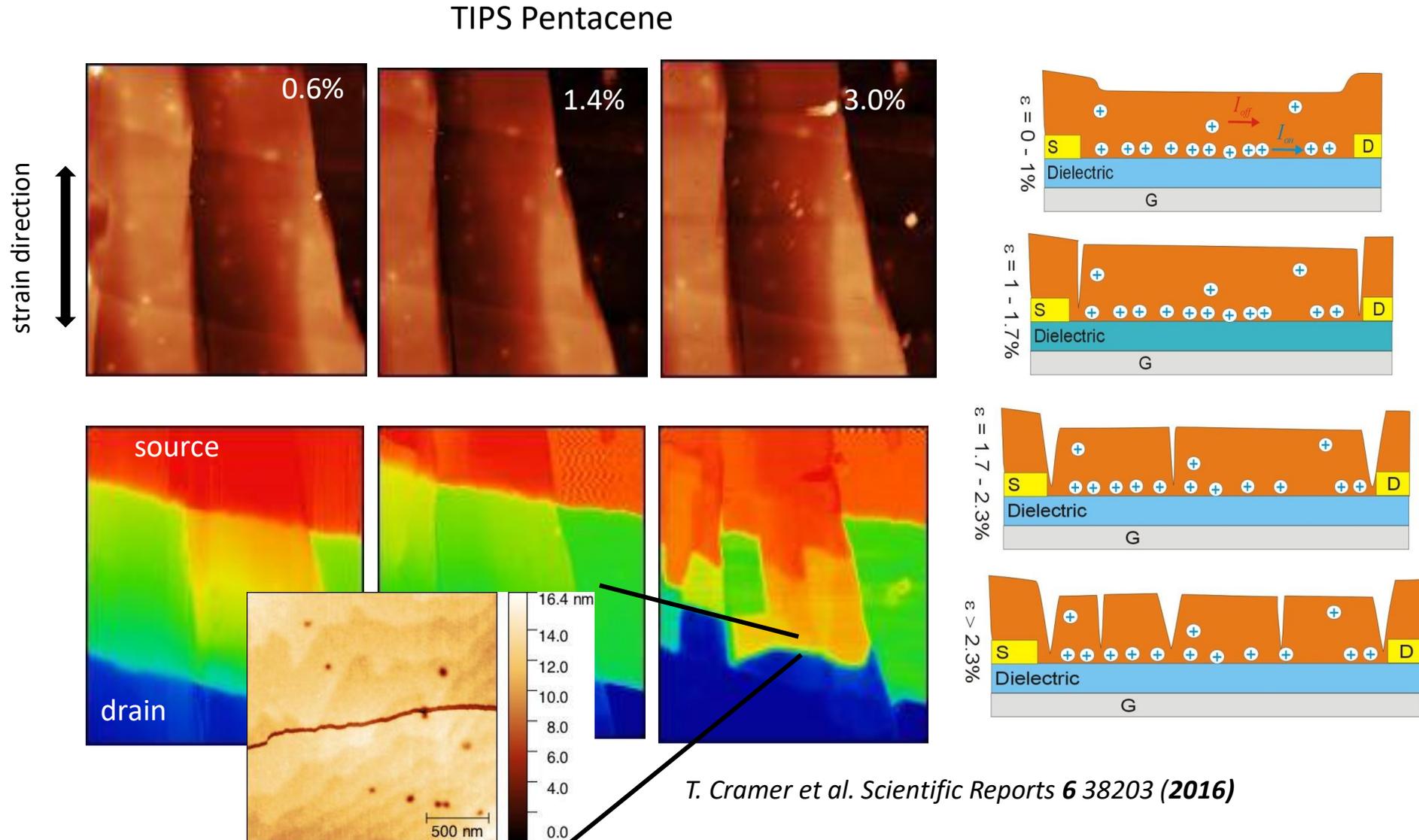
**bending radius of 0.3 cm**  
→ Conformable to human body



