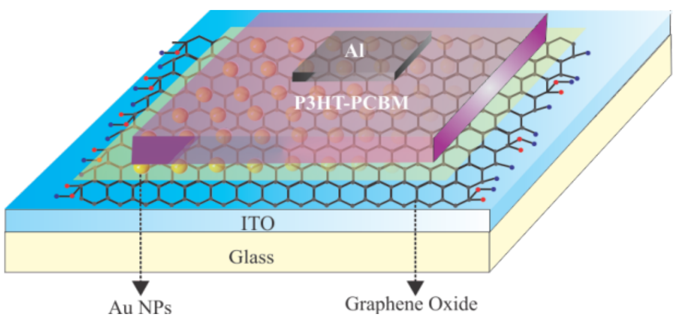
Graphene based HTL

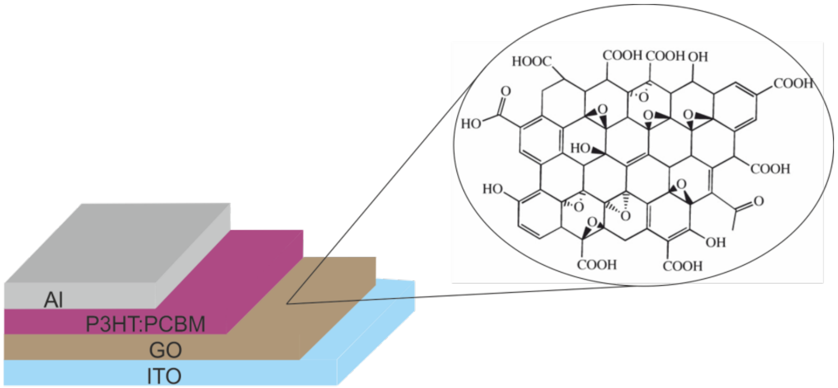
Material	Device Structure	PCE	Used as	Ref.
GO	ITO/GO/P3HT:PCBM	3.6%	HTL	ACS Nano (2010), 4, 3169
pr-GO	ITO/pr-GO/P3HT:PCBM/Ca/Al	3.63	HTL	Adv. Mater. (2011), 23, 4923
GO-SWCNTs(1:0.2)	ITO/GO:SWCNTs/P3HT:PCBM/Ca/Al	4.10	HTL	Adv. Energy Mater. (2011), 1, 1052
GO & CO-Cs	ITO/GO/P3HT:PCBM/GO-Cs/Al	3.67	HTL & ETL	Adv. Mater. (2012), 24, 2228
GO & Au NPs	**ITO/GO/Au_NPs/P3HT:PCBM/Al**	**2.9**	**HTL**	**Nanoscale (2013), 5, 4144**
GO-Cl	**ITO/GO-Cl/PCDTBT:PCBM/TiOx/Al**	**6.56**	**HTL**	**Nanoscale (2014), 6, 6925**

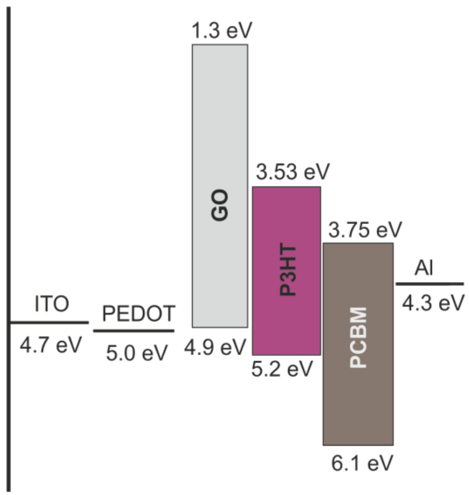


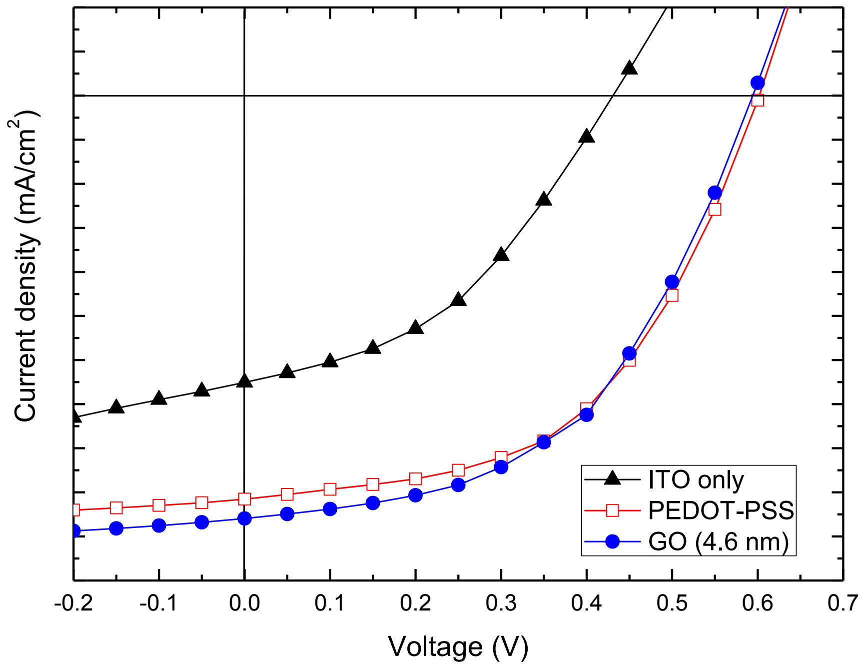
- The plasmonic GO-based devices exhibited a performance enhancement by 30% compared to the devices using the traditional PEDOT:PSS layer.
- They preserved 50% of their initial PCE after ***45 hrs of continuous illumination, contrary to the PEDOT:PSS-based ones that die after 20 hrs***

