Boosting direct X-ray detection in organic thin films by small molecules tailoring

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[2]*National Institute for Nuclear Physics – INFN section of Bologna, Italy*
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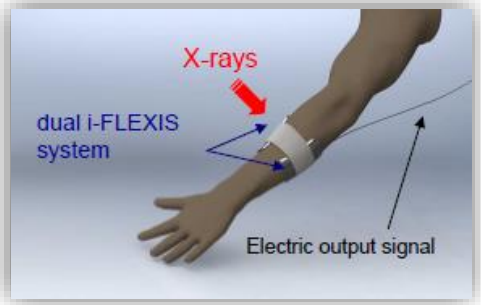
Motivation:
Flexible large area X-rays detectors

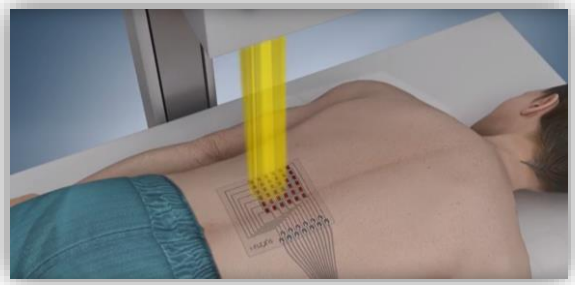
New generation of
low cost, **low power supply**
and **mechanical flexibility**
Thin and **comformable**
sensor panels and patches

Motivation:
Flexible large area X-rays detectors

Wearable health diagnostic applications



Personal dosimetry Medical application



Cultural Heritage

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

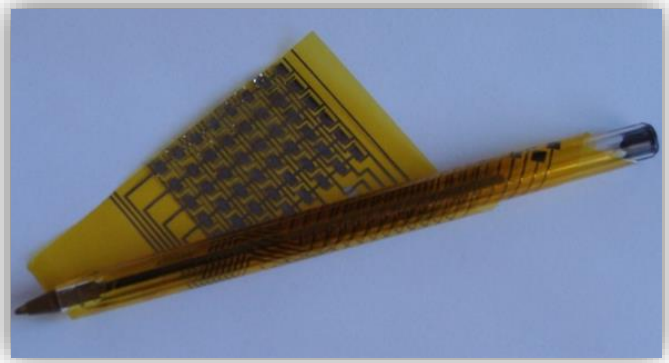
Citizens security: "smart walls/pillars"

Airport security

Defense

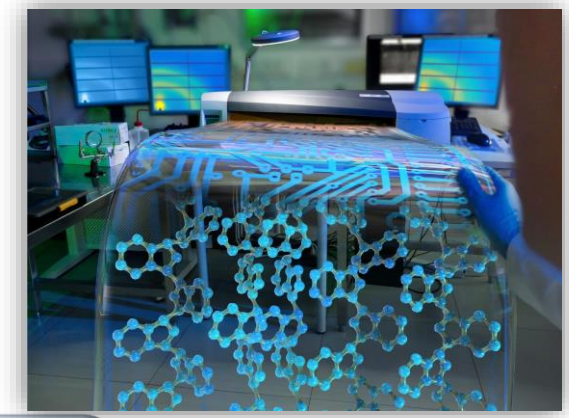
Why Organic Materials?

Light-weight and flexible



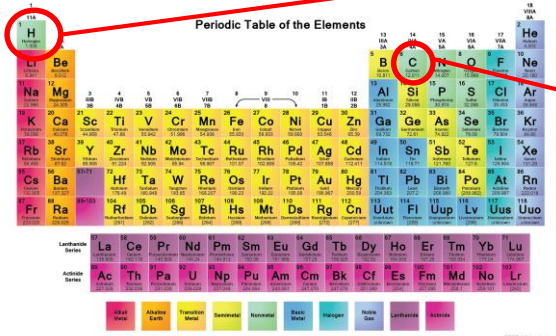
Wearable

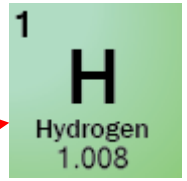
Low-cost printing techniques

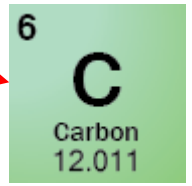


Easy chemical tailoring

Low-Z→ tissue-like



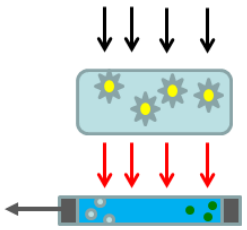




Organic X-Ray detectors

INDIRECT DETECTION

X ray beam

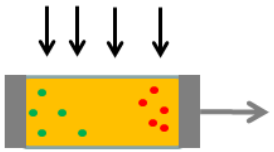


Scintillators
(X - to VIS)

Photodiode
(VIS to electrical
charge carriers)

DIRECT DETECTION

X ray beam



X photons to electrical
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Organic X-Ray detectors

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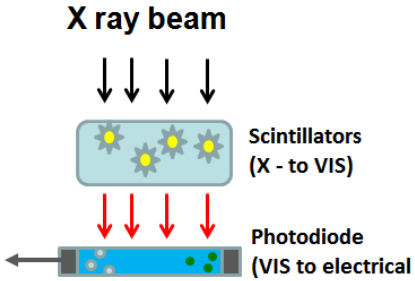
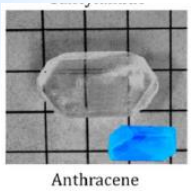
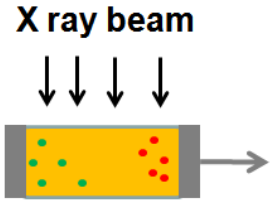


Hull et al., IEEE Trans. on Nucl. Sci.56, 3, (2009)


Anthracene

DIRECT DETECTION



Organic X-Ray detectors

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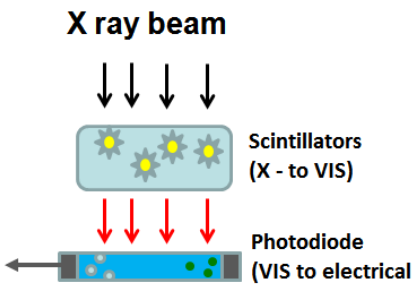

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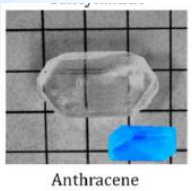
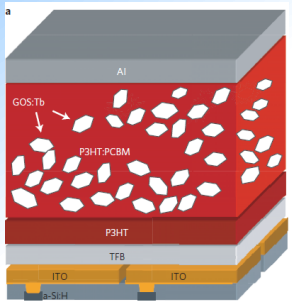


P. Buchele, Nature Photonics,9 (2015)



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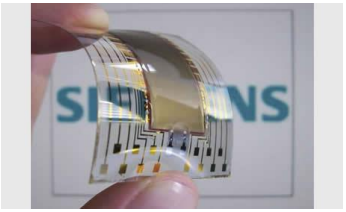

http://www.siemens.com/innovation/en/news/2013/e_inno_1305_1.htm

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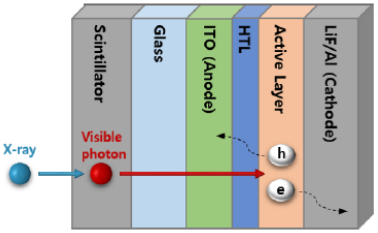

Seon et al. DOI 10.1109/TNS.2016.2645228, IEEE Trans. Nuc. Sci. (2017)

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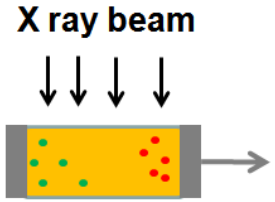

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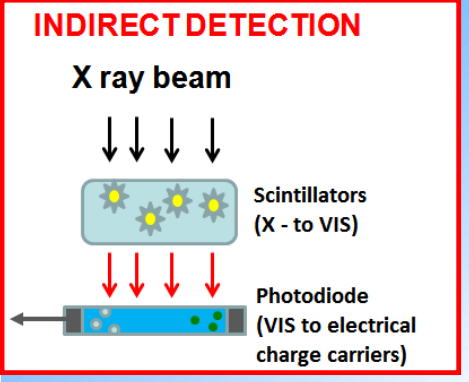

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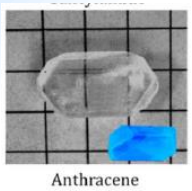

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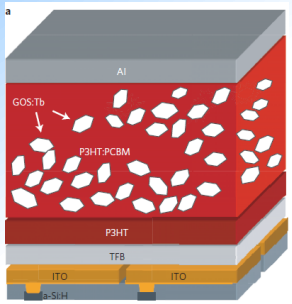

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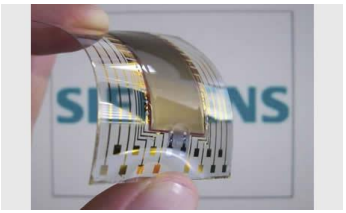

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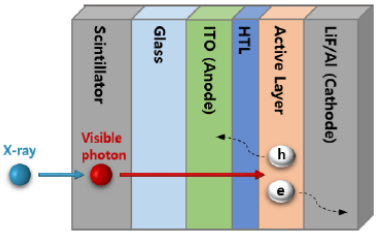

Seon et al. DOI 10.1109/TNS.2016.2645228, IEEE Trans. Nuc. Sci. (2017)

Organic single crystals

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

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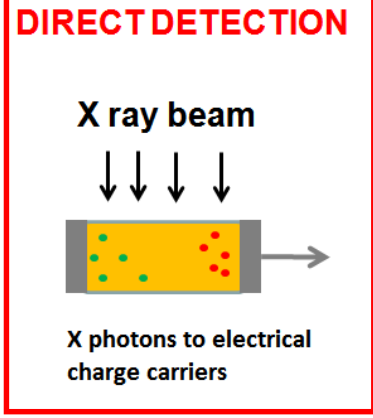

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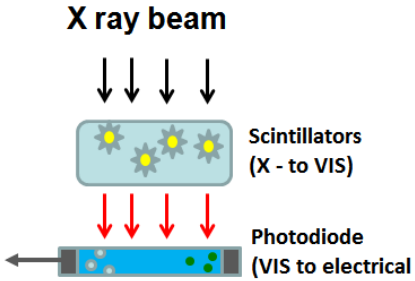

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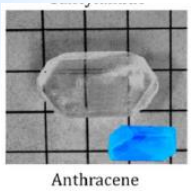

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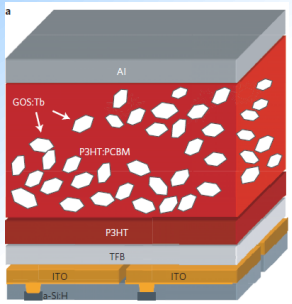

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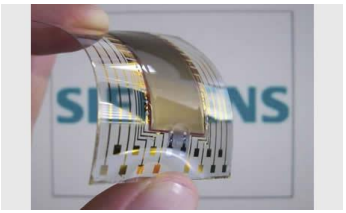

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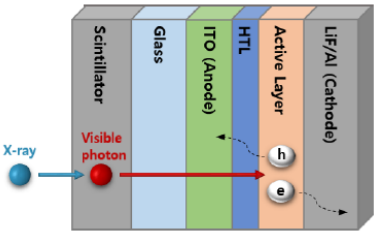

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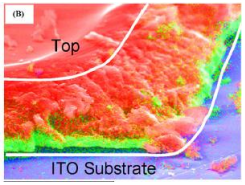

PTAA/Bi_2O_3 NPs

Intaniwet, et al., Journ. App. Phys., 106, (2009)
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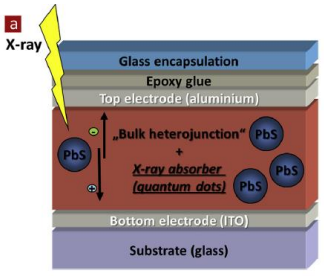
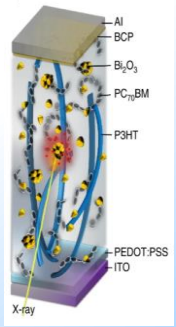
Sensitivity up to 480 nC/mGy/cm^3 @50-100V

P3HT:PCBM:PbS QDs BHJ

Ankah et al. Organic Electronics 33 (2016)





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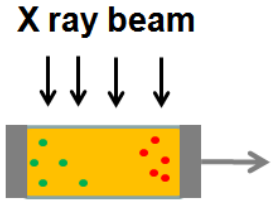

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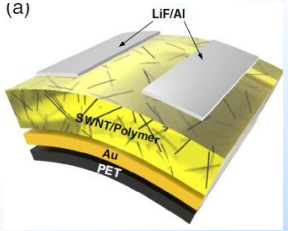

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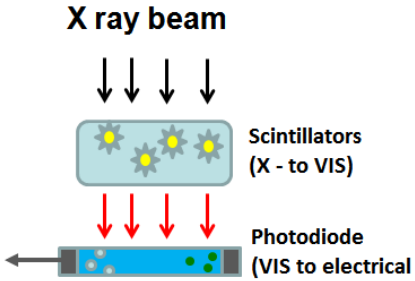

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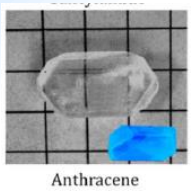

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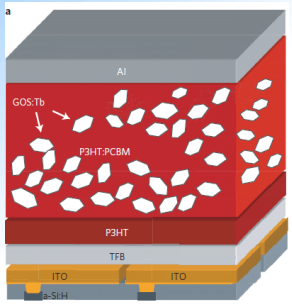

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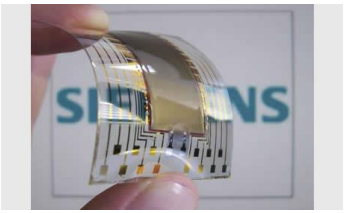

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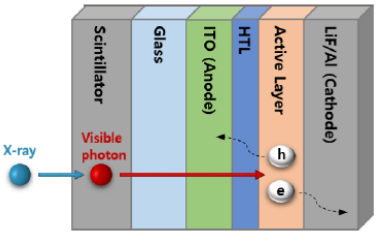

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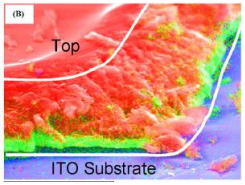

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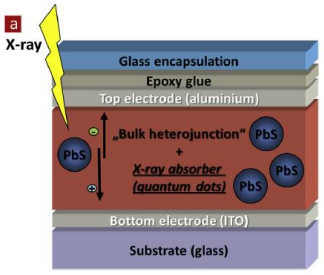