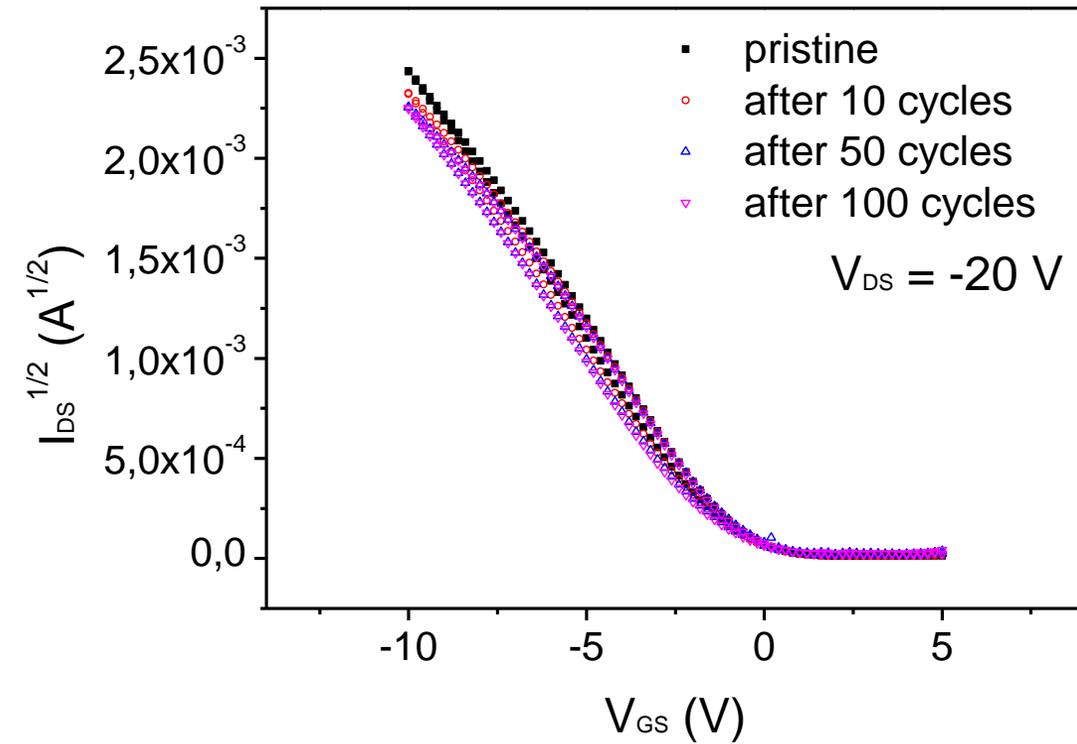
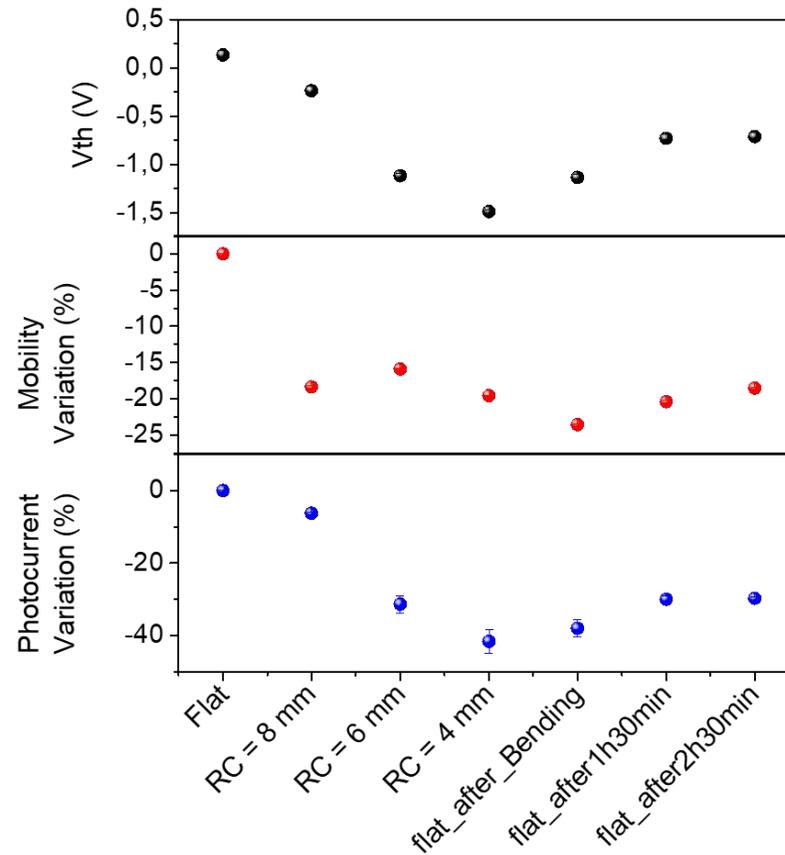
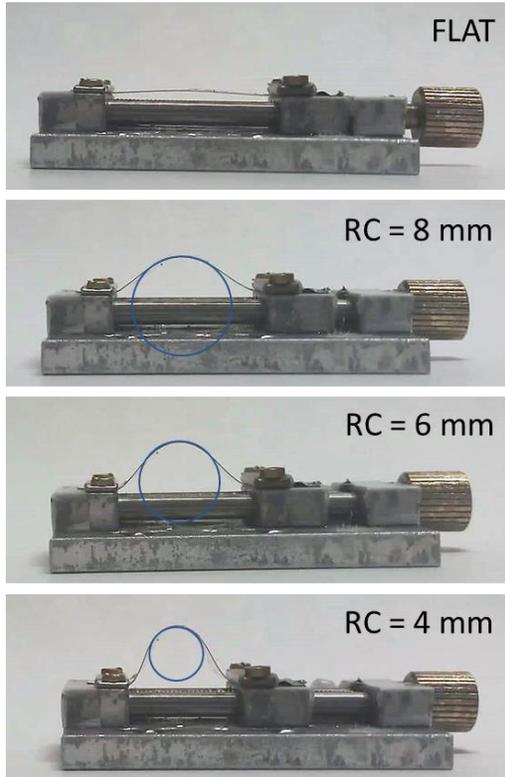
- Decrease of about 50% after first bending
- Stability over 100 bending cycles.