- ***Main reason➔ Stability improvement***

Photovoltaic Devices - GO replacing PEDOT



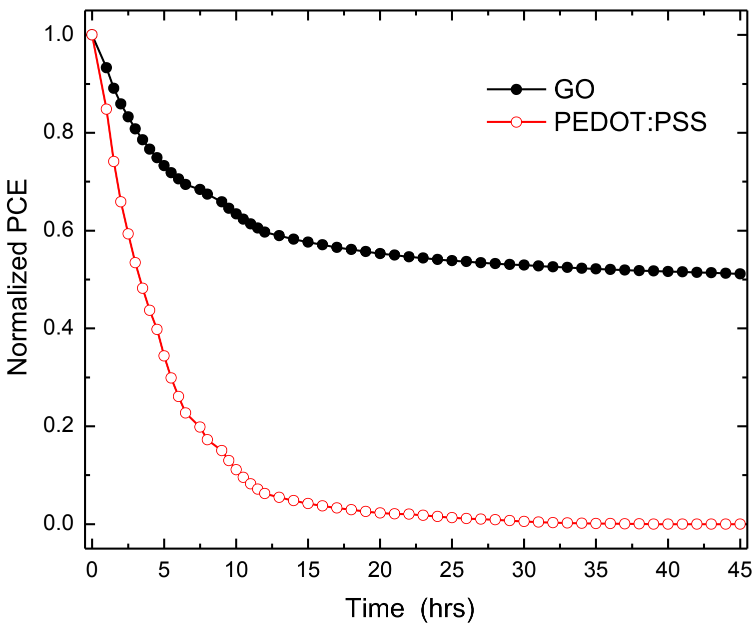


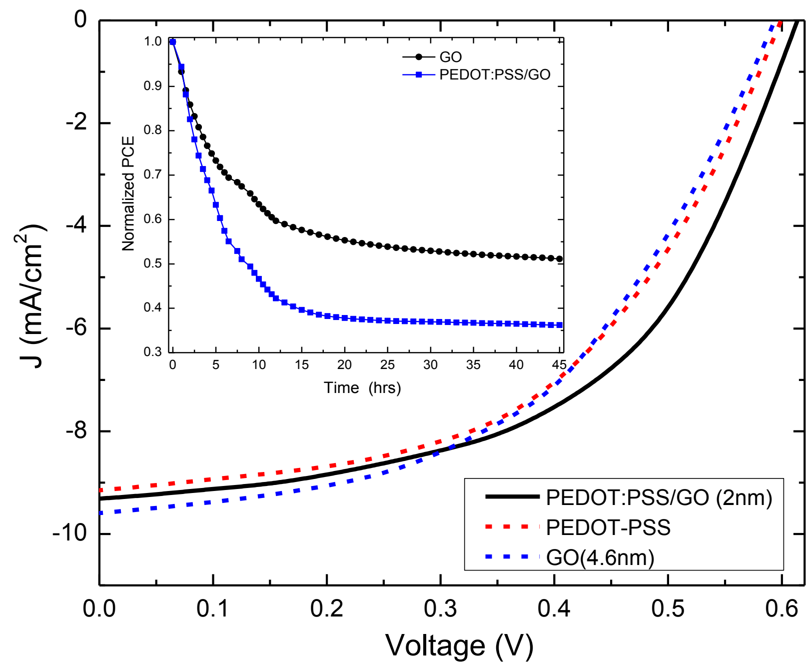


HTL	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
ITO only	0.42	6.51	42.5	1.16
PEDOT:PSS	0.60	9.15	51.7	2.86
GO (2.0 nm)	0.43	6.65	36.2	1.04
GO (2.6 nm)	0.54	9.04	43.8	2.09
GO (4.3 nm)	0.60	9.59	50.7	2.90
GO (5.2 nm)	0.60	8.02	44.1	2.12

Nanoscale (2013), 5, 4144-4150

GO replacing PEDOT – Stability enhancement

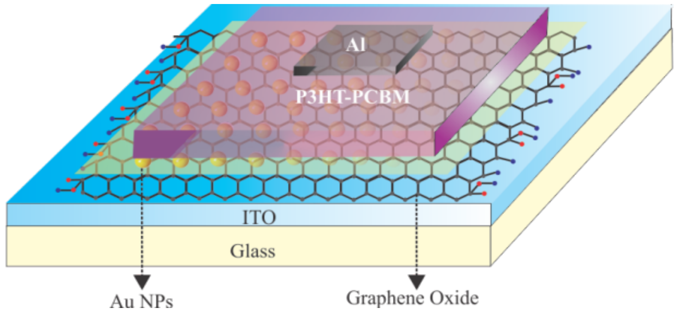


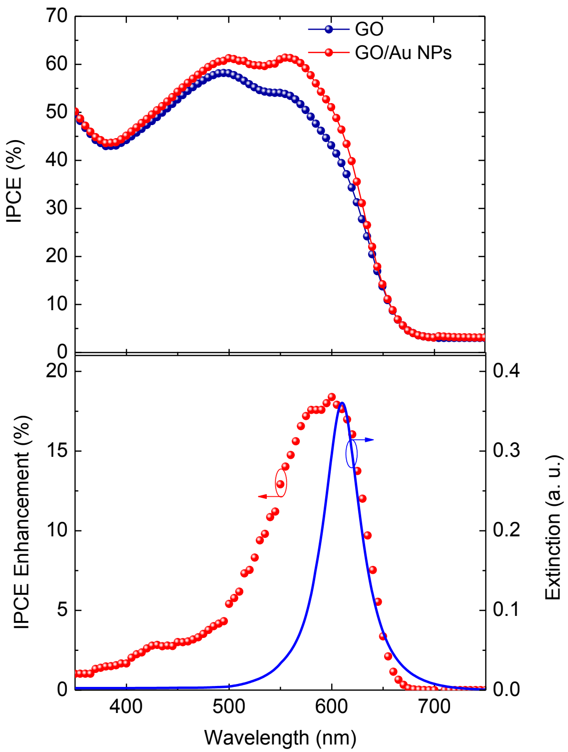


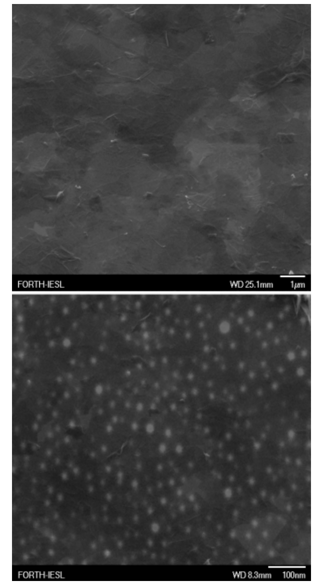
- The devices with GO as the HTL preserve more than 50% of their initial efficiency after 45 hrs of continuous illumination
- The PEDOT:PSS based devices die after 20 hrs

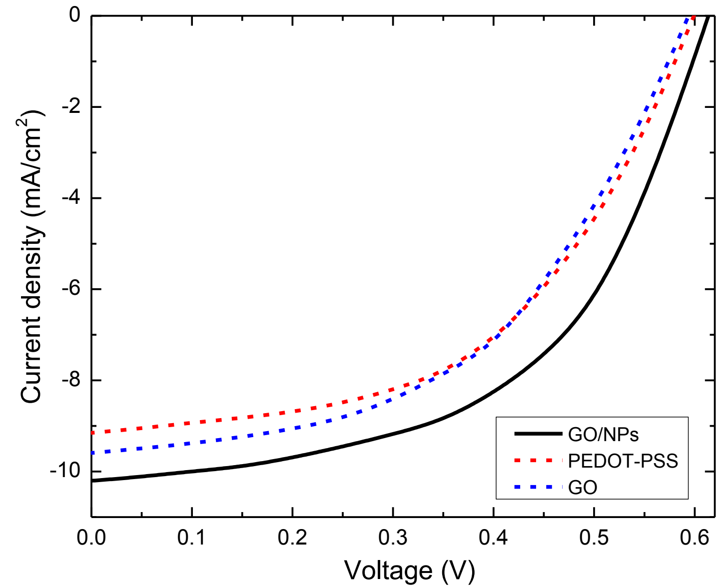
Nanoscale (2013), 5, 4144-4150

Combining Plasmonics with Graphene



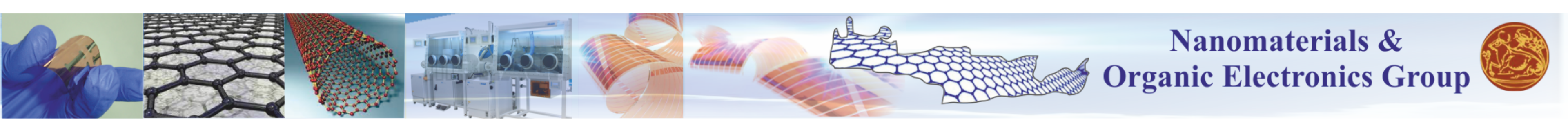




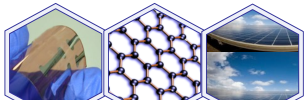


- The plasmonic GO-based devices exhibited a performance enhancement by 30% compared to the devices using the traditional PEDOT:PSS layer.
- They preserved 50% of their initial PCE after 45 hrs of continuous illumination, contrary to the PEDOT:PSS-based ones that die after 20 hrs