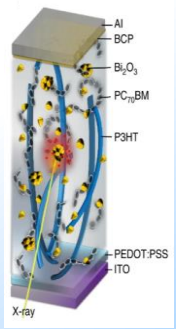

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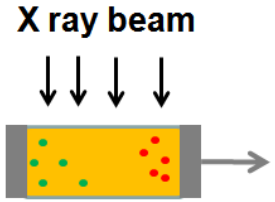

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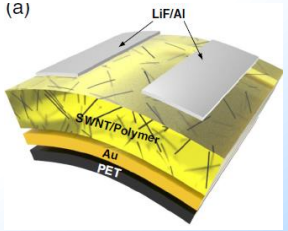

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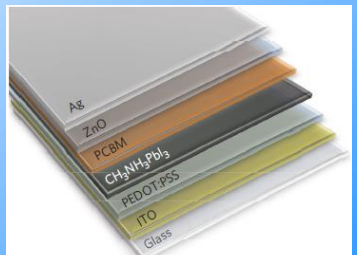
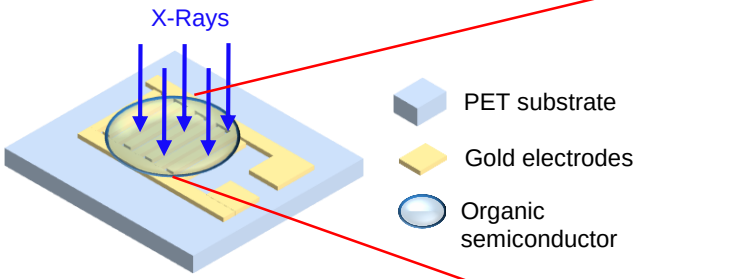
Solution-processed **lead halide perovskites**

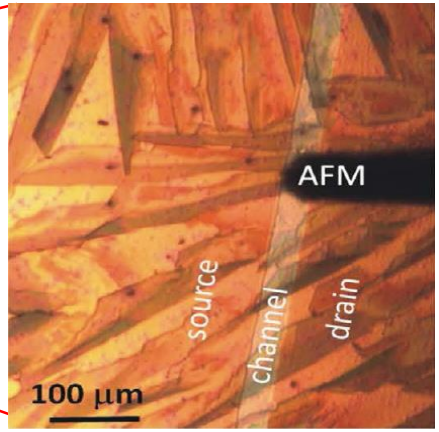
S. Yakunin et al, Nature Photonics, 9 (2015)

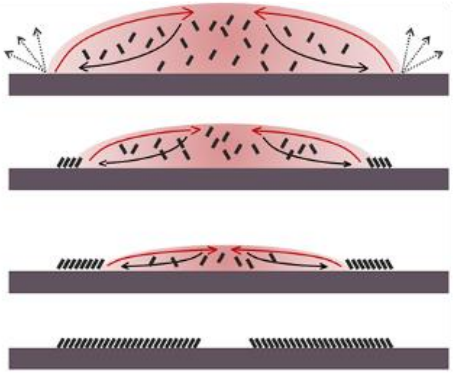
Basiricò et.al. Adv.Func.Mat. 2019

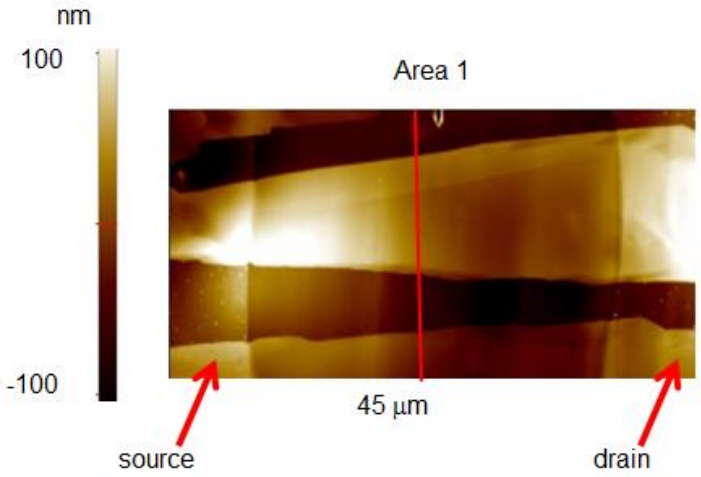


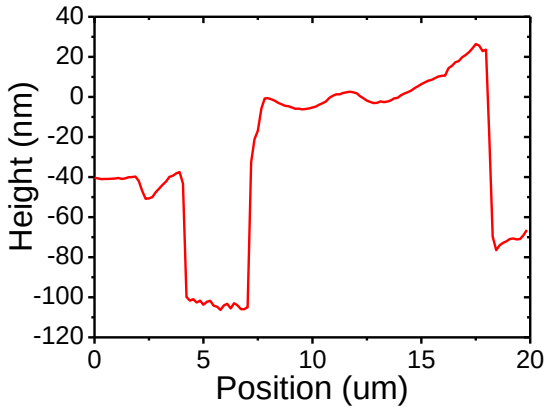
Flexible direct X-ray detectors based on Organic thin films

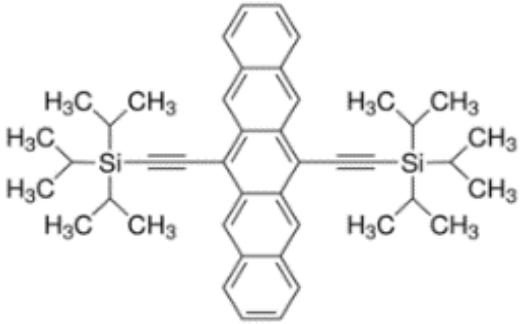








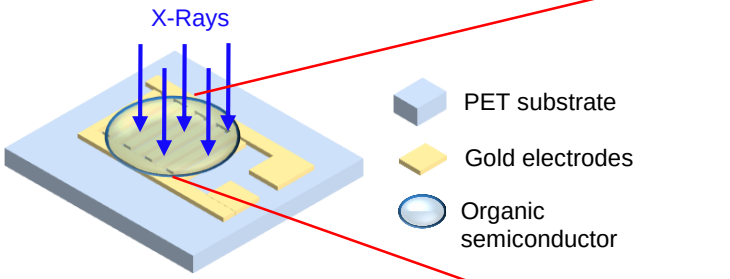


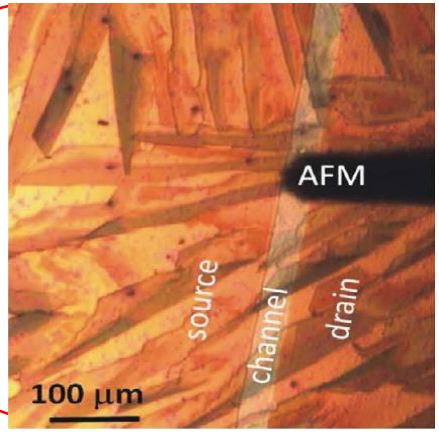


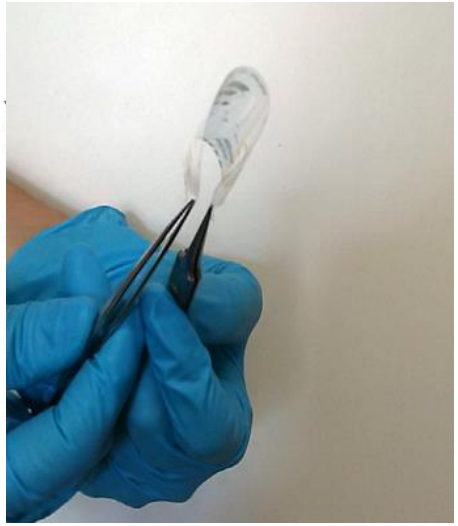
TIPS Pentacene

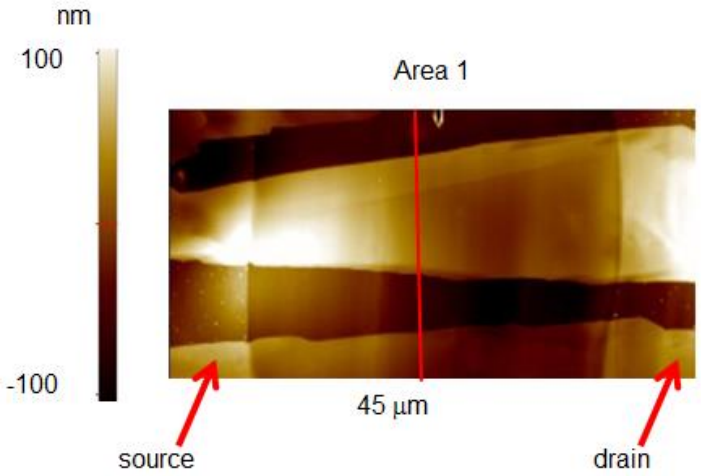
Single crystals (100 microns long and 100nm thick) bridge source and drain electrodes

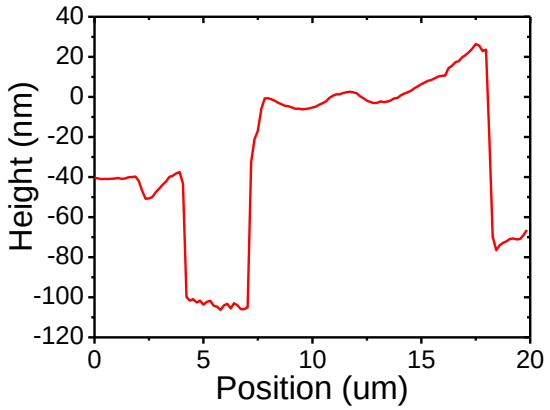
Flexible direct X-ray detectors based on Organic thin films

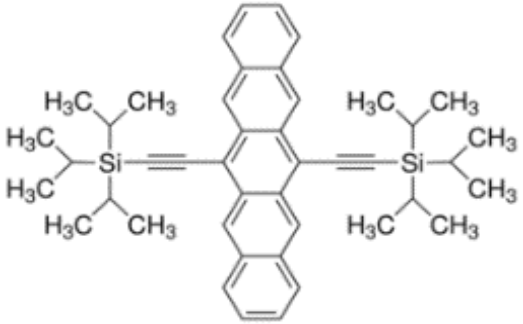












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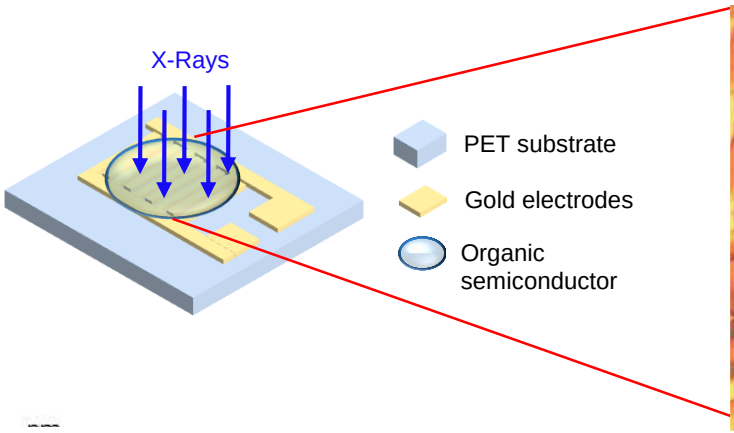
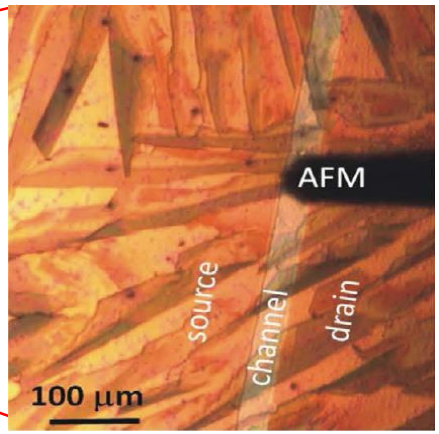
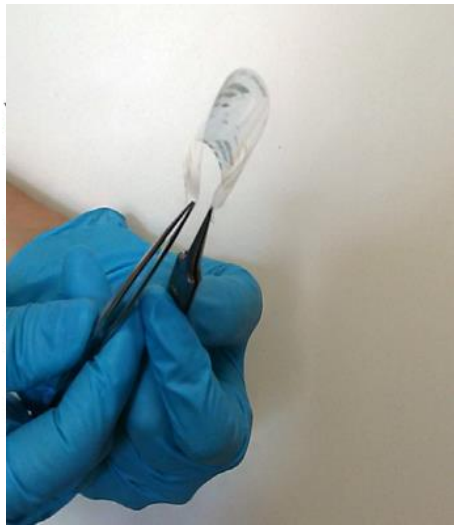
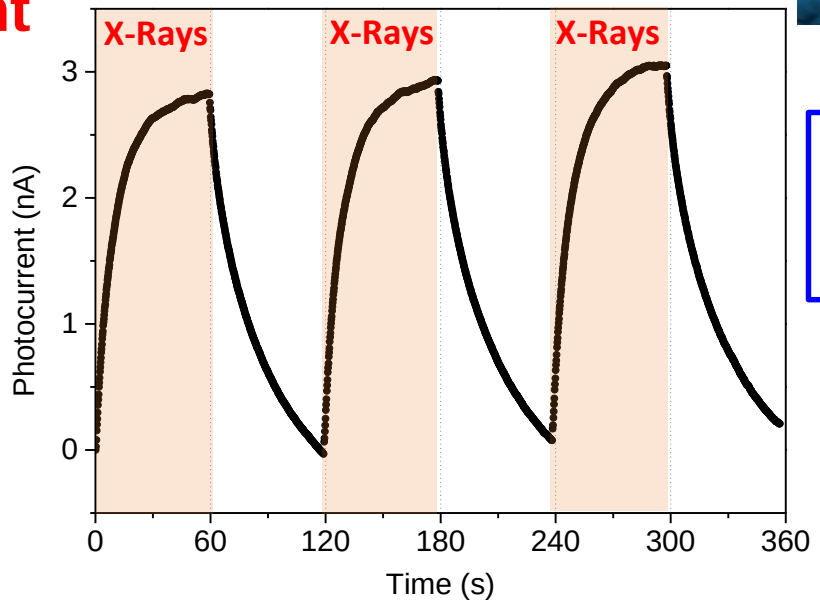






X-ray photocurrent

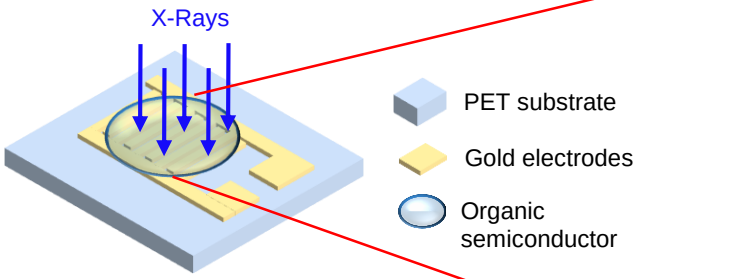
Synchrotron X-ray beam
Energy 17 keV
Dose rate 19 mGy/s