# KPFM on strained transistors: nanocrack formation



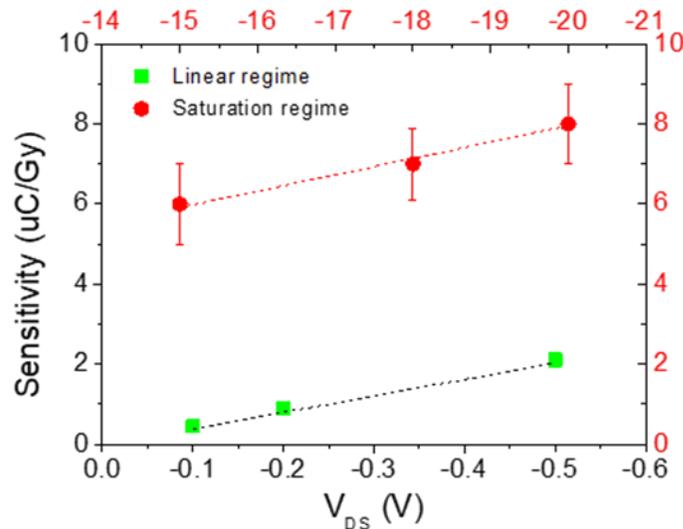
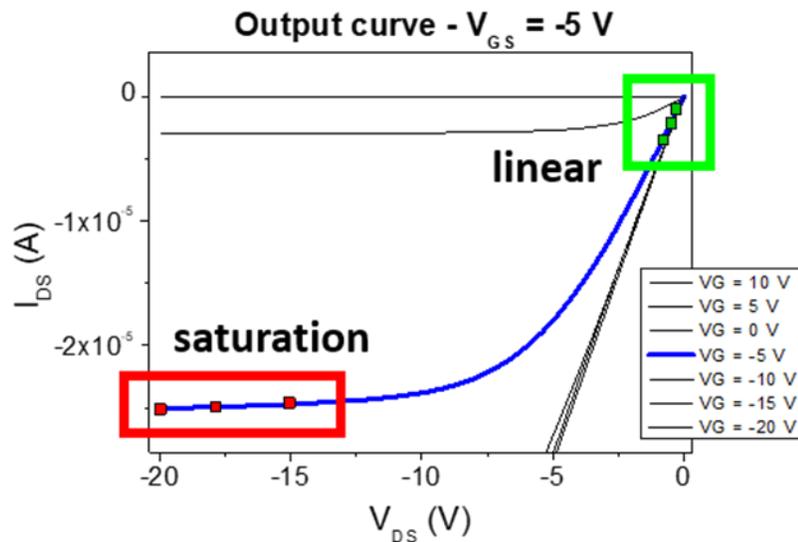
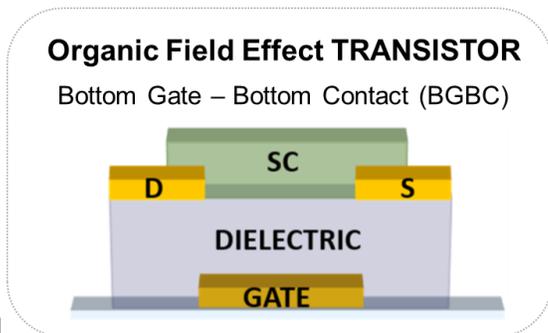
# Mechanical flexibility



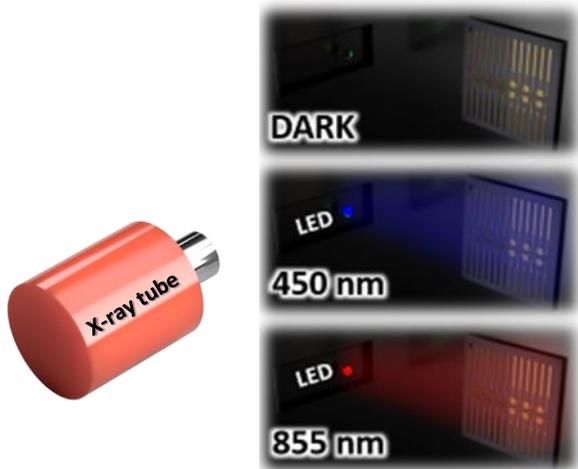
# Printed organic detector architectures

## OFETs advantages

- ✓ Multiparametric device
- ✓  $V_{GS}$  → switch ON/OFF, allow to address the pixel
- ✓  $V_{DS}$  → tuning of the sensitivity



# Role of photoactive traps states



## Quenching of photoconductive gain by visible light

LED (450, 593 and 855 nm) have been selected because they correspond to three different and crucial position in the Photocurrent spectrum.

