UNIVERSITY OF OXFORD

Nano-Day 2020

Pierluigi Mondelli

ITN Marie-Curie PhD student
Advanced Functional Material and Devices Group
Condensed Matter Physics | Physics Department | University of Oxford
Clarendon Laboratory, Parks Road
Oxford OX1 3PU, England, UK

email: pierluigi.mondelli@physics.ox.ac.uk

On-line meeting

17-06-20

European Commission | Horizon 2020 European Union funding for Research & Innovation

SEPOMO

MERCK

Previous Experience at Sapienza University of Rome

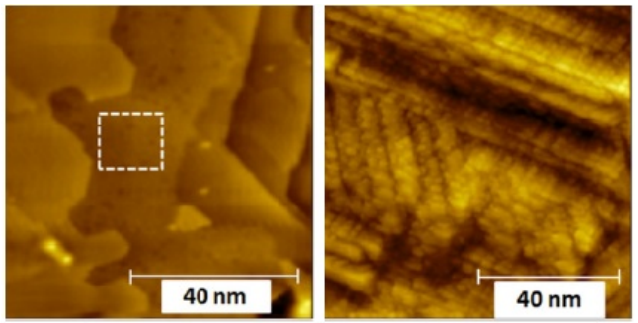
BSc in Mechanical Engineering and MSc in Nanotechnology

Improving the graphene quality for silicon-based electronics (MSc thesis project)

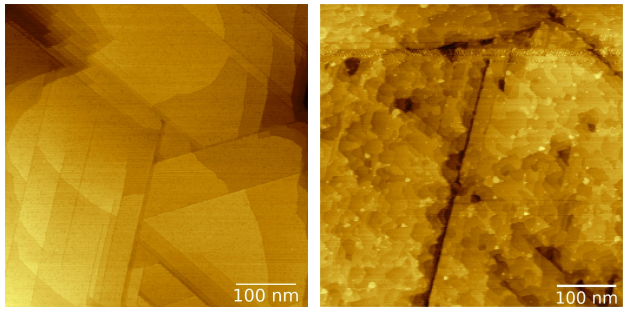
Supervisors: Prof C. Mariani, Prof N. Motta (QUT)

Treated Un-treated

Effect of substrate polishing

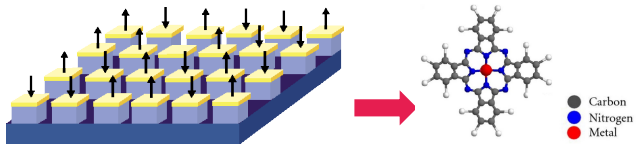


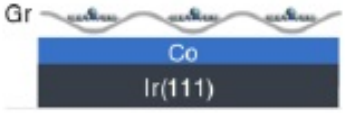
Effect of hydrogen etching



Single-Molecule Magnet (SMM) self-assembling for spintronics (1-year research fellowship)

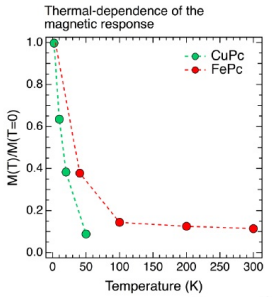
Supervisors: Prof C. Mariani and Prof M. G. Betti (Sapienza)

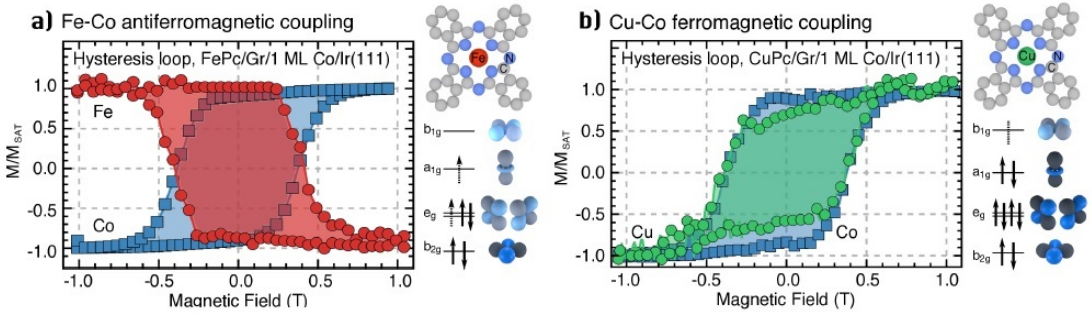




Ordered network of SMMs

The different d-state occupancy leads to different interaction mechanism with Co (super-exchange or direct exchange)