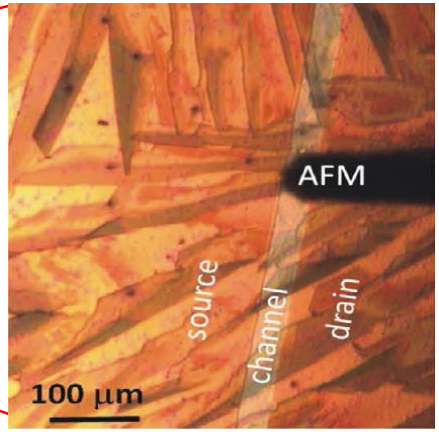


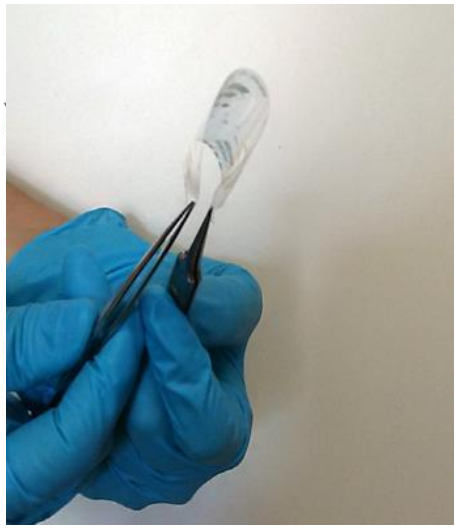
ΔI = 3 nA (max 30nA)

bias voltage 0.2 V

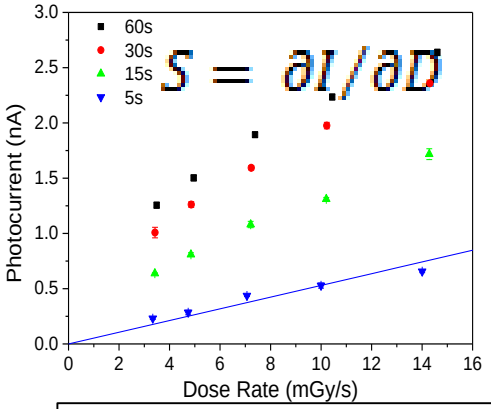
Flexible direct X-ray detectors based on Organic thin films







180 nC/Gy (72000 nC/mGy cm³) @0.2V



Sensitivity: one order of magnitude higher than thin polymeric films or bulk organic single crystals(biased at several tens of volts)

Room temperature and real-time operation

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

Literature:
S ≈ 450nC/mGycm³
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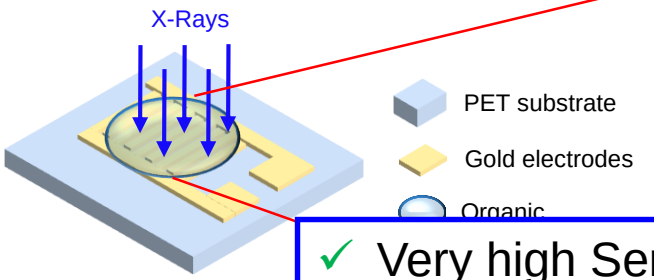
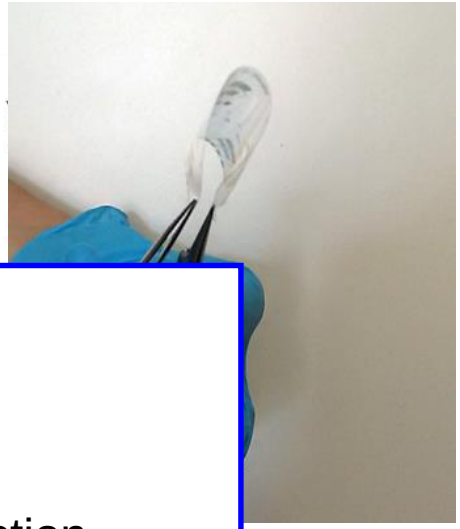
A. Intaniwet et al. / Organic Electronics 12 (2011) 1903

Flexible direct X-ray detectors based on Organic thin films

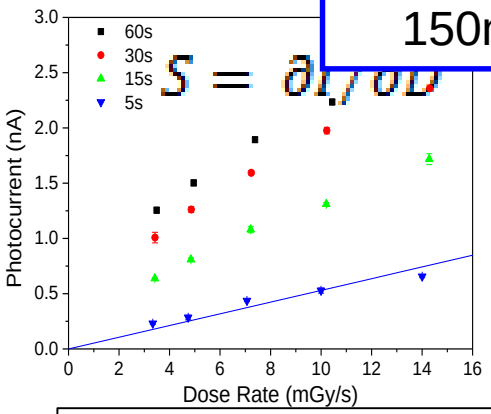




- ✓ Very high Sensitivity
- ✓ Flexible (100nm thin film)
- ✓ Very low voltage (0.2V)

- X Extremely low Q.E. (0.0015% attenuated fraction 150nm thick film)

180 nC/



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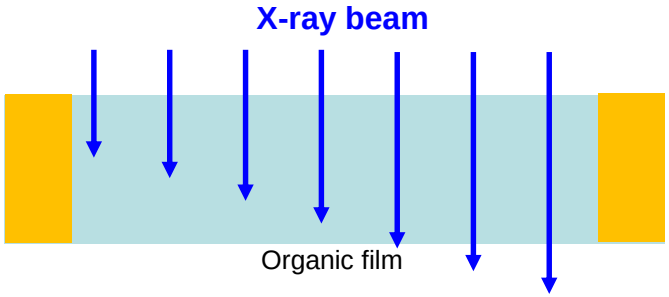
Charge Collection Model



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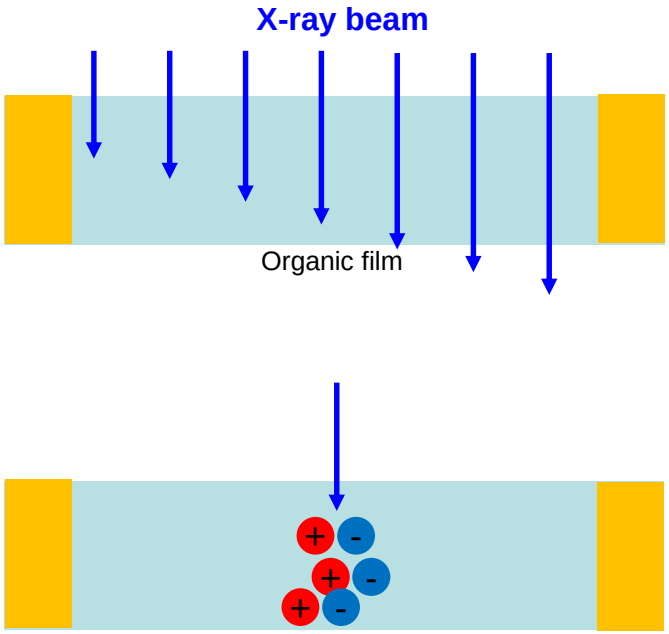

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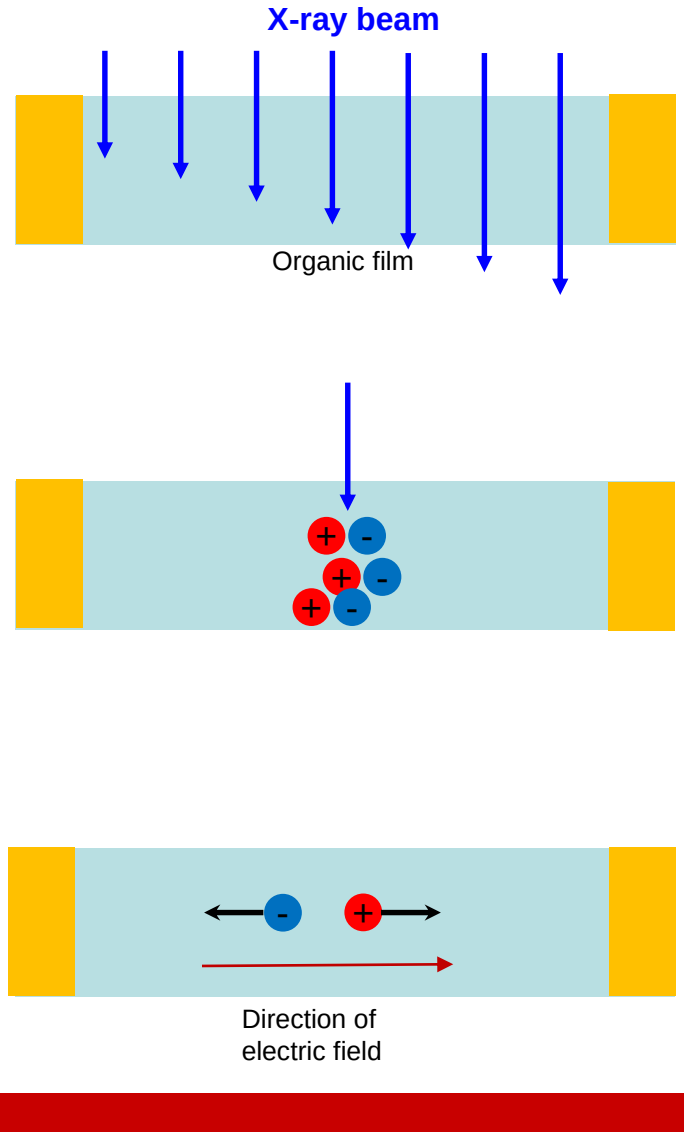

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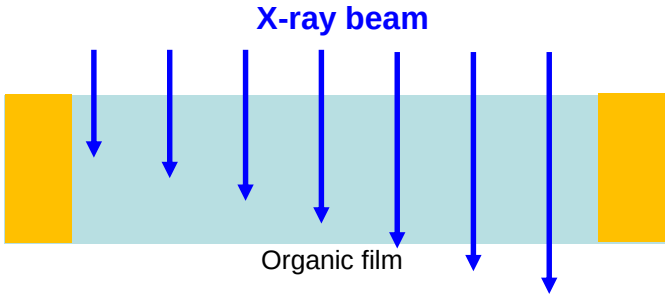
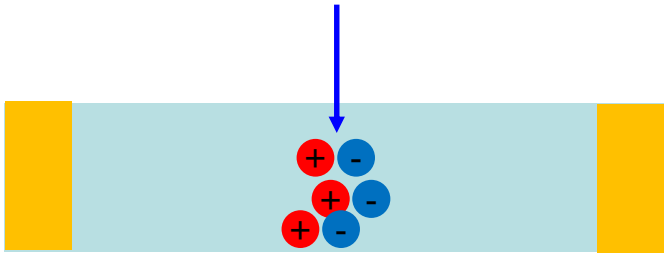


X-ray photocurrent

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

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



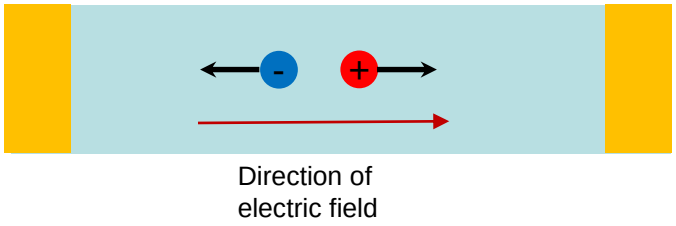
Absorbed photon flux

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

E_{ph} is the photon energy
$E_g \approx 2$ **eV** the energy gap of TIPS-pentacene.
A = 0.015 cm² - active area
***h* = 100 nm** – thickness
λ = 0.68 cm - attenuation length
absorption = 0.0015 %

photogenerated carriers/photons

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



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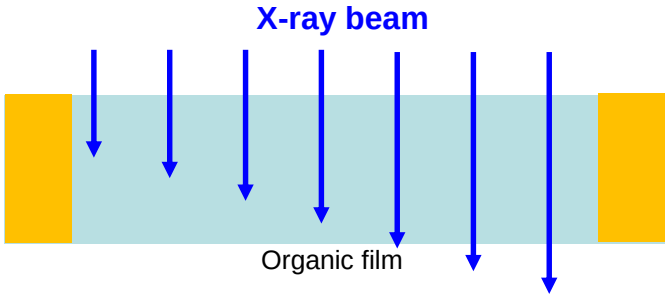

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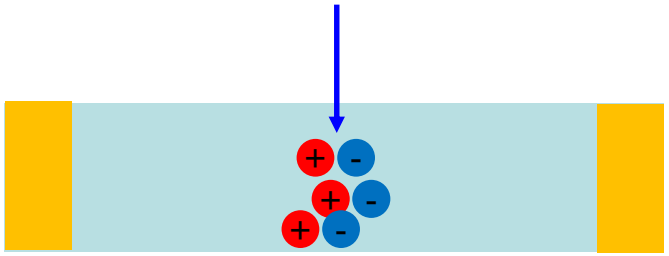

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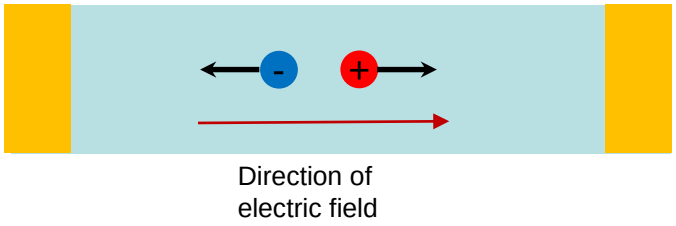
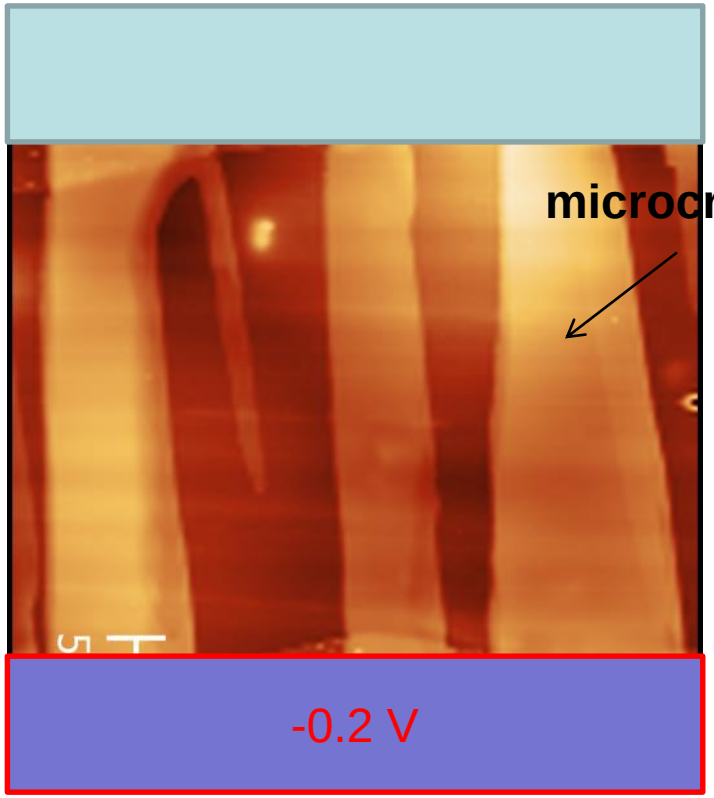


photogenerated carriers/photons

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

$I_{CC,max} < 2$ pA

Why is it working?
charge traps and photoconductive gain



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.