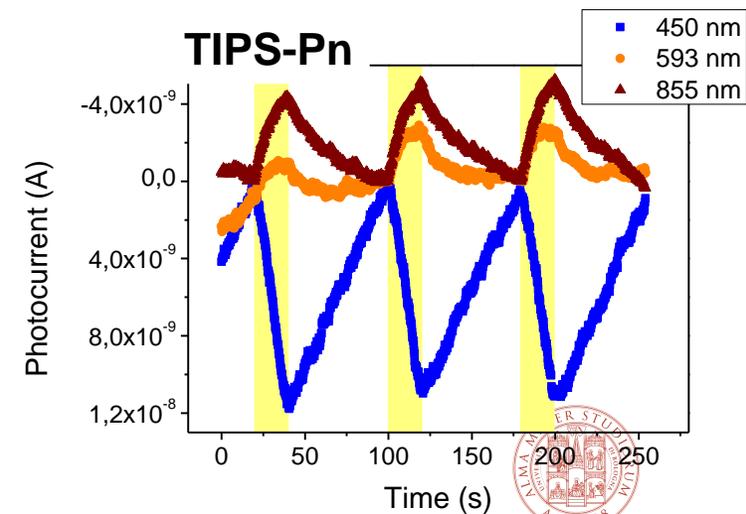
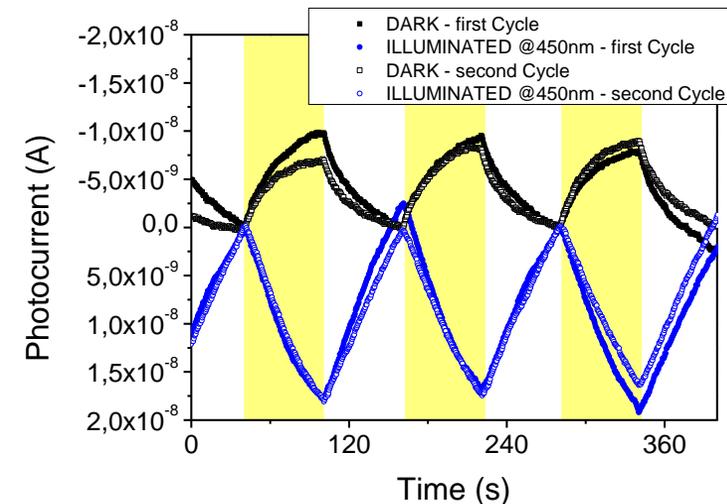
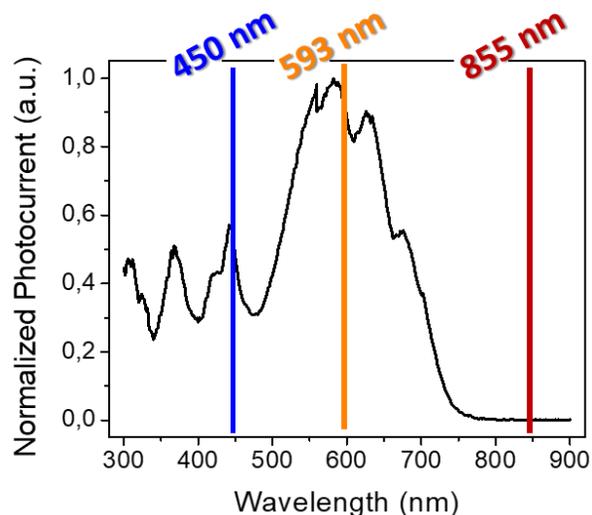
**855 nm** light (below bandgap) have no effect on X-ray response.

**590 nm** light only partially reduce the X-ray response (absorption of visible photons above band gap, correlated to a large conductivity increase and a decrease in the X-ray signal-to-noise ratio)

**450 nm** light completely quench the X-ray induced signal → decrease of current → X-rays facilitate a recombination between the electron already trapped and the hole already present/generated.