Previous Experience at Sapienza University of Rome

Summary of the Results - Publications

Improving the graphene quality for silicon-based electronics (MSc thesis project)

- **Mondelli P.**, Gupta B., Betti M. G. et al. High quality epitaxial graphene by hydrogen-etching of 3C-SiC(111) thin-film on Si(111) *Nanotechnology* 28 (2017) 115601
- Gupta B., di Bernardo I., **Mondelli P**, et al. Effect of Substrate Polishing on the Growth of graphene on 3C-SiC(111)/Si(111) by high temperature annealing *Nanotechnology* 27 (2016) 185601

Single-Molecule Magnets (SMM) self-assembling for spintronics (1-year research fellowship)

- Avvisati G., Gargiani P., **Mondelli P.** et al. Metal phthalocyanines interaction with Co mediated by a moiré graphene superlattice, *The Journal of Chemical Physics* 2019, Vol. 150, Issue 5
- Avvisati G., **Mondelli P.**, et al. Graphene-mediated interaction between FePc and intercalated cobalt layers, *Applied Surface Science* 2018, Vol. 438 2-6
- Avvisati G., Gargiani P., **Mondelli P**. et al. Superexchange pathways stabilize the magnetic coupling of MnPc with Co in a spin interface mediated by graphene, *Physical Review B* 98, 115412 (2018)
- Gargiani P., Lisi S., Avvisati G., **Mondelli P**. et al. Mixing of MnPc electronic states at the MnPc/Au(110) interface, *The Journal of Chemical Physics* 2017, Vol. 147, Issue 13

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The Molecular Organization of Non-Fullerene Acceptors for Organic Photovoltaics

From Single-Crystals to Solar Cells

PhD Candidate: Pierluigi Mondelli
Supervisor: Prof Moritz Riede (University of Oxford)
Co-supervisor: Dr Graham Morse (Merck Chemicals Ltd)

On-line meeting

17-06-20

European Commission | Horizon 2020 European Union funding for Research & Innovation

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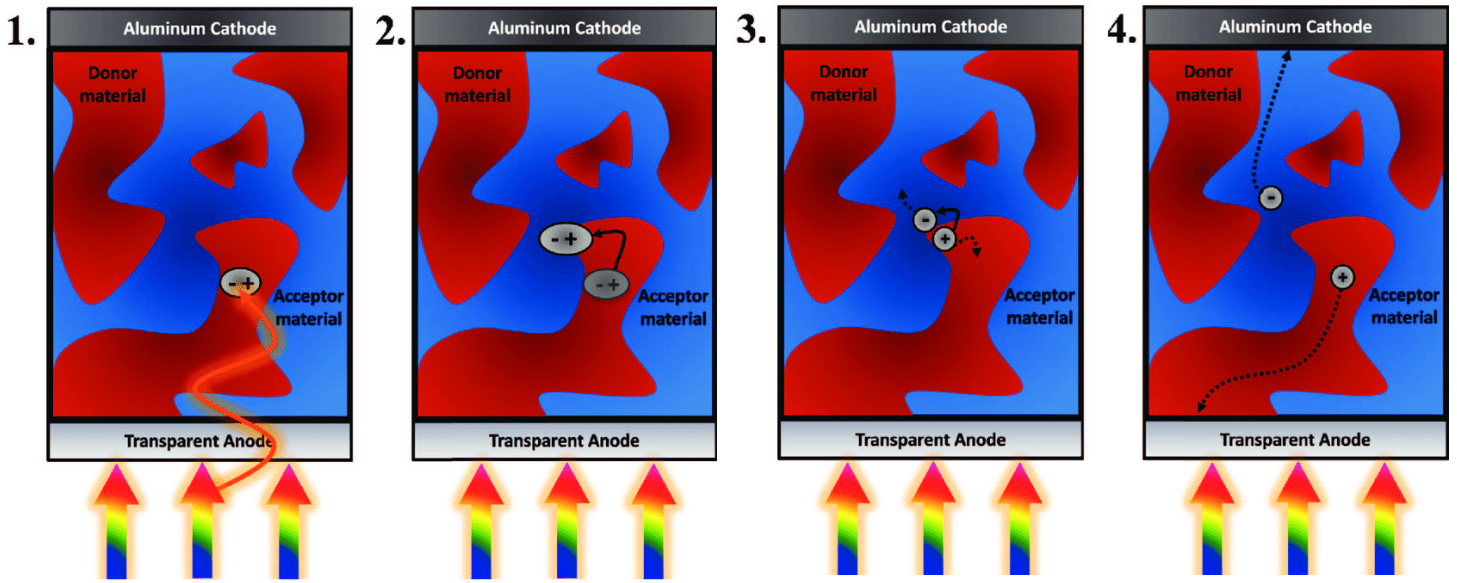
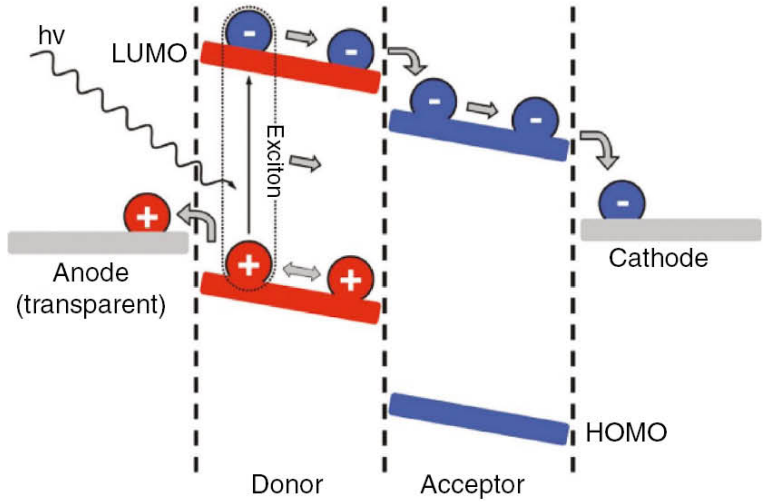
Principles of Organic Photovoltaics (OPV)