- trapping of n-type carriers
- injecting contacts

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

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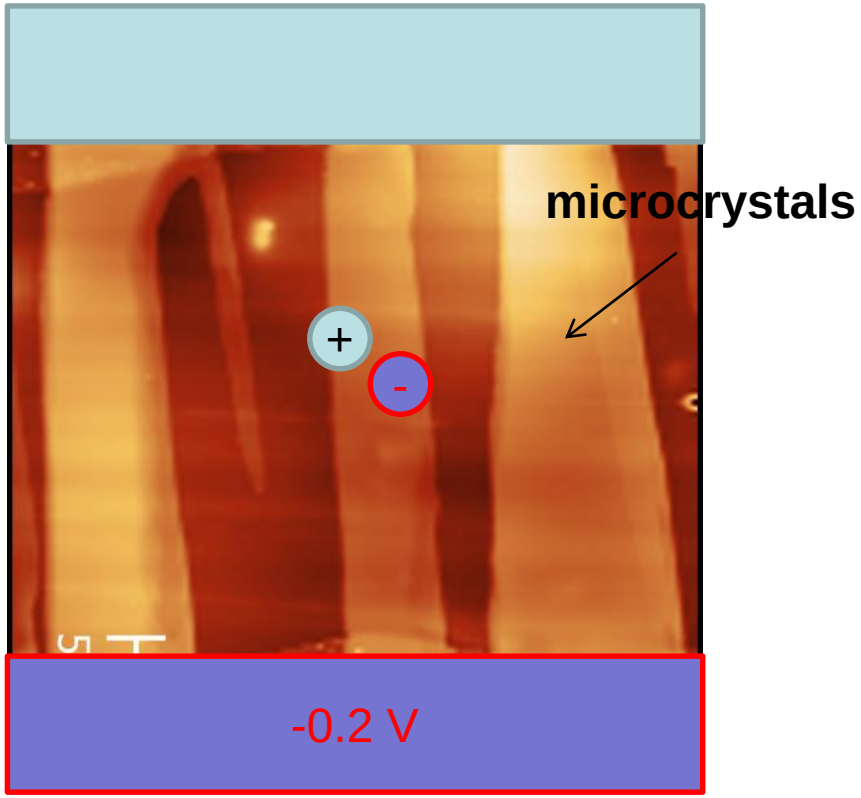

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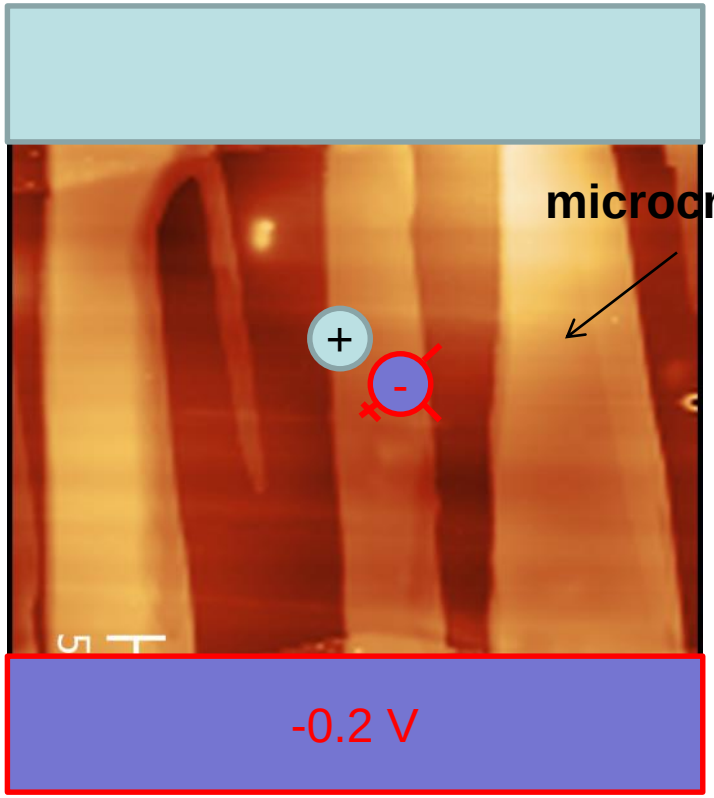

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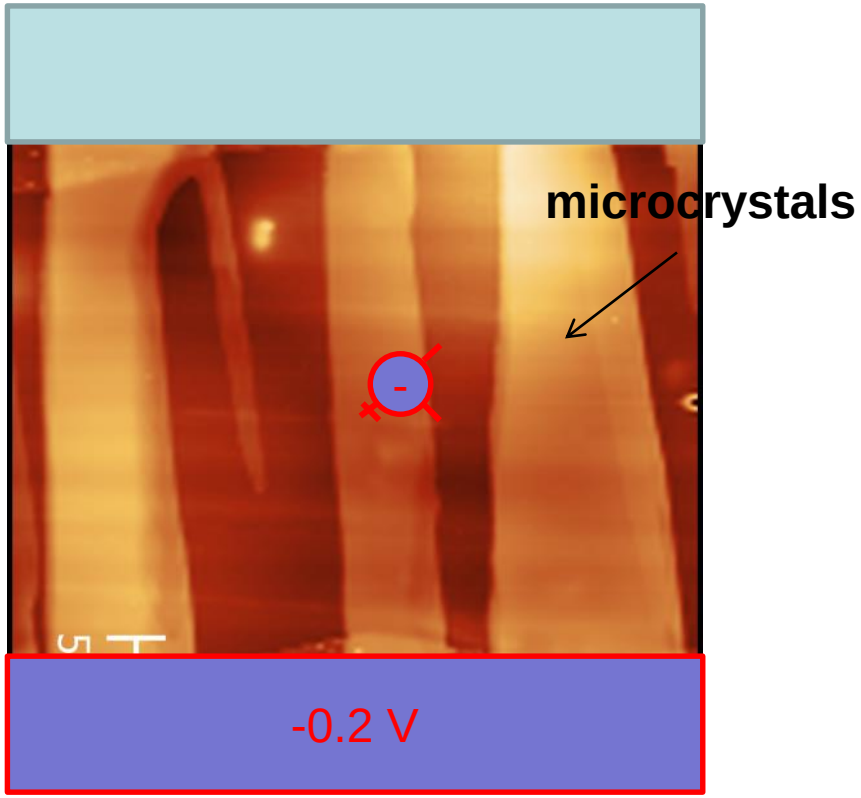

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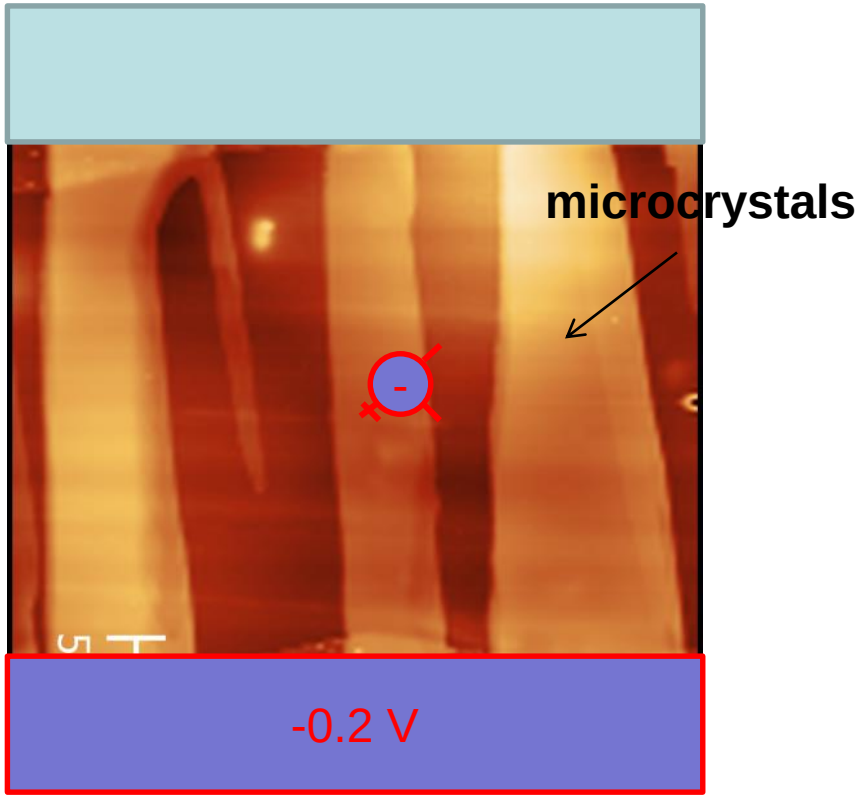

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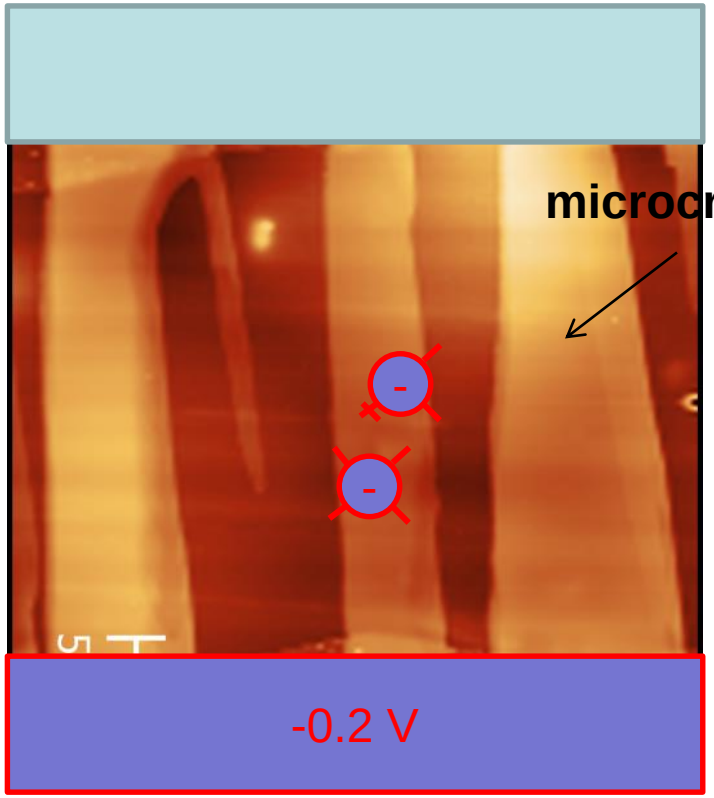

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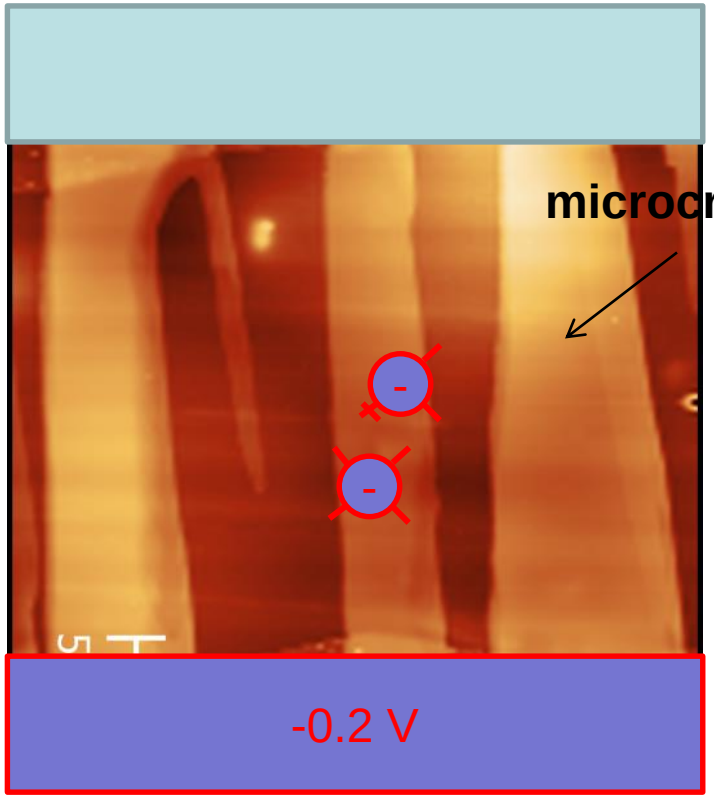

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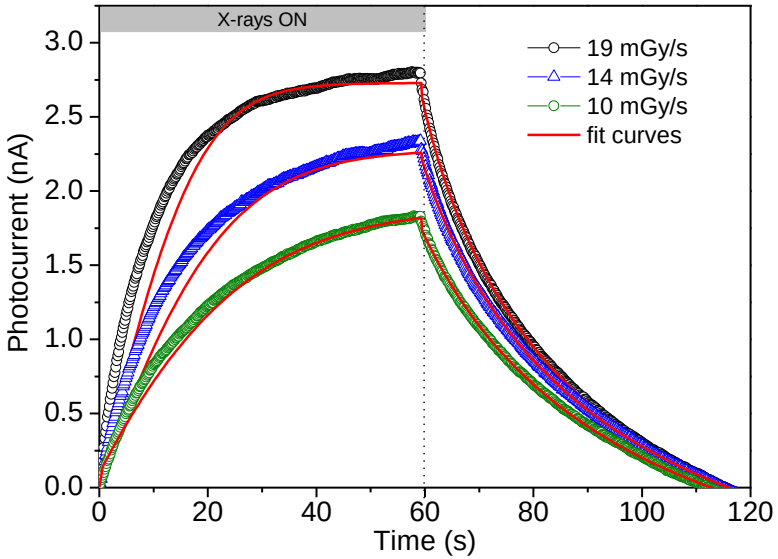
$$\Delta I_{PG} = G\, I_{CC}$$