I. Kymissis et al., *IEEE Trans. Electron Devices* **57**, 380–384 (2010).

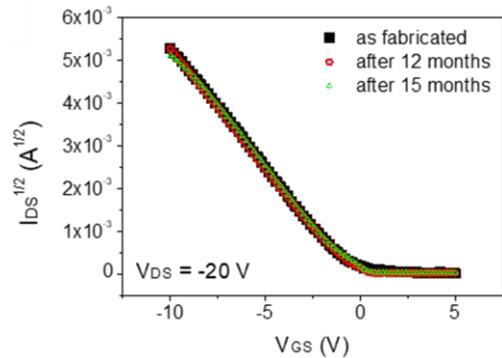
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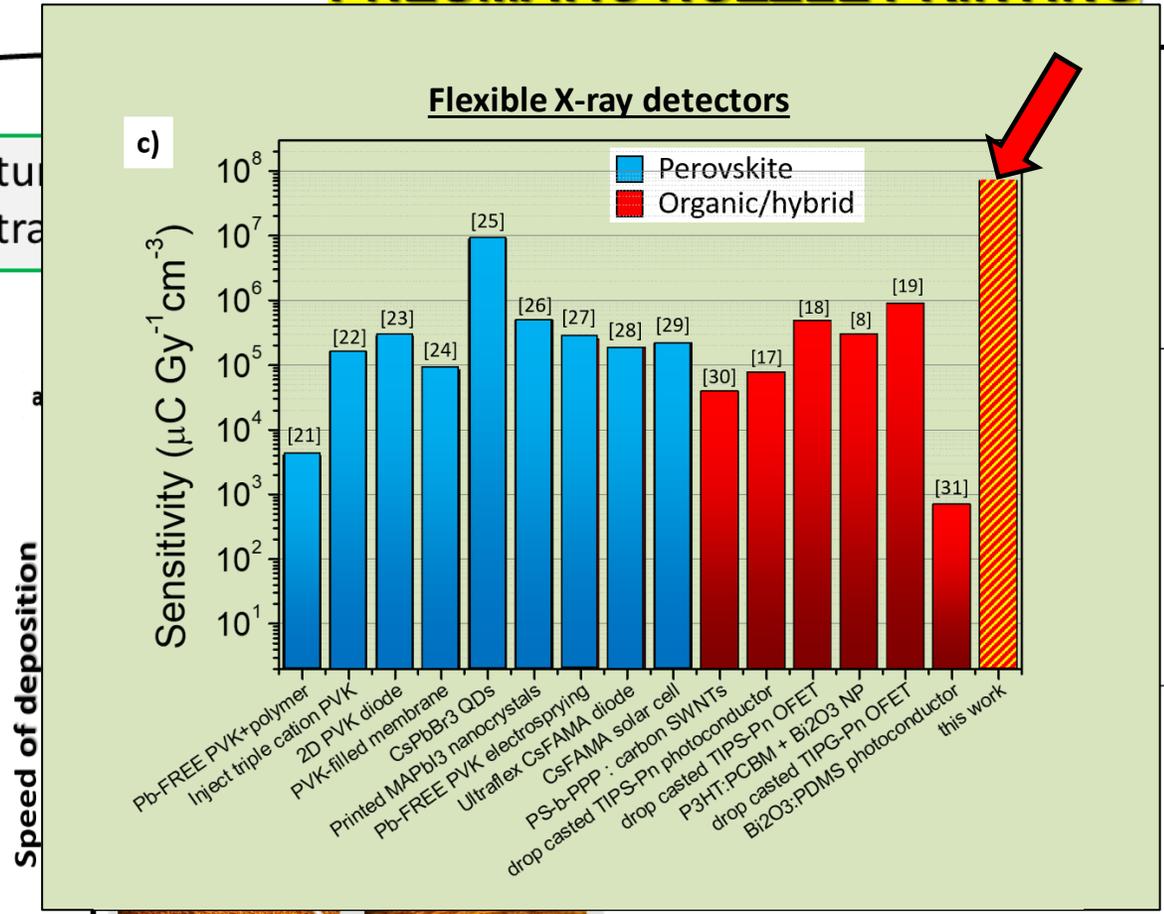
# CONTROL STRATEGIES OF DEVICE PARAMETERS

## PNEUMATIC NOZZLE PRINTING

Transistor architecture onto **FLEXIBLE** substrates



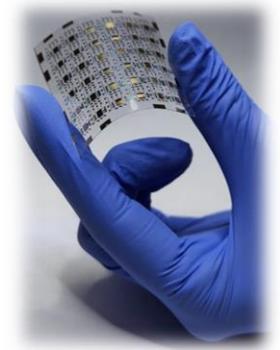
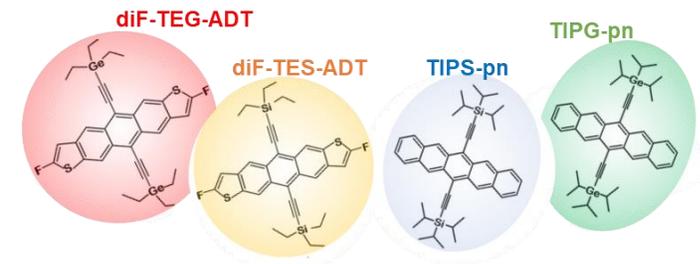
c)