Formation and splitting of strongly-bound excitons (Frenkel excitons)

From light absorption to free charge collection



Energy diagram of an organic BHJ



1. Exciton formation upon light absorption
2. Migration to the donor/acceptor interface
3. Exciton splitting into free charges
4. Charge collection at the electrodes

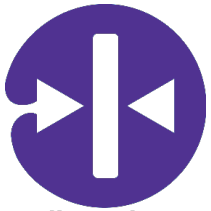
In inorganic semiconductors the free charges are generated upon light absorption (Wennier exciton with very low binding energy $E_B << kT \approx 25$ meV)

Printed Electronics and Organic Photovoltaics (OPV)

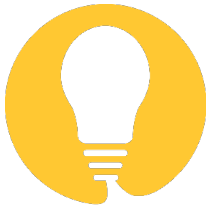
A technology for solution-processing addressing multiple needs

Indoor applications

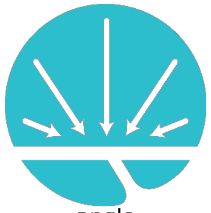
Self-powered indoor electronic devices



efficient from both sides



efficient indoor



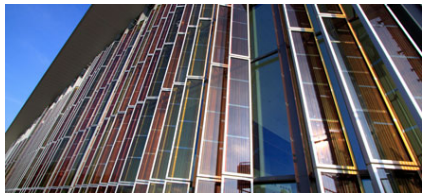
angle independent

choice of colors

opaque or semi transparent

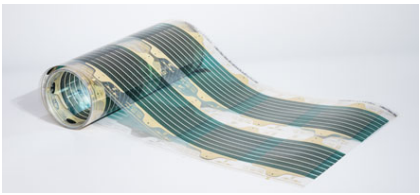
temperature independent

Outdoor applications

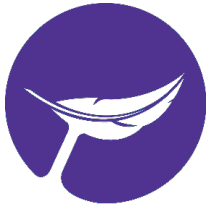


BIPV (Building Integrated Photovoltaics)