G = photoconductive gain ≈ 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 μm, 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

$$G = \frac{\tau_r}{\tau_t} = \frac{29.4}{1.1\times10^{-3}} = 2.6\times10^4$$

Carriers lifetime

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

Transit time

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

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** (BUT low dark current)

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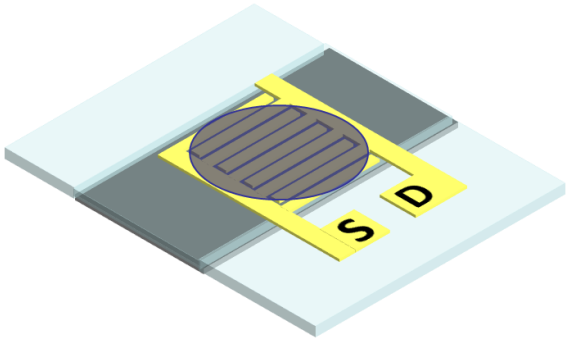
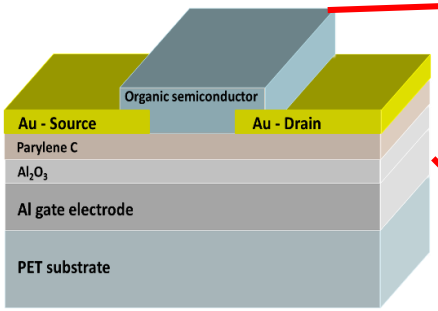
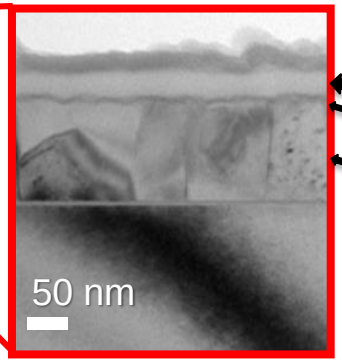
Towards Scaling-up the system: Low Voltage Organic Field Effect Transistors as X-ray detectors

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







Parylene C

Al_2O_3

Al

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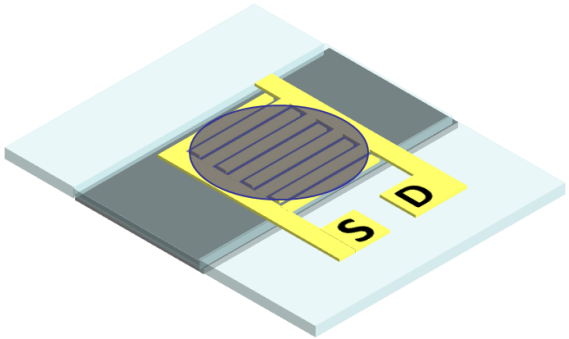
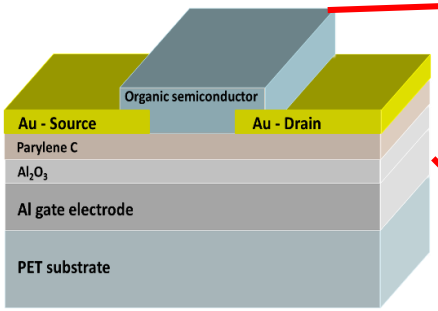
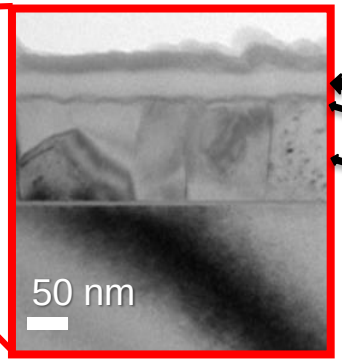
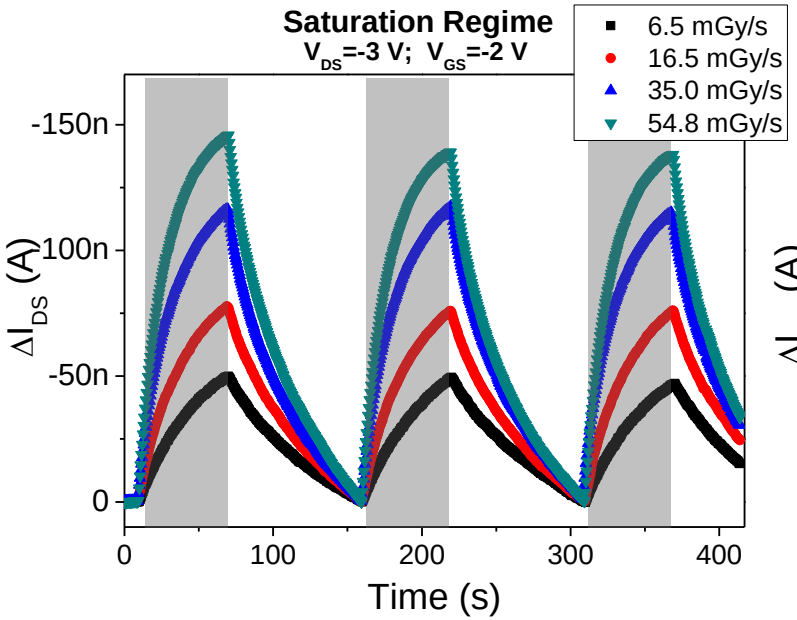
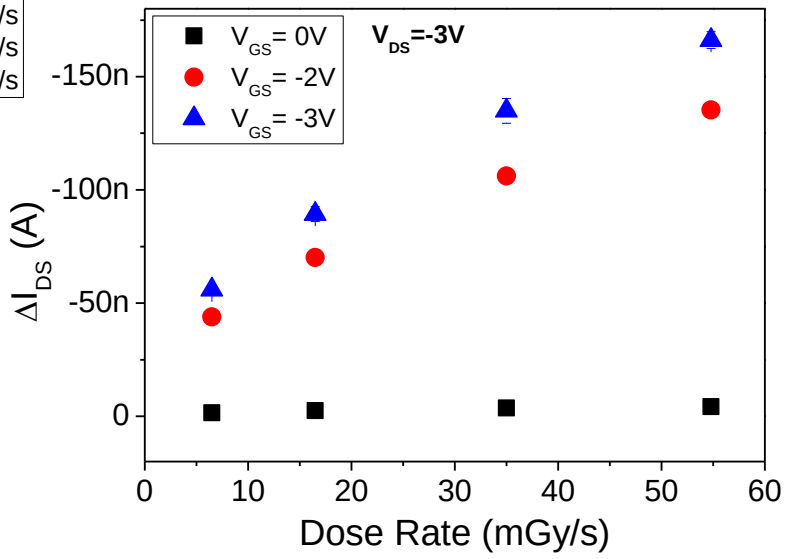
P.Cosseddu et al., AppPhysLett,100 (2012)







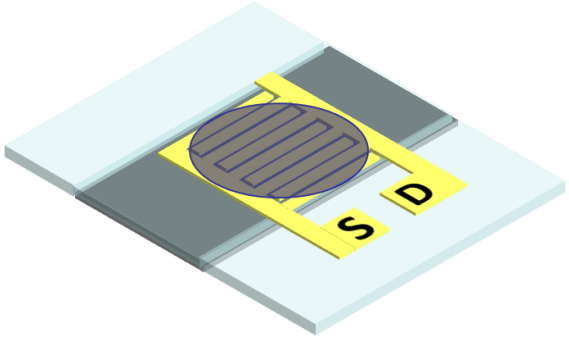


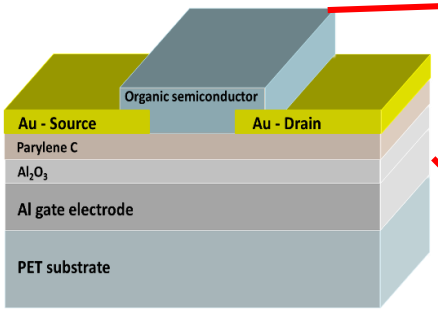


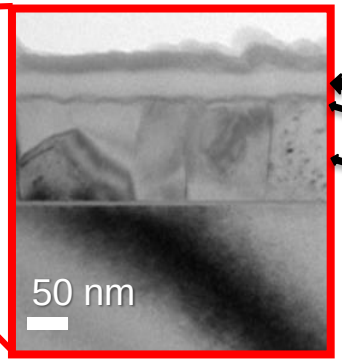
S.Lai et al., Adv. El. Mat., in press, 2017

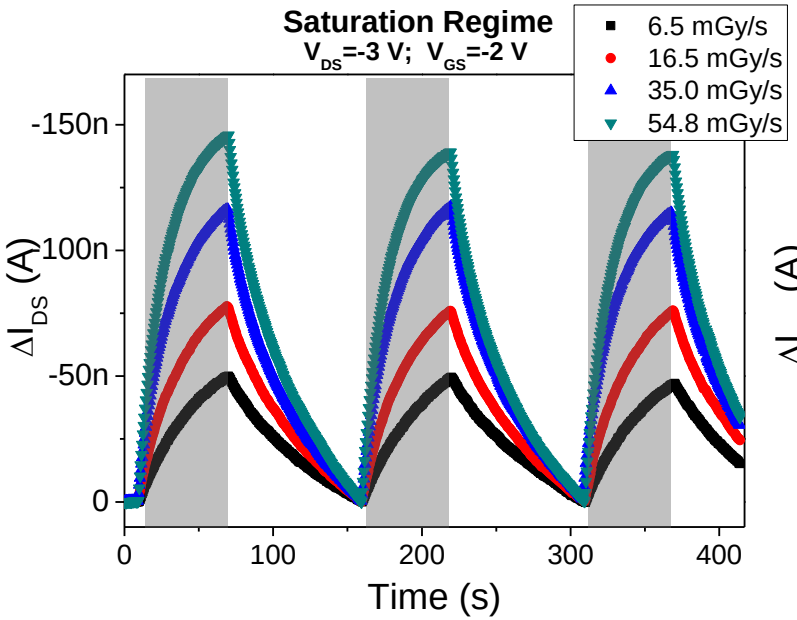
Towards Scaling-up the system: Low Voltage Organic Field Effect Transistors as X-ray detectors

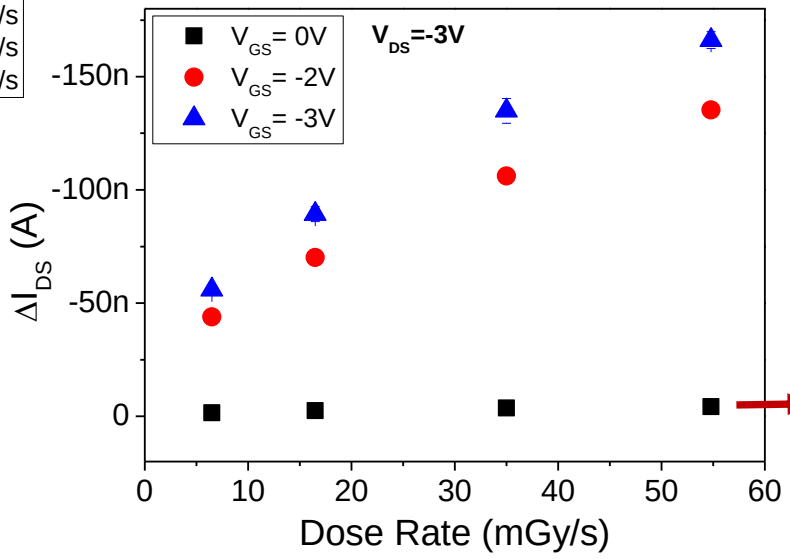
P.Cosseddu et al., AppPhysLett,100 (2012)











Transistor/Sensor OFF

S.Lai et al., Adv. El. Mat., in press, 2017

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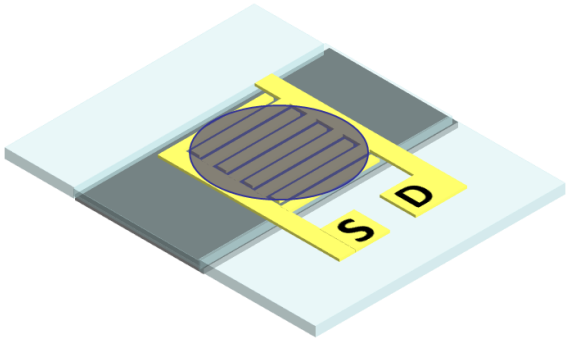