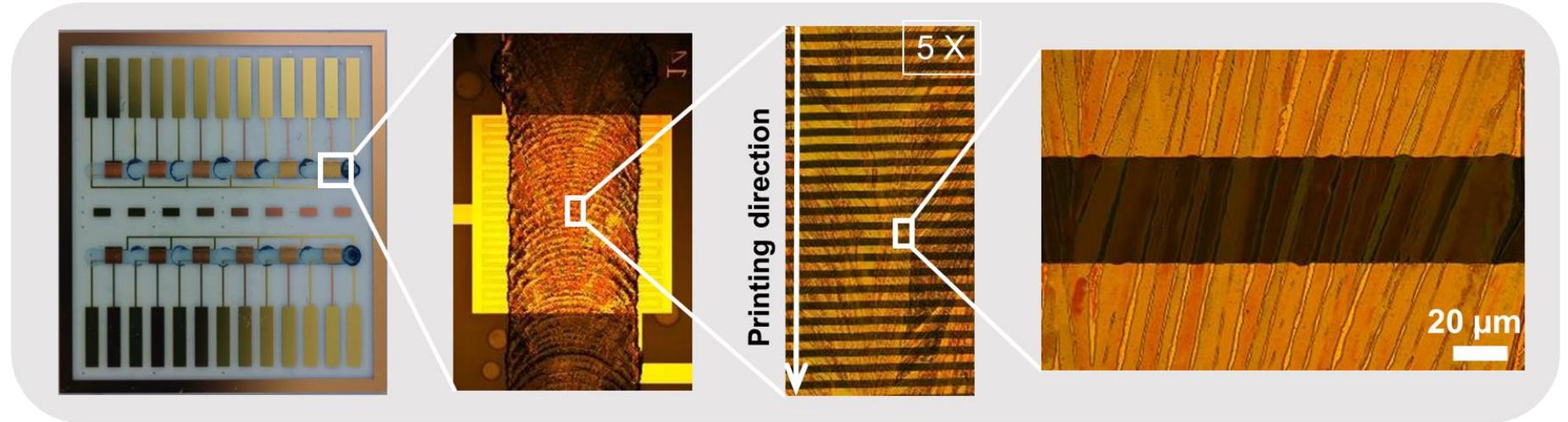
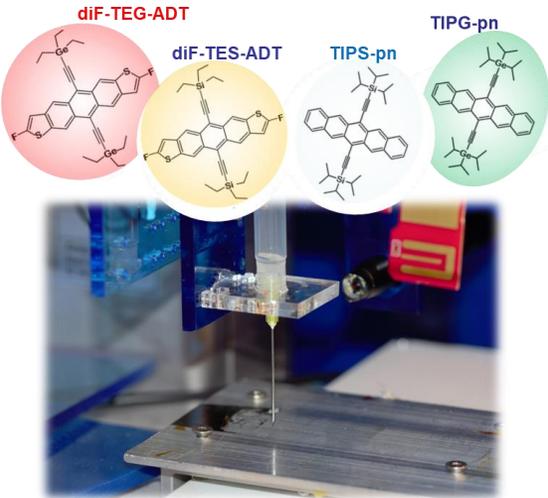
Speed of deposition