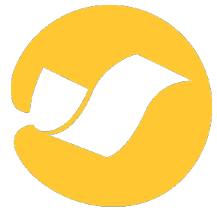
Processability



Mobile and flexible off-grid power supply



low weight



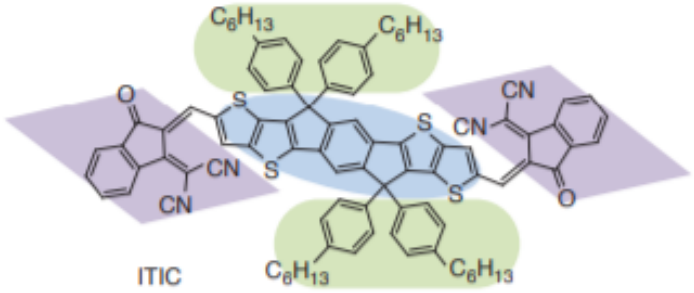
bendable & easy to laminate

tailor made

Non-Fullerene Acceptors (NFAs) vs Fullerenes

Chemical Structure → Solid-State Packing → Charge Transport

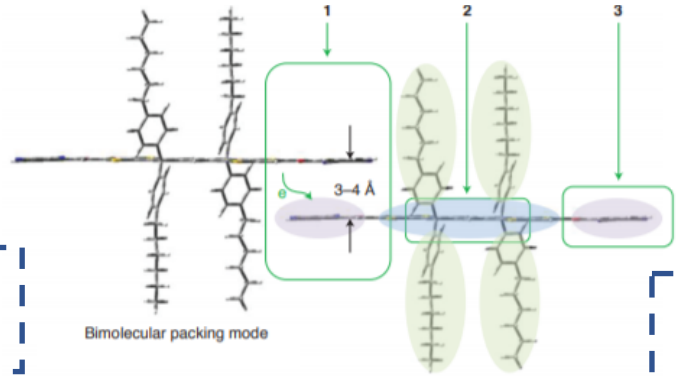
Acceptor-Donor-Acceptor π-conjugated molecule



Bulky non-conjugated
Electron pushing
Electron pulling

π-π stacking

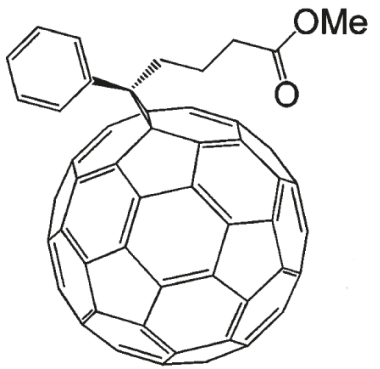
anisotropic domains



PCE > 16%
μe ~ 10^{-4} cm^2 V^{-1} s^{-1}

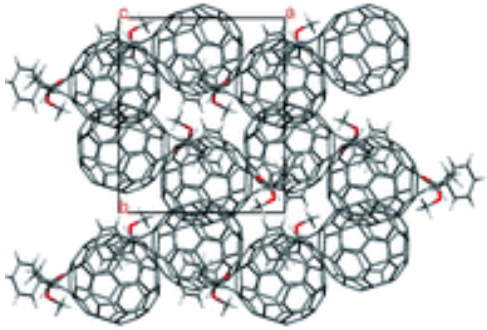
Hou, J. et al. *Nature Materials 17,* **2018,** 119

PCBM near-ball shape



π-π interactions

3D network

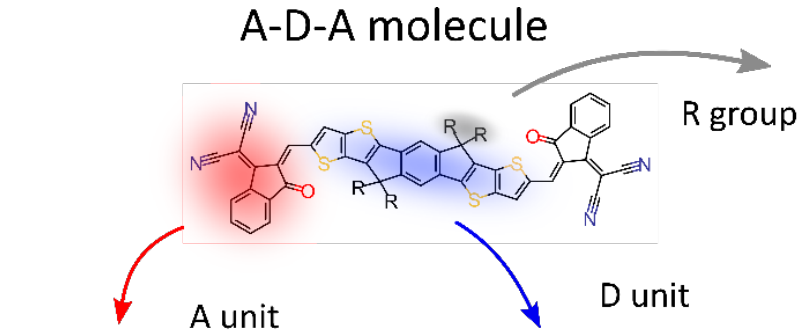


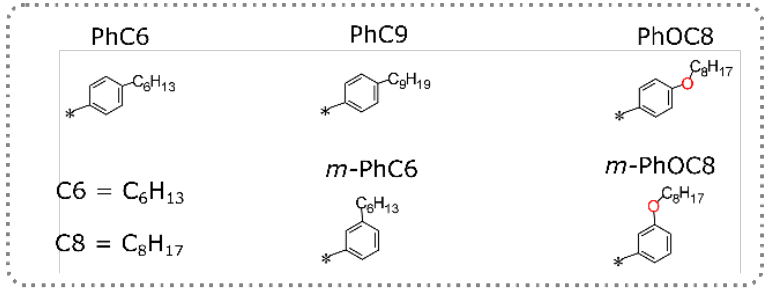
PCE ~ 10%
μe ~ $7x10^{-3}$ cm^2 V^{-1} s^{-1}

- NFAs outperformed fullerenes thanks to their higher absorption of the solar spectrum, energy levels tunability and better stability (despite the lower electron mobility)
- Study of the arrangement of the molecules in the solid-state gives insight about the propensity of the material to build favorable systems in terms of charge transport
- Microstructure-property relations for NFAs