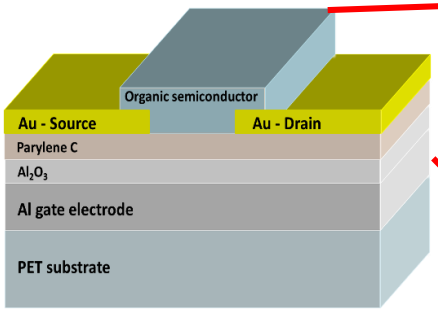

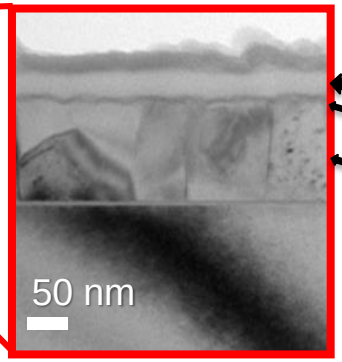

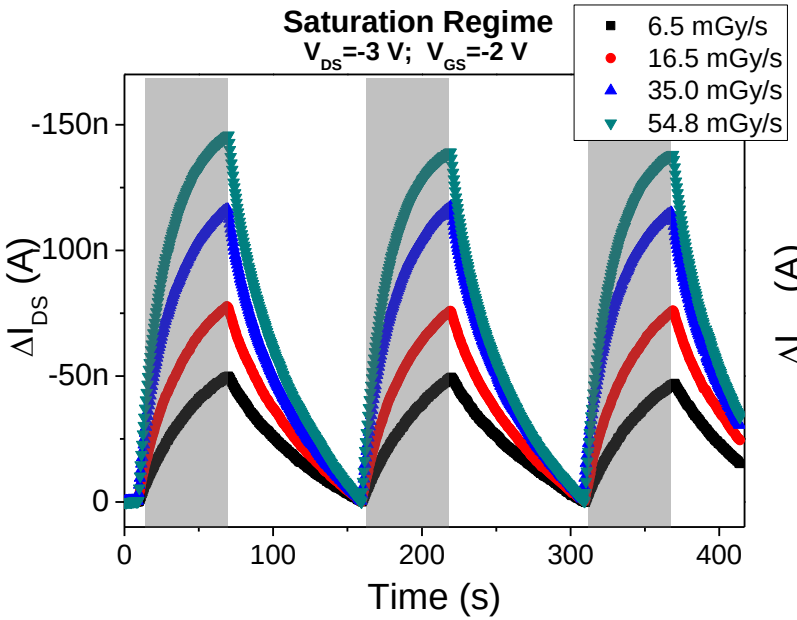

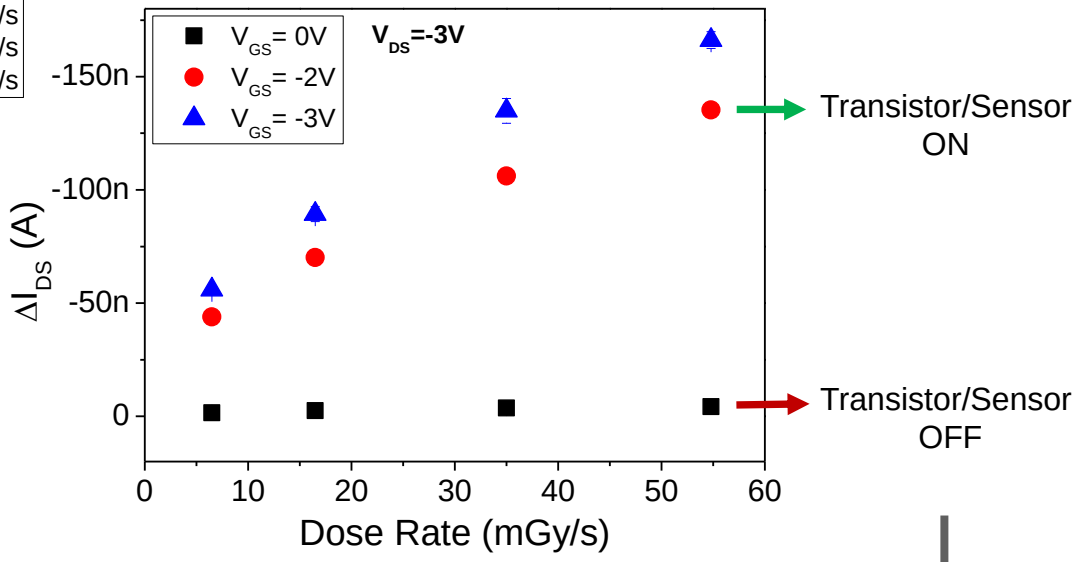

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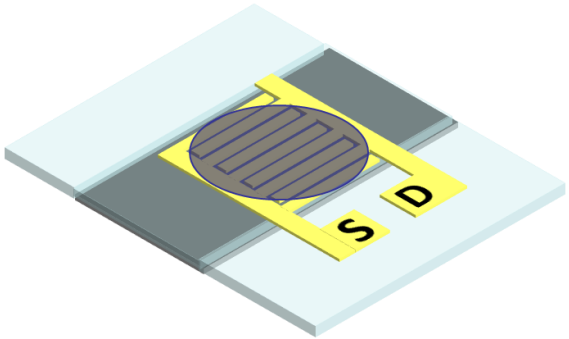

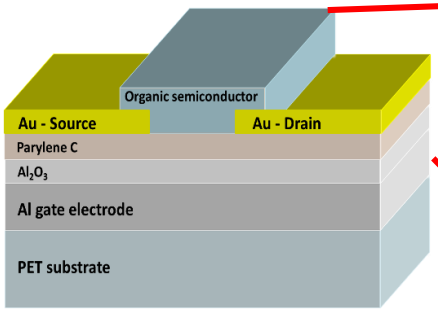

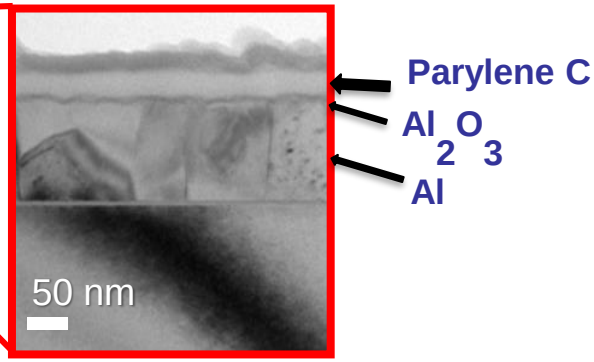

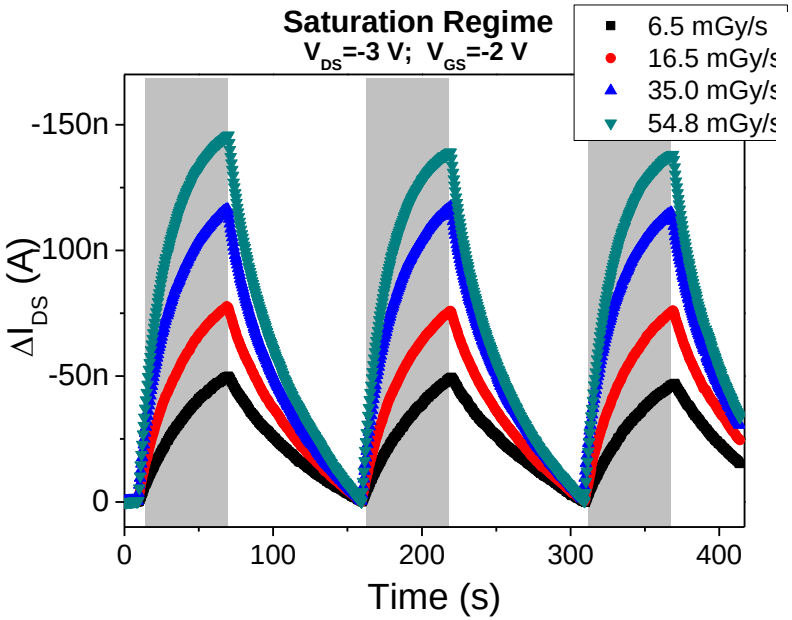

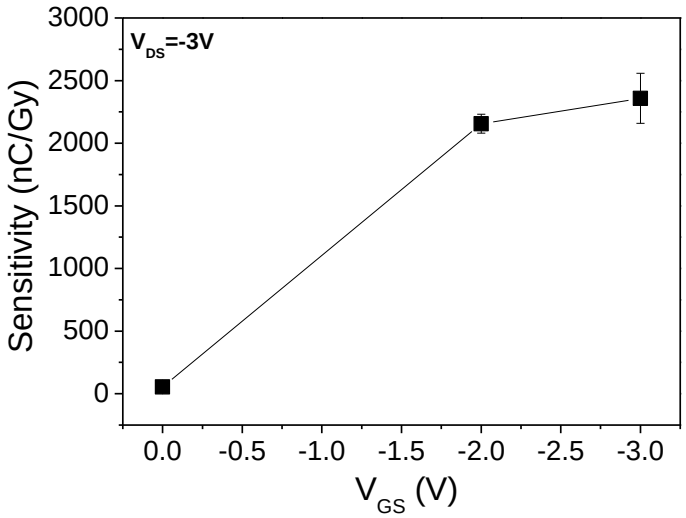

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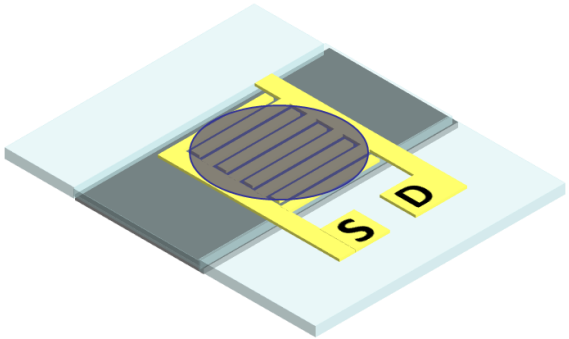
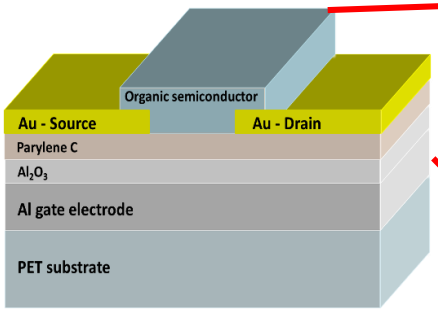
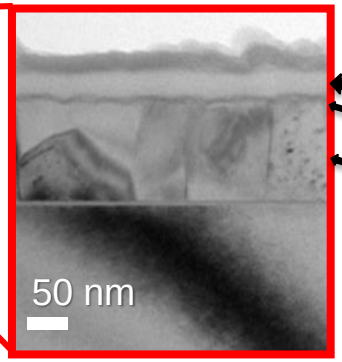
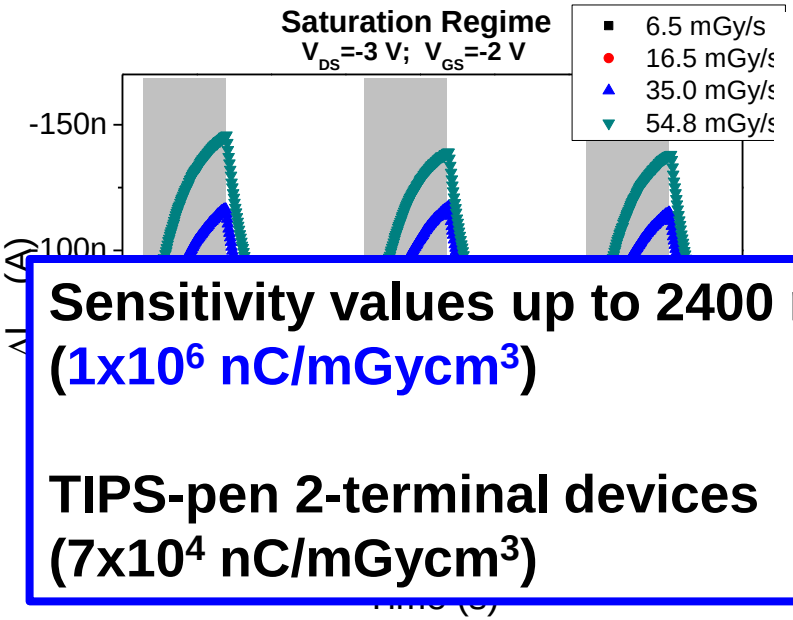
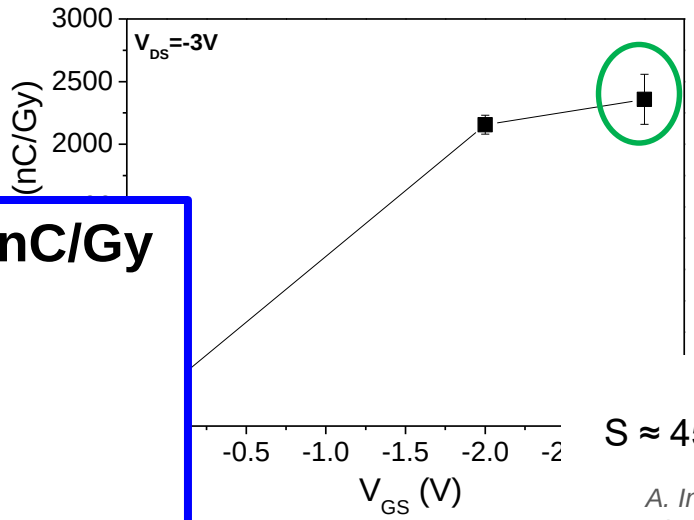










Sensitivity values up to 2400 nC/Gy (1x10^6 nC/mGycm^3)

TIPS-pen 2-terminal devices (7x10^4 nC/mGycm^3)

Literature:
S ≈ 450nC/mGycm^3 @40V

A. Intaniwet et al. / Organic Electronics 12 (2011) 1903–1908

S.Lai et al., Adv. El. Mat., in press, 2017

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)

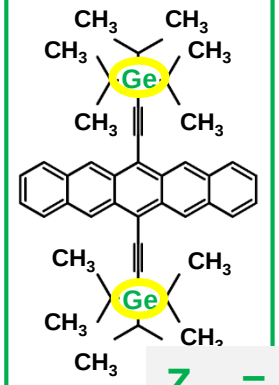
Increasing attenuated fraction & mobility: TIPGe

NOVEL MOLECULE!

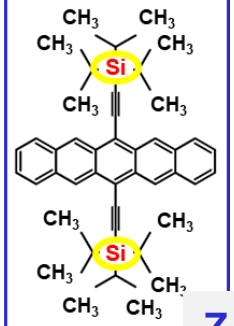
University of Kentucky

Prof. John Anthony

TIPGe -pentacene

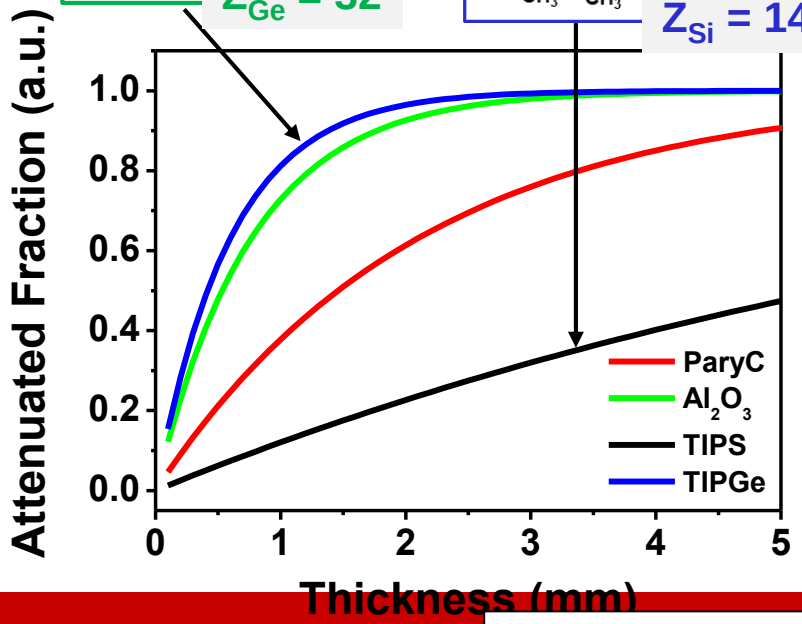


TIPS -pentacene



$Z_{Ge} = 32$

$Z_{Si} = 14$



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Increasing attenuated fraction & mobility: TIPGe

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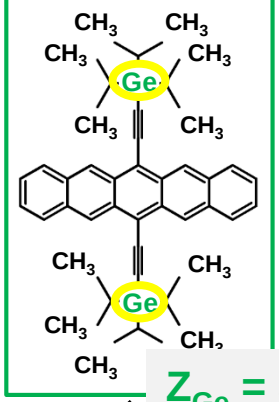