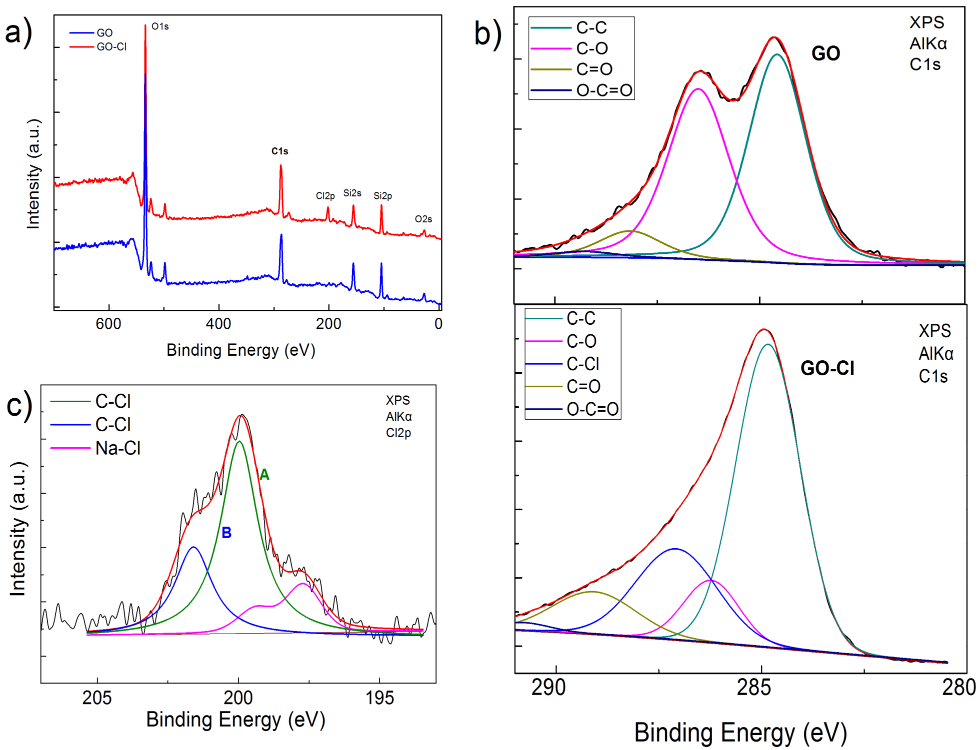
Nanoscale (2013), 5, 4144-4150

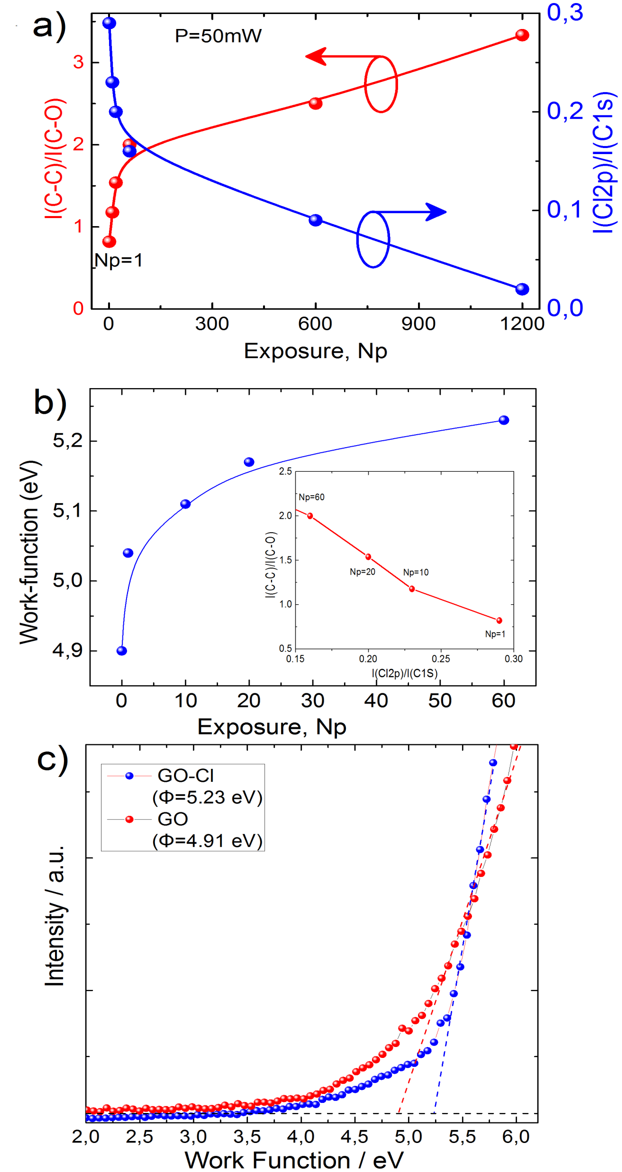


Doped GO replacing PEDOT



- R2r compatible photochemical method for the simultaneously partial reduction and doping of GO films through ultraviolet laser irradiation in the presence of Cl_2 precursor gas
- Photochlorination → grafting of chloride to the edges and the basal plane of GO
- Tailoring of the GO work-function from 4.9 eV to a maximum value of 5.23 eV.