Single-Crystal Growth (Solvent-Vapour Diffusion)

Structure determination by X-Ray Diffraction (XRD)





Molecule	*Motif*	*π-π order*	*Cryst. ref.*
ITIC	Herringbone	0D	This work
4TICO	Herringbone	0D	This work
m -4TICO	Brickwork	2D	This work [3]
IDIC	Brickwork	2D	This work
m -ITIC	Brickwork	2D	This work
ITN-C9	Brickwork	2D	1
ITzN-C9	Brickwork	2D	1
IDIC-4H	Brickwork	2D	2
4TIC	Reticular	3D	3
o-IDTBR	Reticular	3D	4
ITCT-DM	Reticular	3D	5
IEICO	Reticular	3D	This work

Mondelli P. et al. *Materials Horizon* 2020, DOI: 10.1039/c9mh01439j

- New crystals obtained in addition to structures known from literature
- Identification of the packing motif and the dimensionality of the π-π overlap

[1] Swick et al. *PNAS* **2018,** DOI: 10.1073/pnas.1807535115. [2] Qu et al. *ACS Appl Mater Interfaces*, **2018**, 10, 39992-40000. [3] Shi et al. *Chemistry of Materials*, **2017**, 29, 8369-8376.

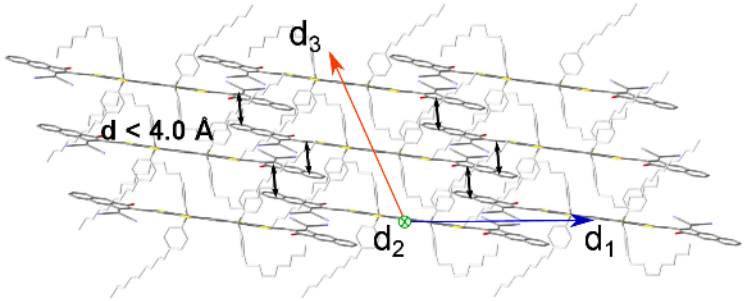
[3] Data courtesy of Sarah Holliday, Imperial College London [5] Yan et al. Advanced Energy Materials, 2018, DOI: 10.1002/aenm.201800204

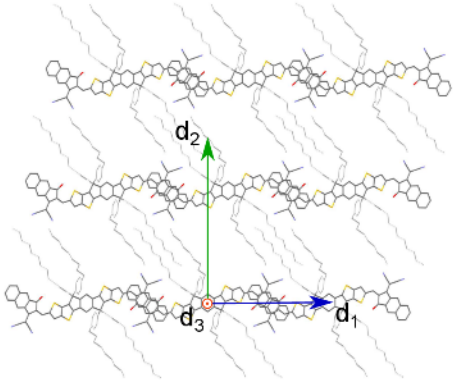
Effect of the Domain Connectivity on the Charge Transport

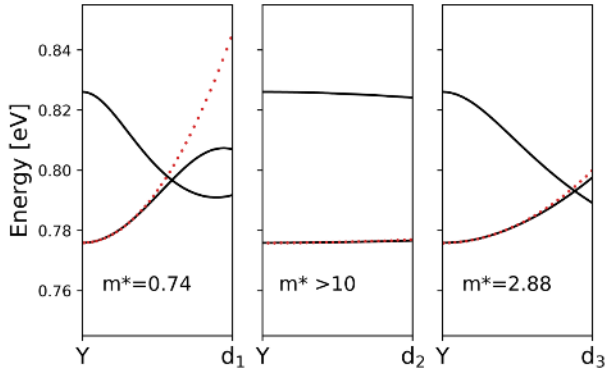
Calculated band structure by DFT and electron effective mass