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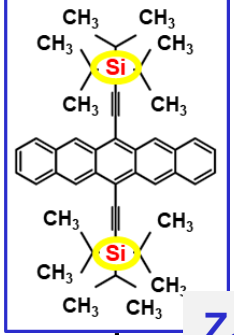

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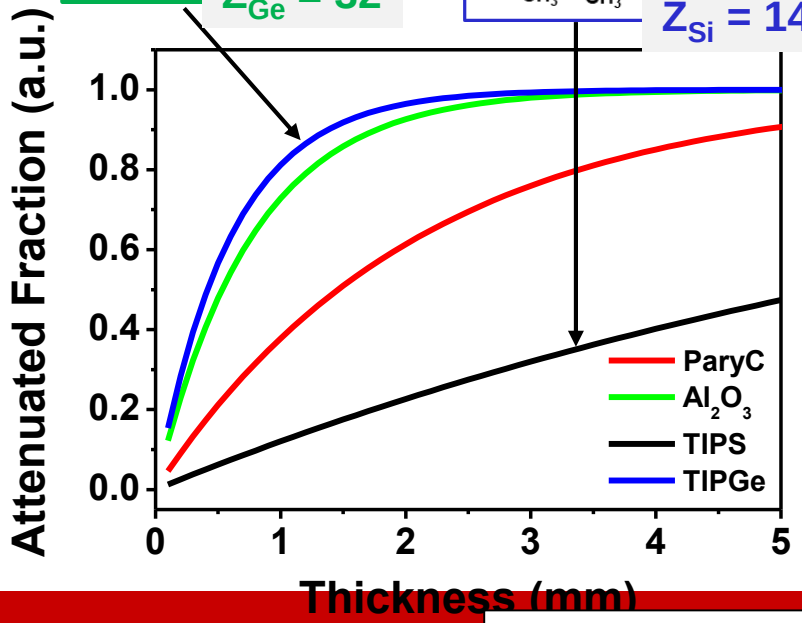

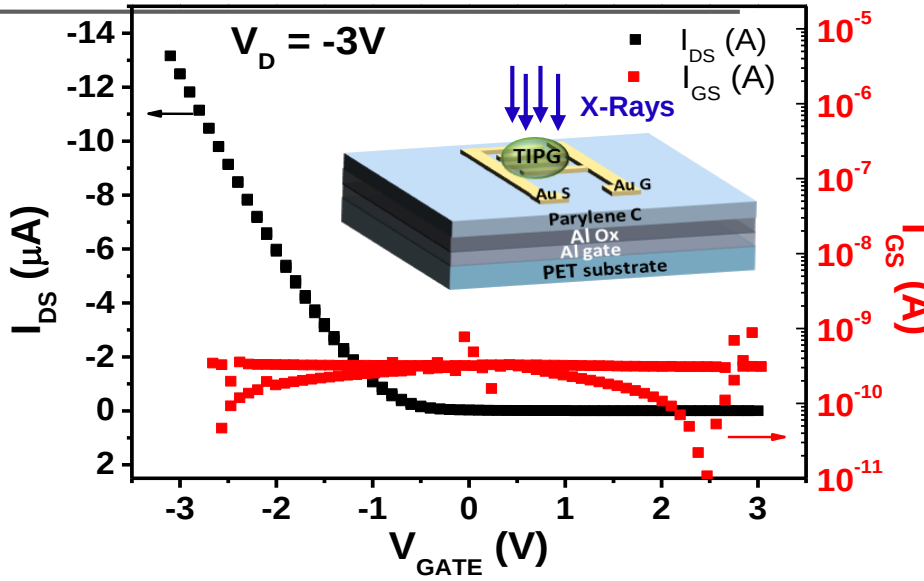

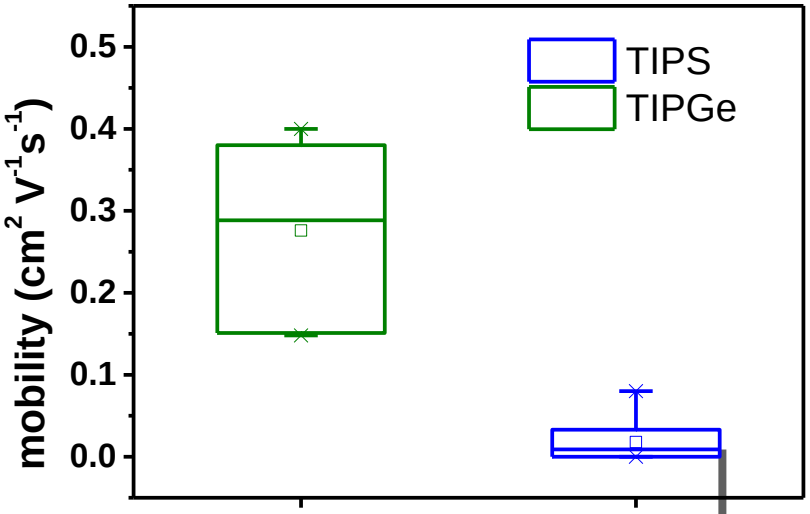

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Increasing attenuated fraction & mobility: TIPGe

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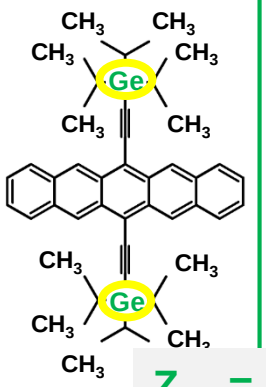

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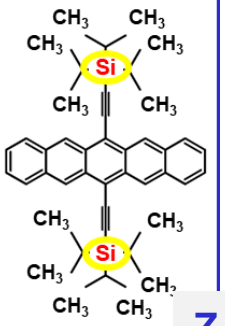

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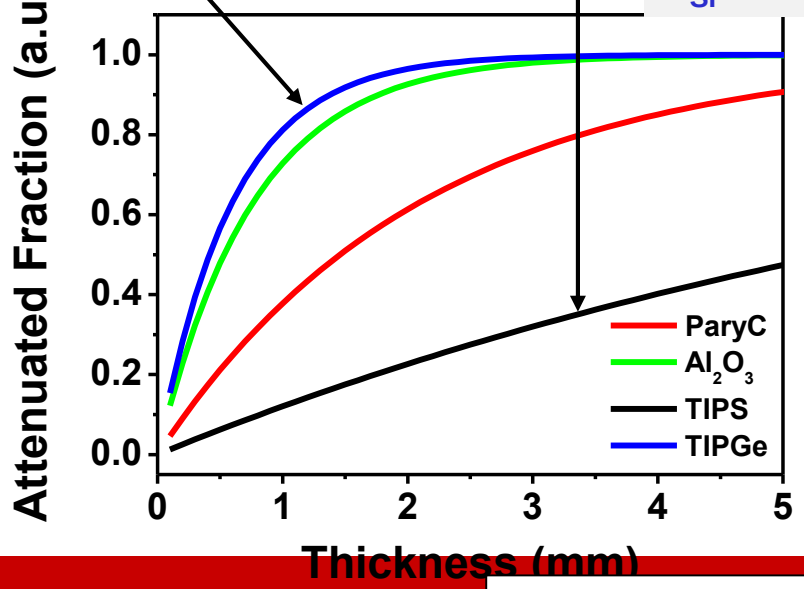
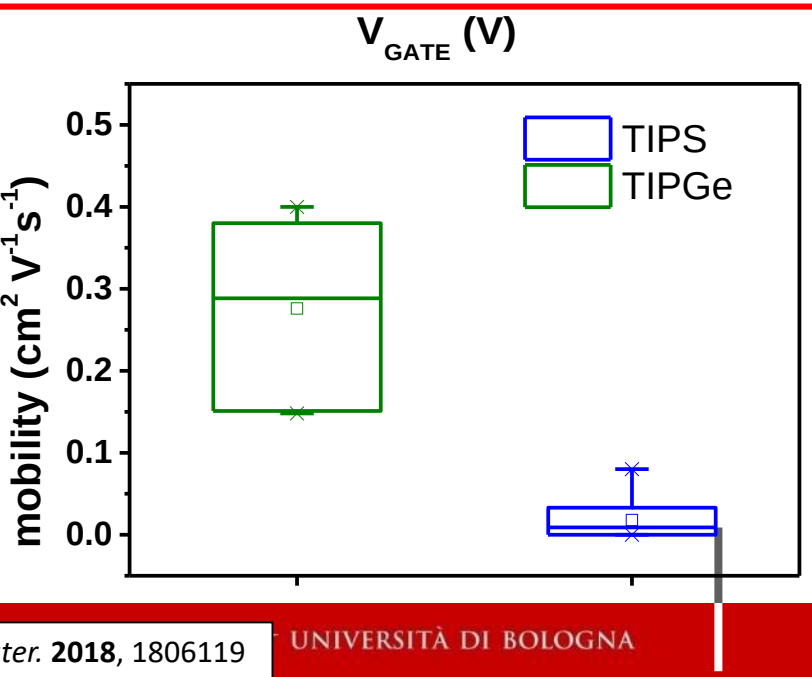


→ TIPGe attenuated fraction 0.03% @ 200nm (x10 times than TIPS film)

→ TIPGe mobility 0.27 cm^2/Vs (x10 times than TIPS film)

Higher sensitivity than TIPS is expected



I_{GS} (A)

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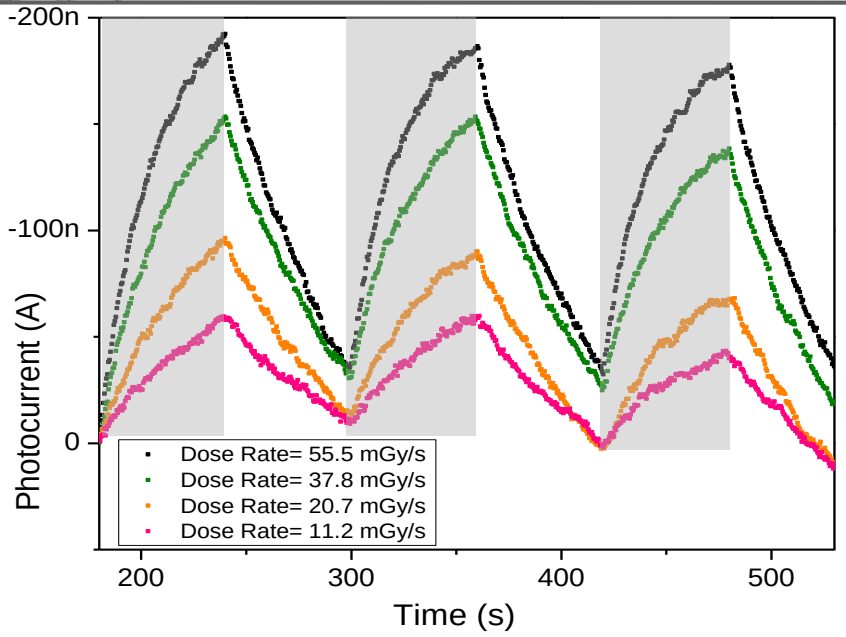
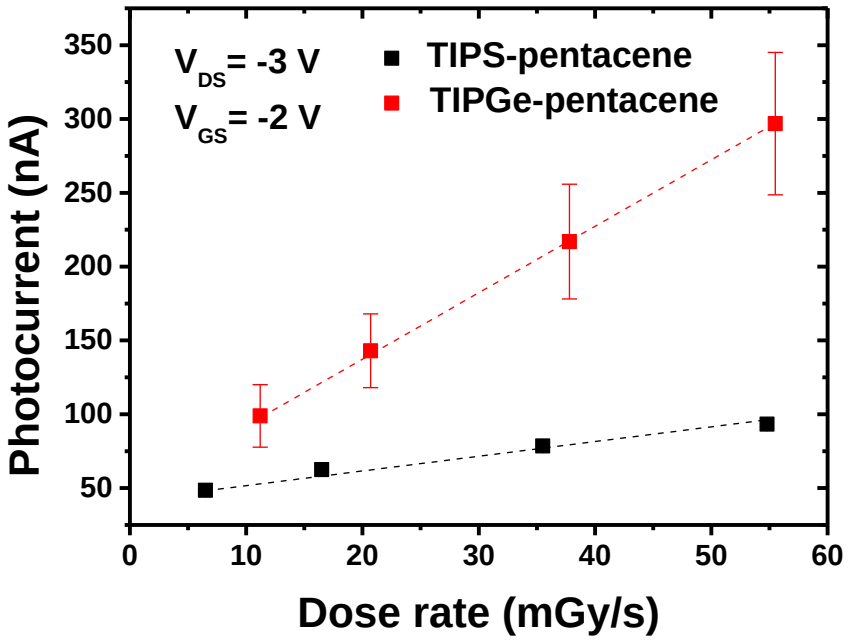
Increasing attenuated fraction & mobility: TIPGe





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

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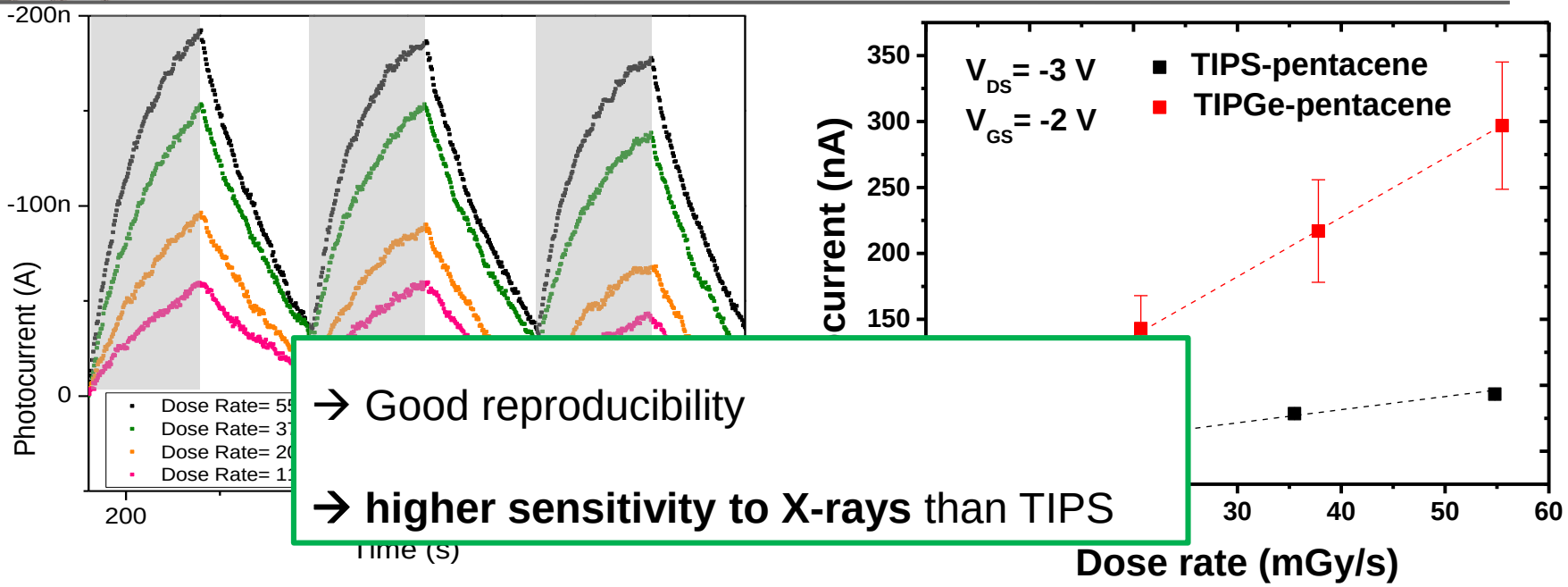
Increasing attenuated fraction & mobility: TIPGe