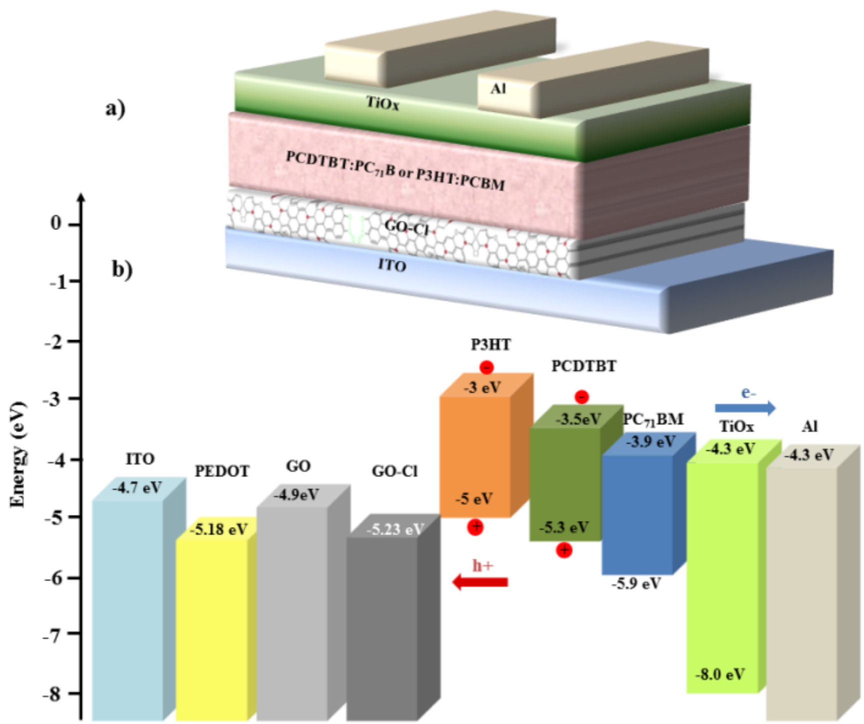


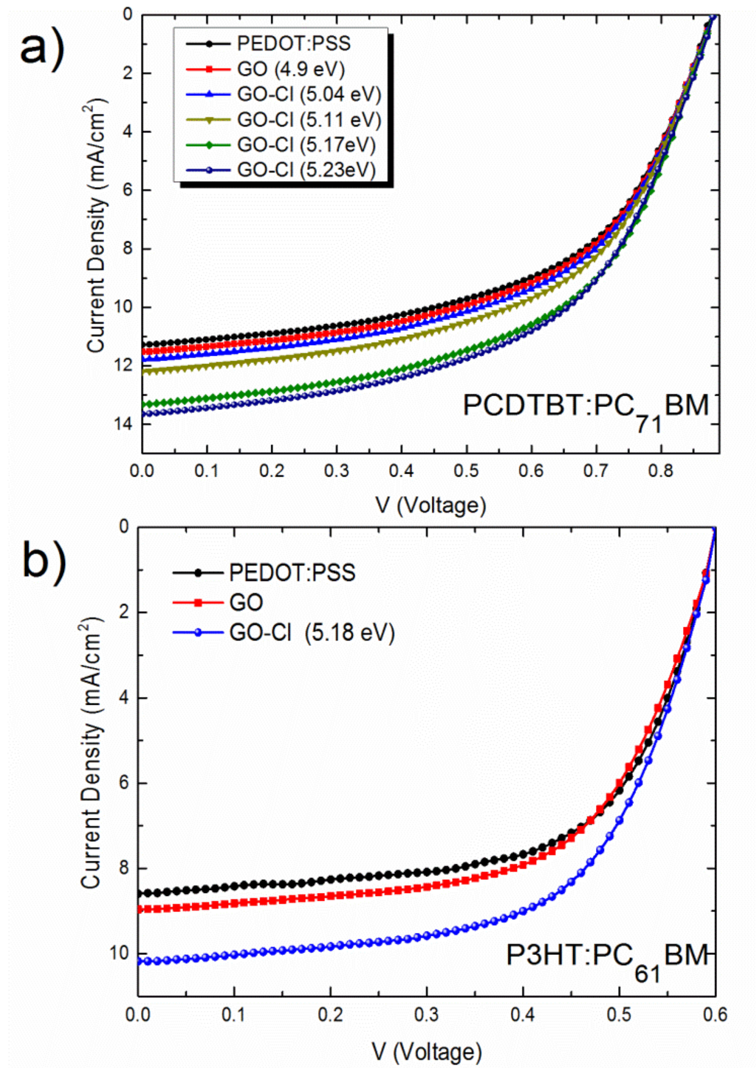


Nanoscale (2014), 6, 6925-6931

Doped GO replacing PEDOT

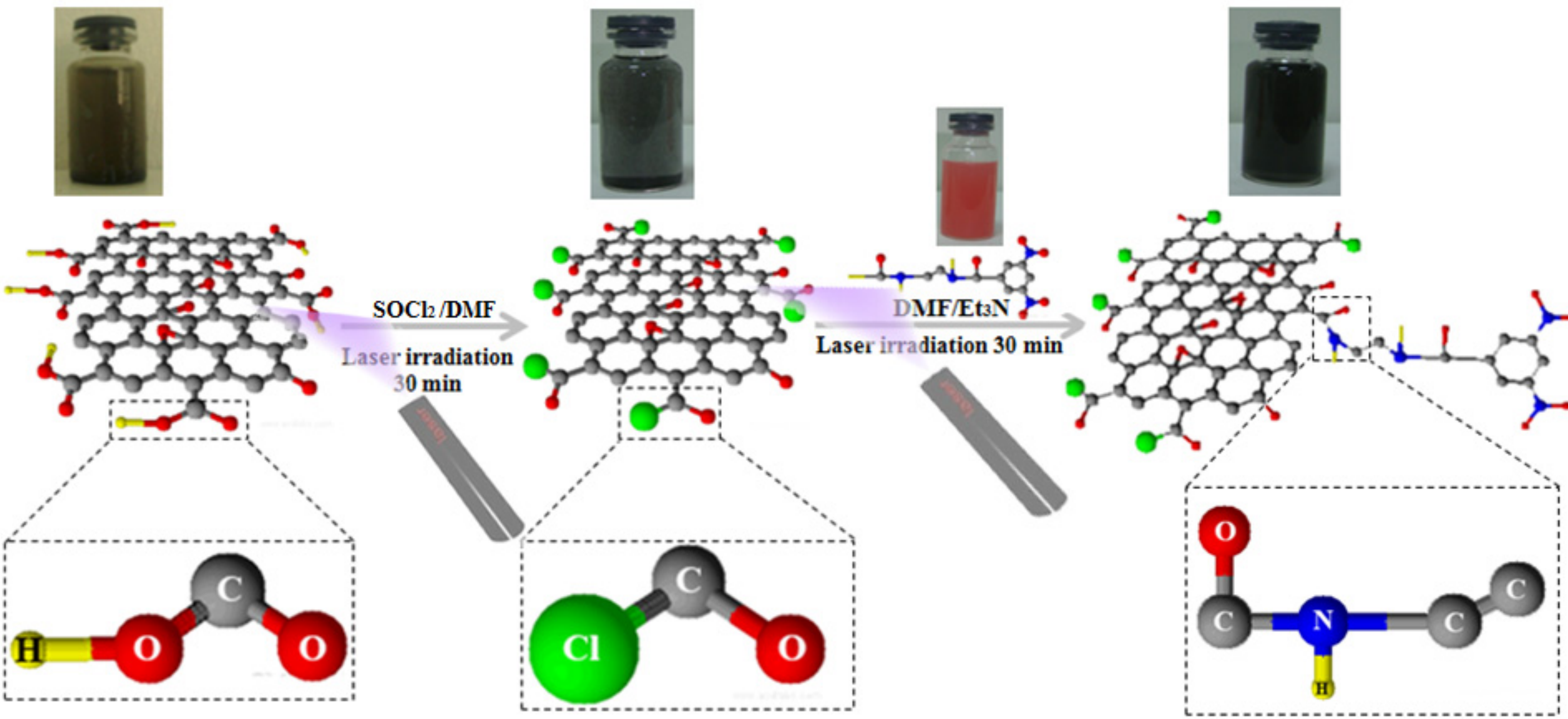
- OPVs with GO-Cl as the hole transporting layer (HTL) were demonstrated with power conversion efficiency (PCE) of 6.56 %
  - ✓ >14.1 than GO based OPVs
  - ✓ > 15.7 % than PEDOT:PSS based OPVs
- The performance enhancement was attributed to more efficient ***hole transportation*** due to the energy levels matching between the GO-Cl and the polymer donor.





Nanoscale (2014), 6, 6925-6931

The Photochemical Synthesis
(Laser assisted)

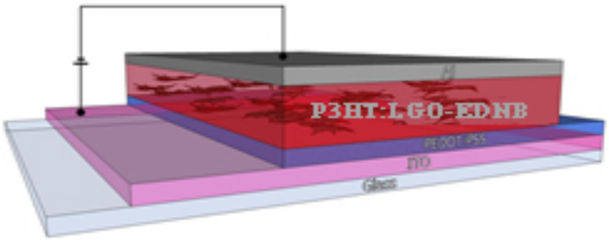


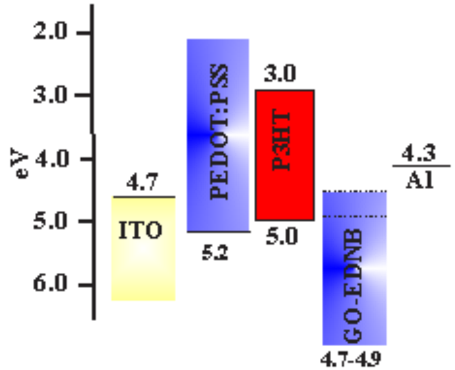
Graphene Oxide (GO) Acylated Graphene Oxide (GO-COCl) LGO-EDNB

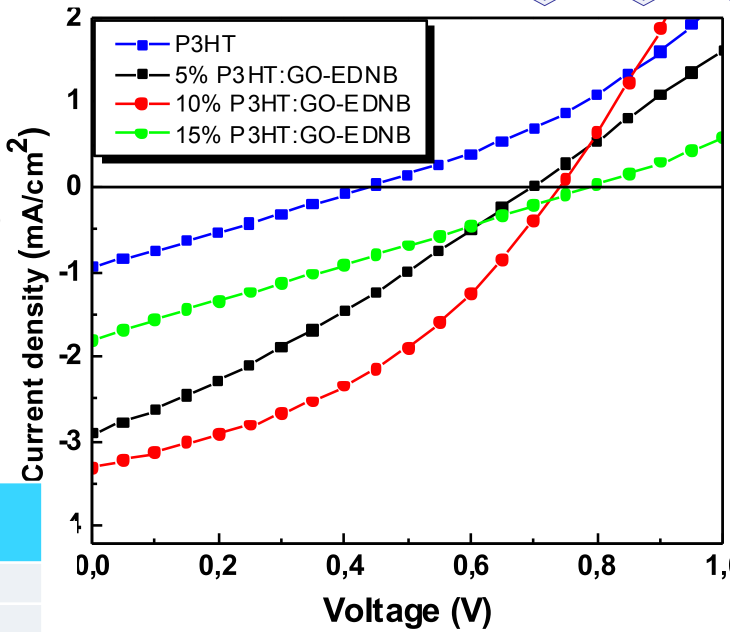
Total time for functionalization: ~ 2 hours !!!!