Enhancing the absorption rate by **Chemical tailoring**



# DEPOSITION TECHNIQUE: PNEUMATIC NOZZLE PRINTING

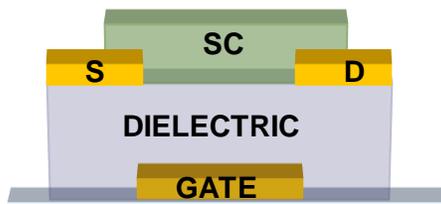


**Microcrystalline structure**  
**Very packed films**

**Thickness** = [100-200] nm

**Width** = varies depending on the deposition parameters

**Bottom Gate – Bottom Contacts OFETs**



## PNEUMATIC NOZZLE PRINTING

Transistor architecture onto **FLEXIBLE** substrate

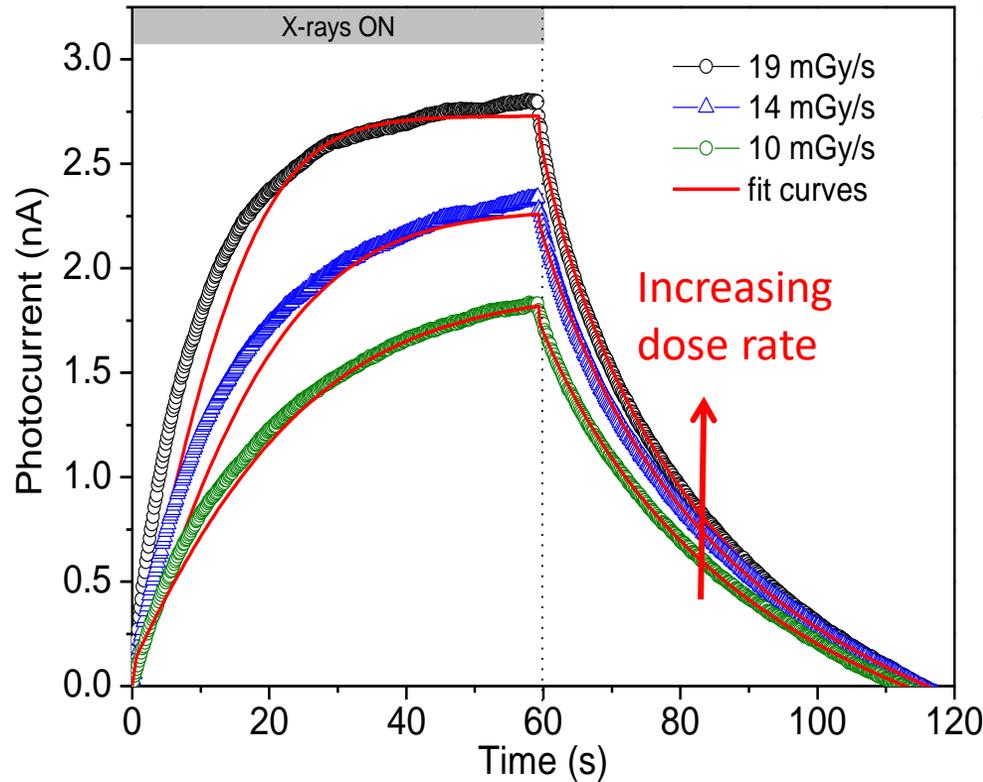
Full control of the **MORPHOLOGY**

Improvement of the **Transport properties**

Enhancing the absorption rate by **Chemical tailoring**



# Photoconductive Gain effect



Good agreement with the saw-tooth shape experimental data, using a single set of fitting parameters

**G = photoconductive gain**

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

**Transit time**

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

**Carrier lifetime**

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

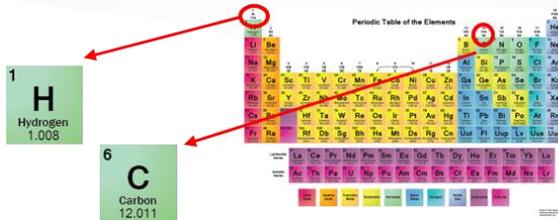
## Dynamics of X-ray response

Device:  $W = 48 \text{ mm}$ ,  $L = 30 \text{ }\mu\text{m}$ , bias 0.2 V Synchrotron 17 keV X-ray beam



# ORGANIC SEMICONDUCTORS FOR IONIZING RADIATION DETECTION - ADVANTAGES

Chemical composition formed by  
**LOW-Z ELEMENTS**



✓ **HUMAN TISSUE EQUIVALENCE**

✓ Medical Applications

✗ **POOR ABSORPTION**

**EXTERNAL QUANTUM EFFICIENCY**  $\approx [0.01 - 0.001] \%$

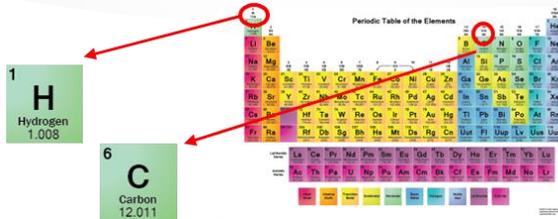
✓ High Radiation Hardness

✓ **But still high Sensitivity!** Why?