2D π-π stacking

ITzN-C9

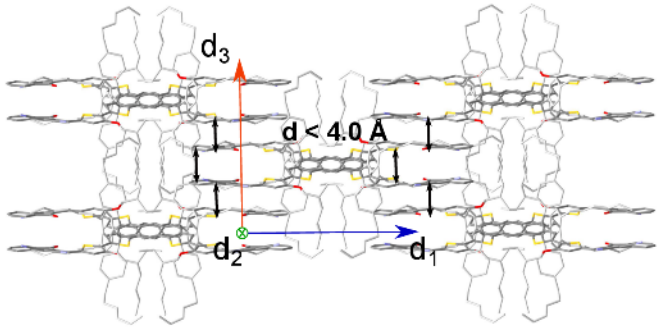


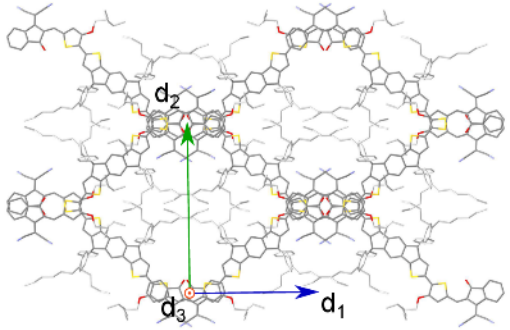


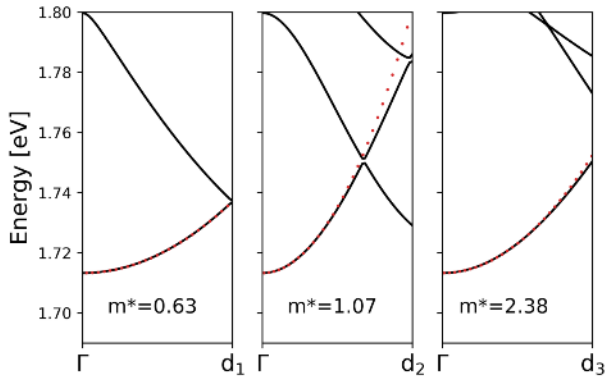


3D π-π stacking

IEICO







- 3D interconnected domains don't present any directional bottleneck for the charge transport
  ⇢ **isotropic transport**

Mondelli P. et al. *Materials Horizon* 2020, DOI: 10.1039/c9mh01439j

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From Single-Crystals to Solar Cells

How can we extend these learnings even further?

i. NFA single crystals

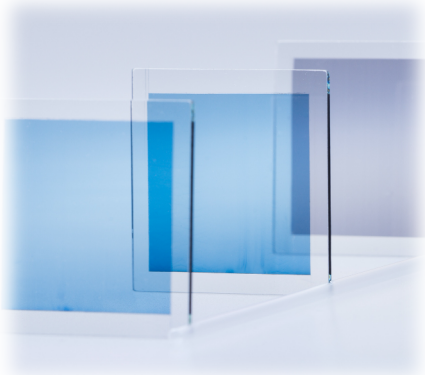
Solid state arrangement and domain anisotropy (DFT)

ii. NFA powder

Polycrystalline ordering (PXRD) and mobility (TRMC and molecular dynamics)

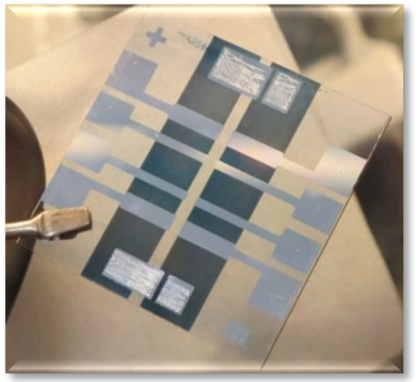
iii. NFA films

Surface morphology (AFM) and film crystallinity (GIWAXS)



iv. Fullerene-free solar cells

Charge mobility (CELIV, RPV TOF) and JV



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Former supervisors and colleagues during my MSc thesis project and 1-year fellowship:

- **Prof Carlo Mariani** (Physics Department, University of Rome "La Sapienza", Rome, Italy)
- **Prof Maria Grazia Betti** (Physics Department, University of Rome "La Sapienza", Rome, Italy)
- **Prof Nunzio Motta** (Science and Engineering Faculty, Queensland University of Technology, Brisbane, Australia)
- **Dr Giulia Avvisati** (Physics Department, University of Rome "La Sapienza", Rome, Italy)

Current supervisors and collaborators:

- **Prof Moritz Riede** (Physics Department, University of Oxford, Oxford, United Kingdom)
- **Prof Dr Carsten Deibel** (Optics and Photonics of Condensed Matter, Technical University of Chemnitz, Chemnitz, Germany)
- **Dr Graham Morse** (PV Physics Fundamentals, Merck Chemicals Ltd, Southampton, United kingdom)
- **Prof Paolo Radaelli** (Physics Department, University of Oxford, Oxford, United Kingdom)
- **Prof David Beljonne** (Chemistry of Novel Materials, University of Mons, Mons, Belgium)
- **Prof Chris-Kriton Skylaris** (Chemistry Department, University of Southampton, Southampton, United Kingdom)