Advanced Optical Materials 2015, 5, 658-666

Chemical functionalization

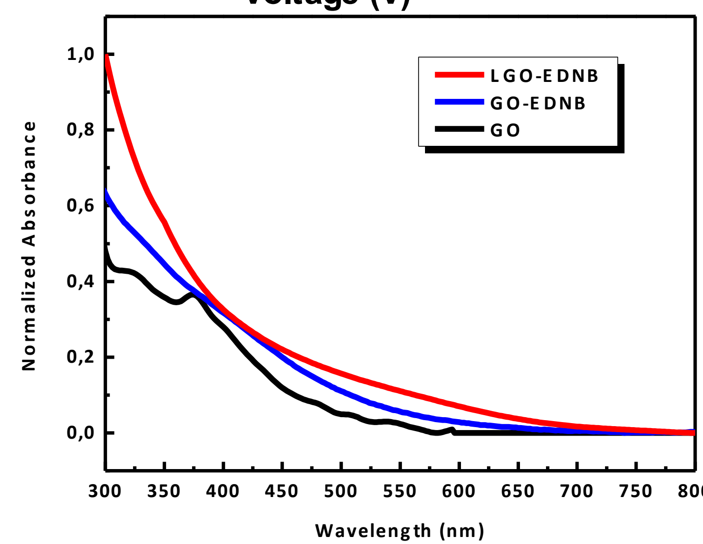




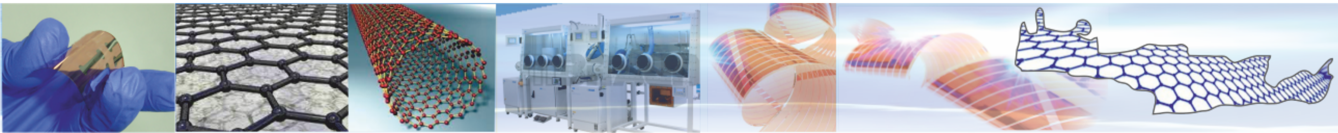


Chemically functionalized GO-EDNB	J_{sc} (mA/cm²)	V_{oc} (V)	FF (%)	PCE (%)
ITO/PEDOT:PSS/P3HT/Al	0.04	0.40	28	0.004
ITO/PEDOT:PSS/P3HT:GO-EDNB (5%)/Al	2.90	0.67	32	0.62
ITO/PEDOT:PSS/P3HT:GO-EDNB(10%)/Al	**3.32**	**0.72**	**40**	**0.96**
ITO/PEDOT:PSS/P3HT:GO-EDNB(15%)/Al	1.78	0.76	28	0.38
Photochemically functionalized LGO-EDNB				
ITO/PEDOT:PSS/PCDTBT:GO-EDNB(5%)/Al	1.99	1.1	25	0.55
ITO/PEDOT:PSS/PCDTBT:GO-EDNB(10%)/Al	3.98	1.09	31	1.34
ITO/PEDOT:PSS/PCDTBT:GO-EDNB(20%)/Al	**5.29**	**1.17**	**39**	**2.41**
ITO/PEDOT:PSS/PCDTBT:GO-EDNB(30%)/Al	2.91	1.12	29	0.95

Voltage (V)