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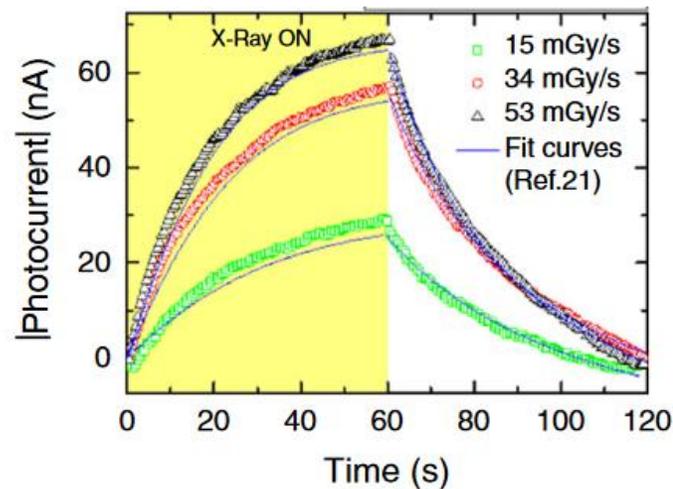
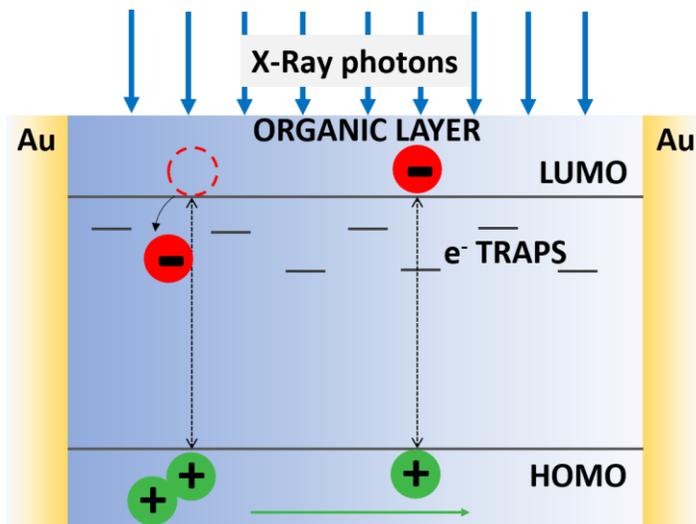
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✓ High Radiation Hardness

✓ **But still high Sensitivity!** Why?

## PHOTOCONDUCTIVE GAIN EFFECT



**VERY HIGH**

**INTERNAL QUANTUM EFFICIENCY**

**Carriers lifetime**

$$G = \frac{\tau_r}{\tau_t}$$

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

**Transit time**

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

COLLECTION EFFICIENCY

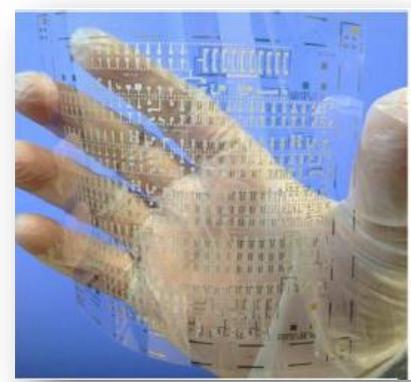
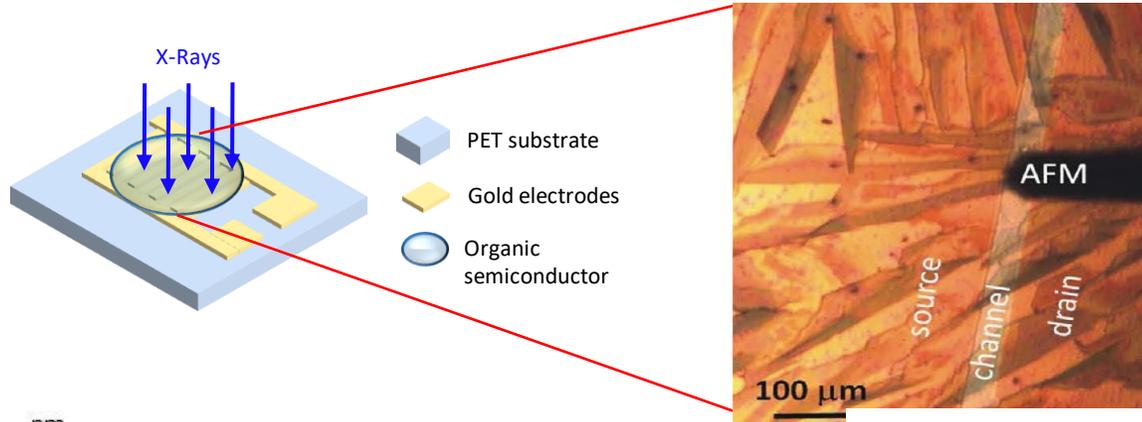
GAIN up to  $10^7$

L. Basiricò et al., Nat. Commun., vol. 7, 13063, 2016

I. Temiño, L. Basiricò et al., Nat. Commun., vol. 11, 2136, 2020



# FLEXIBLE ORGANIC RADIATION SENSORS



**Sensitivity:  $7 \times 10^6 \mu\text{C}/\text{Gy cm}^2 @ 0.2\text{V}$**   
 >> than polyCZT or a-Se

Room temperature

Linear response (**dosimetry**)