→ Good reproducibility

→ **higher sensitivity to X-rays** than TIPS

Molecule	Solution	Mobility [$cm^2V^{-1}s^{-1}$]	Operative voltage	Film thickness [nm]	Average Sensitivity [nC Gy^{-1}]	S_V [µC Gy^{-1} cm^{-3}]
TIPS	0.5wt.% in toluene dropcast	0.02 ± 0.01	V_{DS} = -3V V_{GS} = -2V	150	960 ± 70	$(2.6 \pm 0.2) \cdot 10^5$
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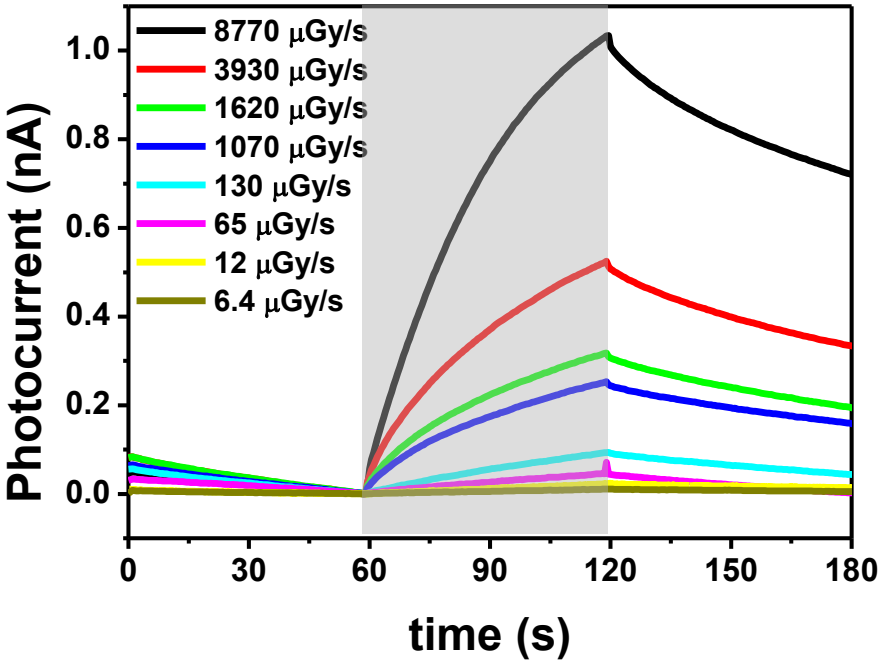
Increasing attenuated fraction & mobility: TIPGe



LIMIT OF DETECTABILITY

Lowest detectable dose rate	Lowest detectable total dose
1.66 µGy s^{-1}	**12 µGy**

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

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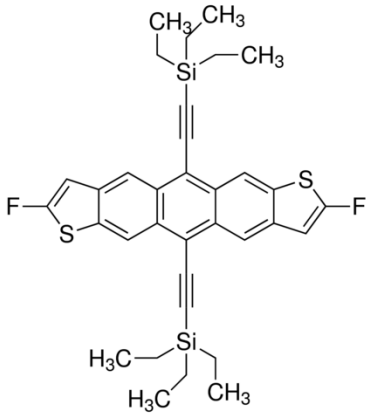
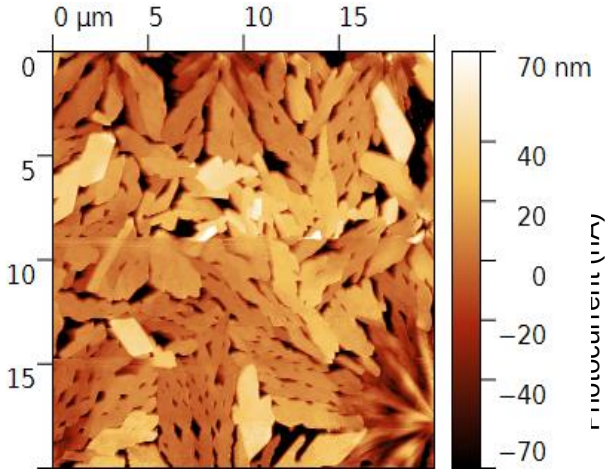
Other Ge-based acenes

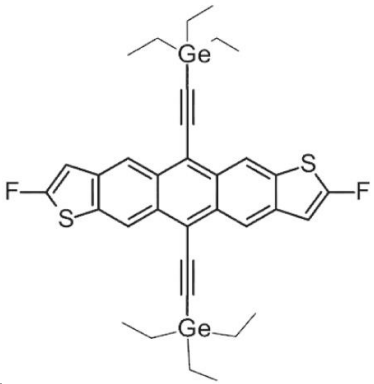
diF-TE**S**-ADT

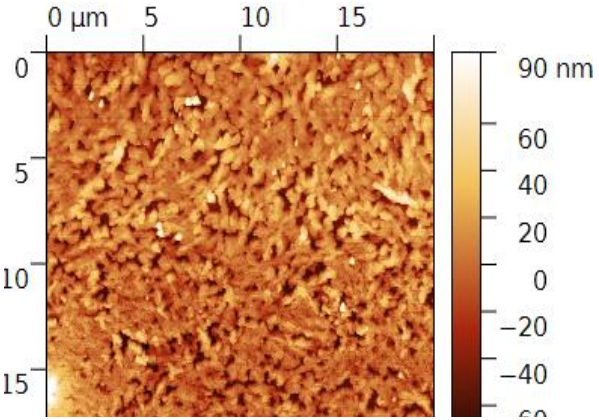




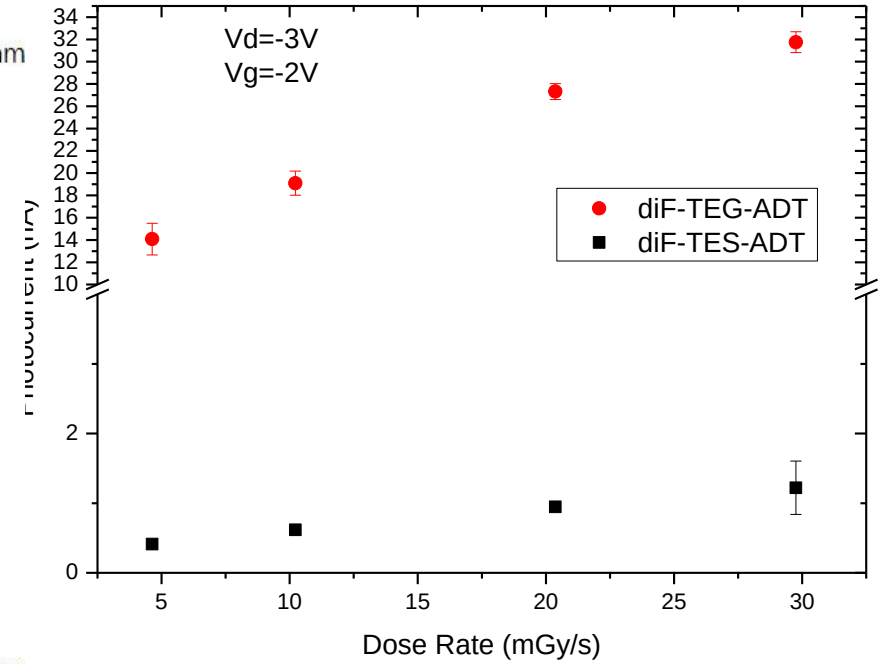
2,8-Difluoro-5,11-bis(triethylsilylethynyl)anthradithiophene

diF-TE**G**-ADT





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

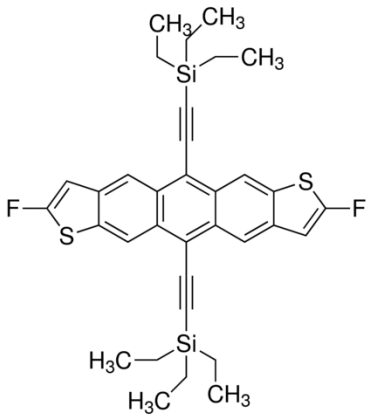


Molecule	Mobility [$cm^2V^{-1}s^{-1}$]	Operative voltage	S_V [$\mu C\ Gy^{-1}\ cm^{-3}$]
diF-TE**S**-ADT	$(5.2\pm0.3)\cdot10^{-2}$	V_{DS} = -3V V_{GS} = -2V	80±10
diF-TE**G**-ADT	$(6.9\pm0.4)\cdot10^{-2}$		3400±400

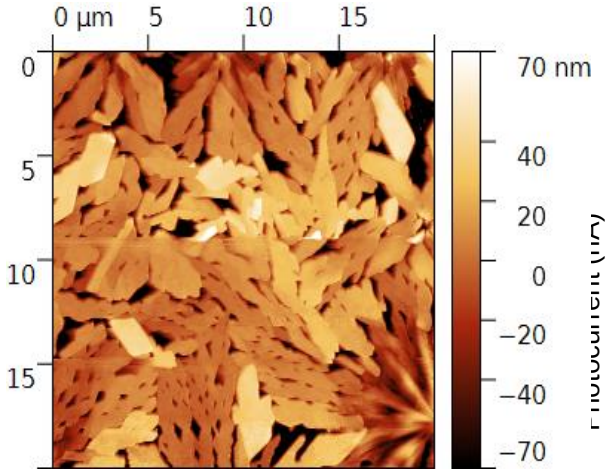
Ciavatti, Basiricò et al., Adv. Funct. Mater. **2018**, 1806119

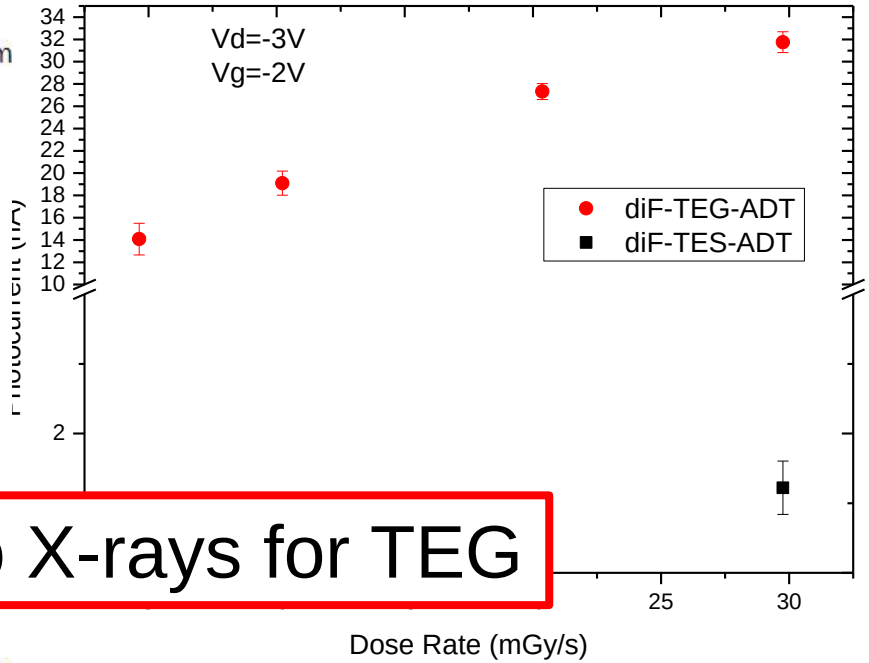
Other Ge-based acenes

diF-TE**S**-ADT



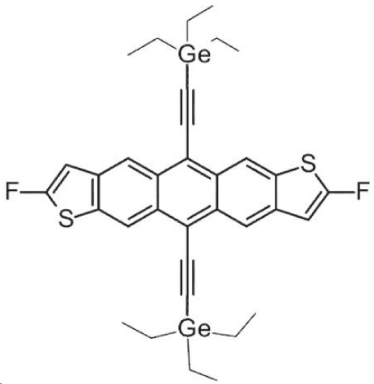
2,8-Difluoro-5,11-bis



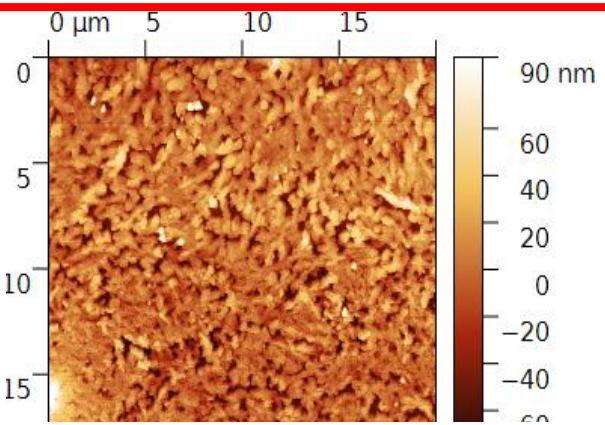


Higher sensitivity to X-rays for TEG

diF-TE**G**-ADT



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



Molecule	Mobility [$cm^2V^{-1}s^{-1}$]	Operative voltage	S_V [$\mu C\ Gy^{-1}\ cm^{-3}$]
diF-TES-ADT	$(5.2\pm0.3)\cdot10^{-2}$	V_{DS} = -3V V_{GS} = -2V	80±10
diF-TEG-ADT	$(6.9\pm0.4)\cdot10^{-2}$		3400±400

Ciavatti, Basiricò et al., Adv. Funct. Mater. **2018**, 1806119

Conclusions

- Novel flexible direct **X-ray detectors** based on organic semiconducting **thin films**, deposited from solution, operating at **very low bias** (< 1V).

- **Amplification** of photoconductive gain effect by the employment of **low voltage OTFTs**.

- Improved the sensitivity of organic thin films, thanks to order of magnitude increasing of **X-ray stopping power** and **mobility** in **Ge-based** molecules**.**

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Thank you for your attention