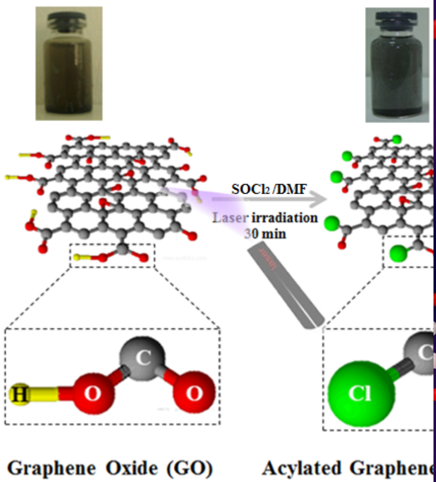


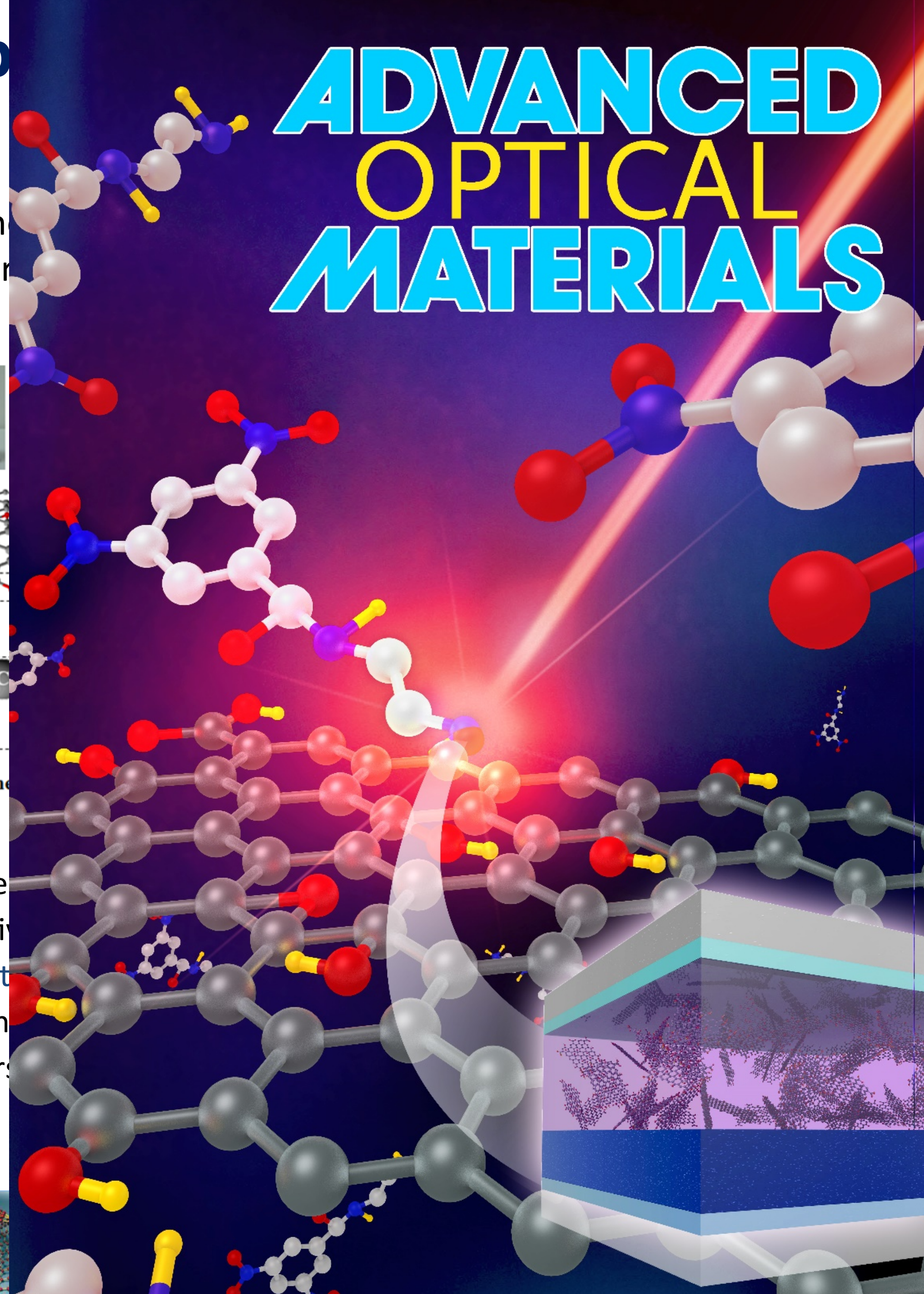
Photochemical functionalization

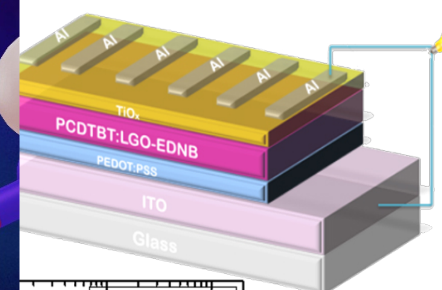


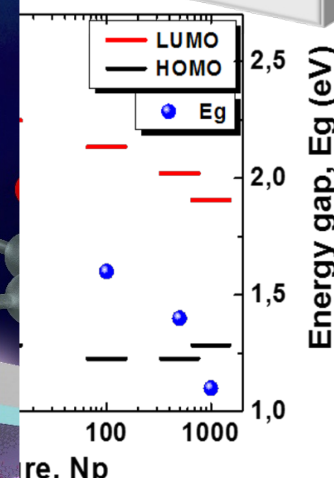
Graphene-b

✓ In summary, a ph dgap graphene-based derivatives GO, thr









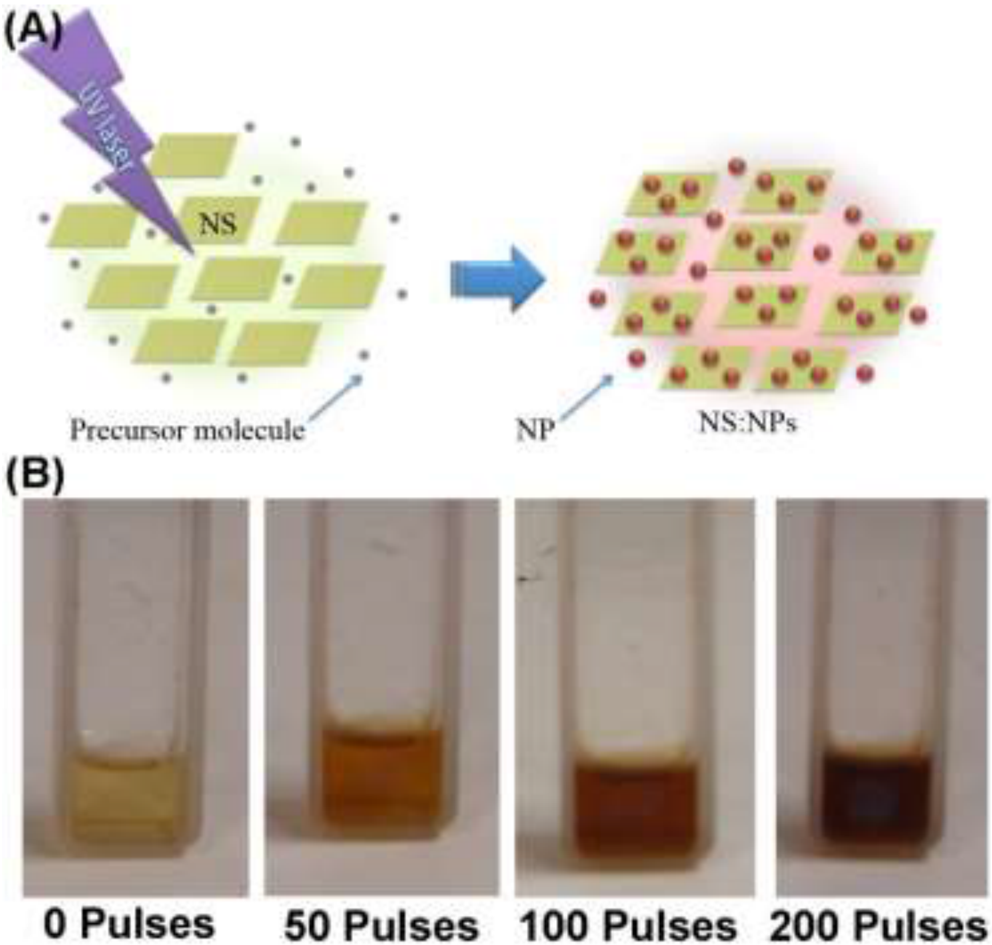
✓ The technique e graphene derivati

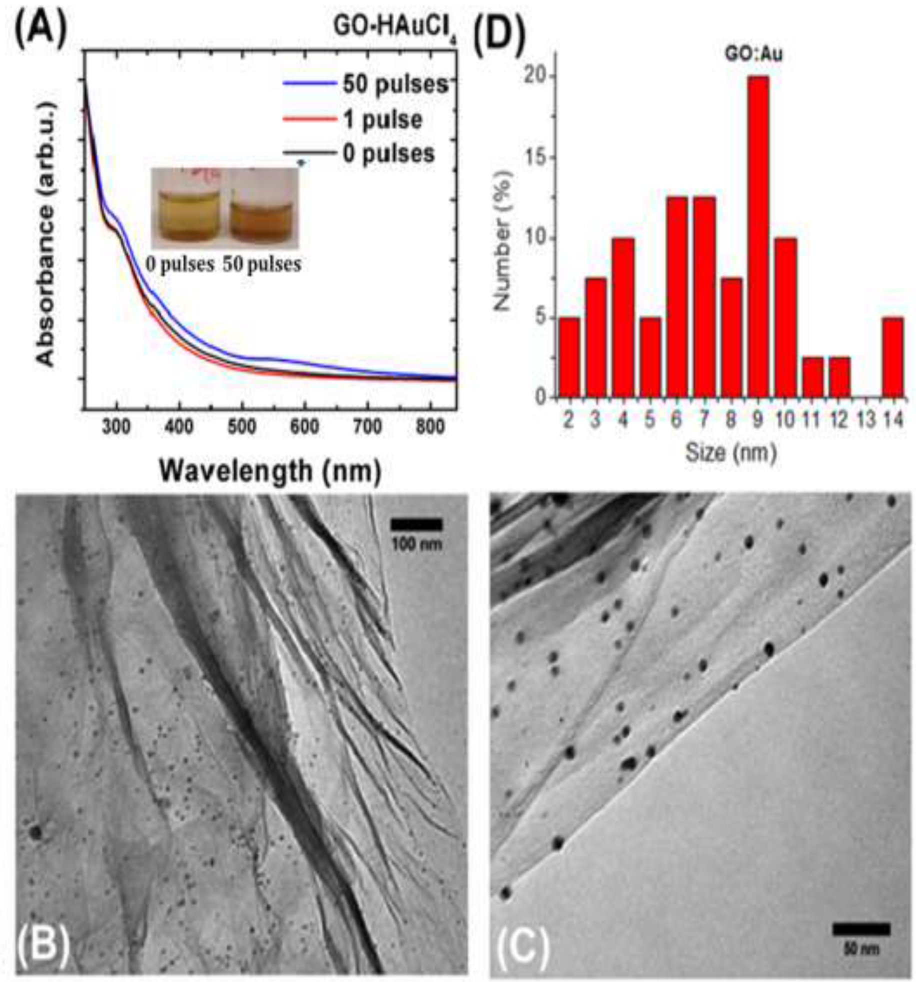
✓ Reduces the react

✓ **PCE of 2.42%,** th electron acceptor

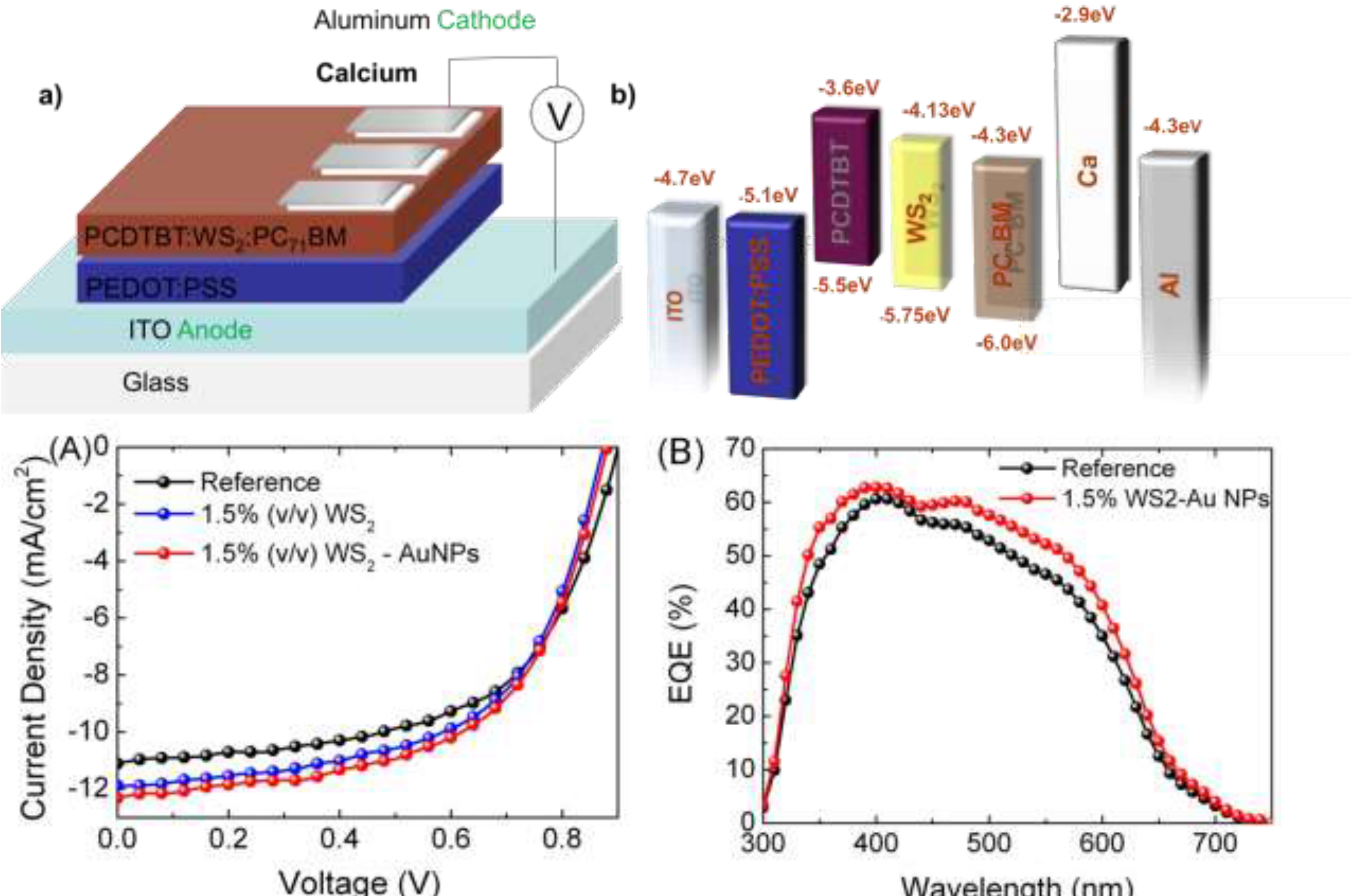
terials (2015), 5, 658-666

2D Materials-based additives in ternary OPVs

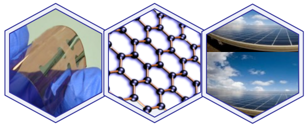




2D Materials-based additives in ternary OPVs

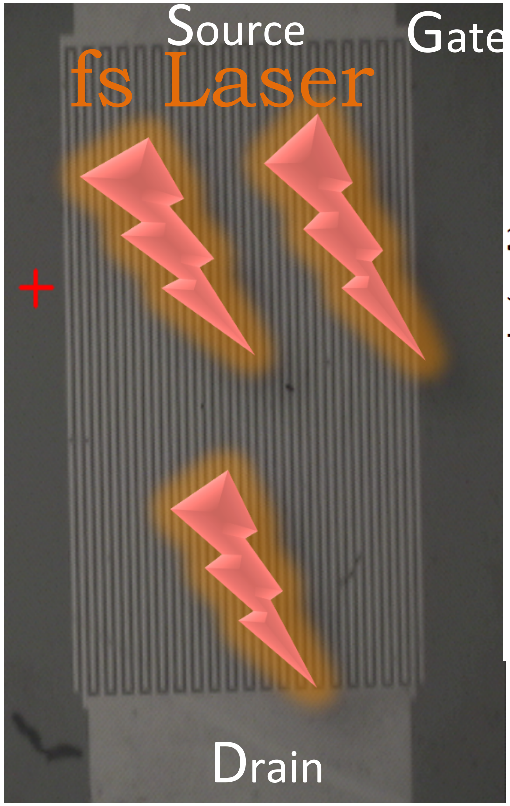


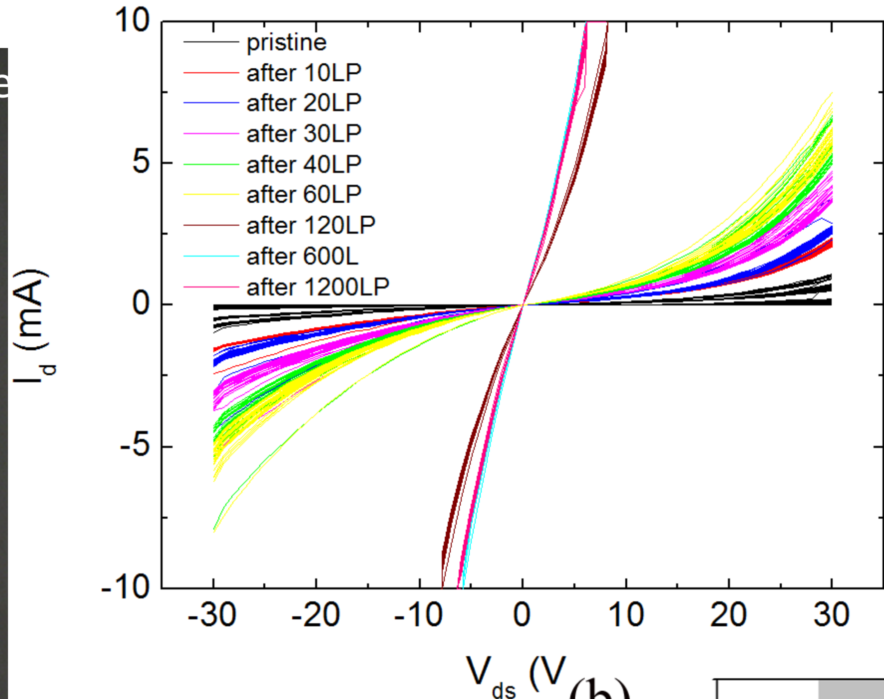
2D Materials-based additives in ternary OPVs

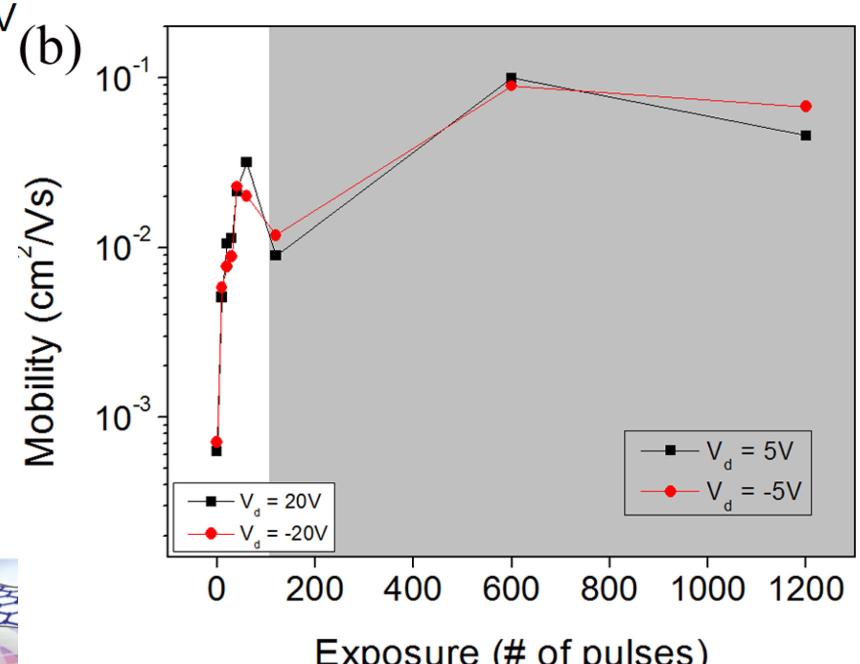


Device structure	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF (%)	PCE (%)
PCDTBT:$PC_{71}BM$	10.6±0.13	0.89±0.1	60.2±0.3	5.6±0.1
1.5% (v/v) WS_2	11.9±0.21	0.87±0.1	59.1±0.2	6.1±0.1
1.0% (v/v) WS_2-Au	11.1±0.18	0.86±0.1	57.3±0.3	5.4±0.2
1.5% (v/v) WS_2-Au	12.3±0.22	0.89±0.1	58.4±0.2	6.3±0.1
2.5% (v/v) WS_2-Au	10.8±0.14	0.86±0.1	57.0±0.4	5.2±0.2

Post-fabrication *in-situ* reduction of GO-based electronic devices







Back-gated GO FET on PET

In collaboration with T. D. Anthopoulos, ICL and G. Eda, NUS

C. Petridis et al., Appl. Phys. Lett. 102, 093115 (2013)

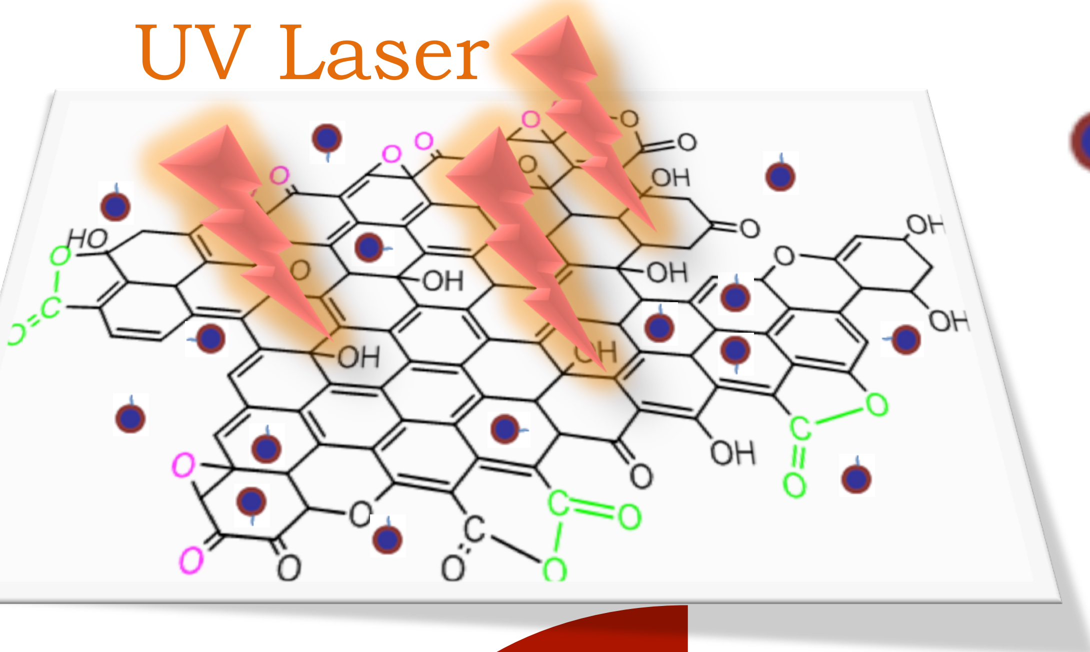
Solution: Chemical modification of reduced GO

Laser based photochemical modification

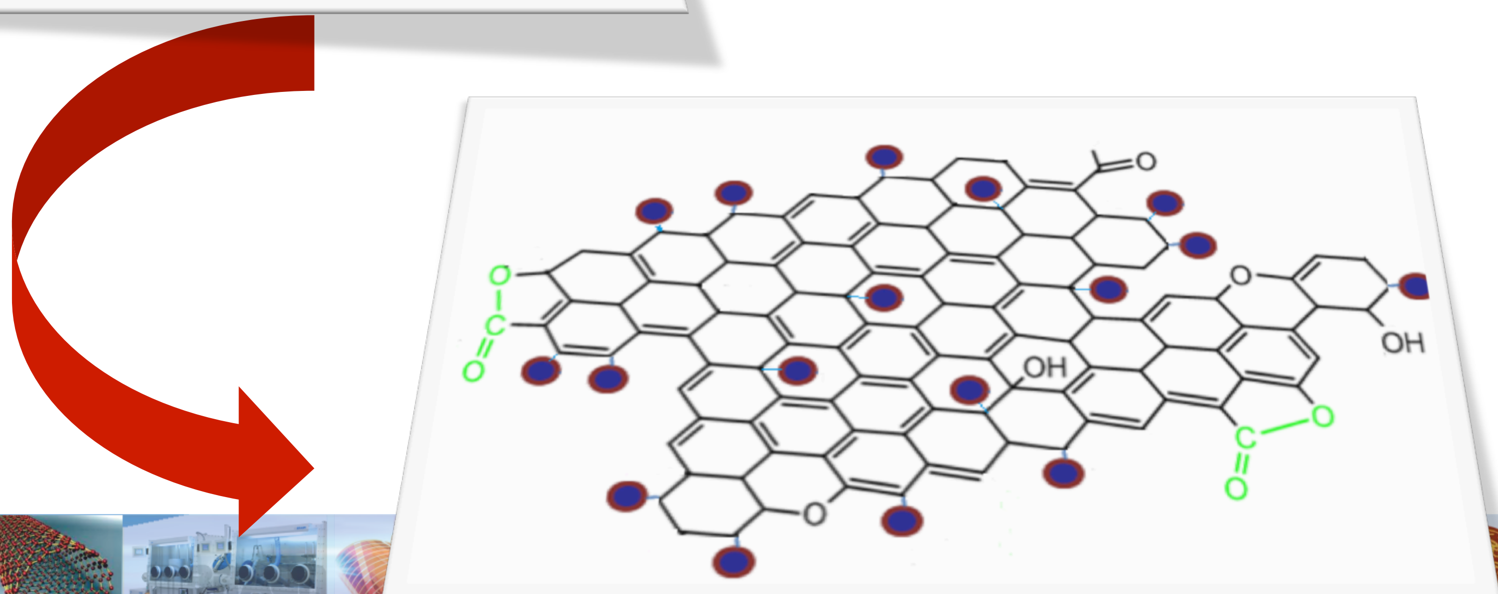
Functionalization	fPOG	→	LrfGO
Doping	dPOG	→	LrdGO

Photochemical functionalization of GO from liquid phase

2

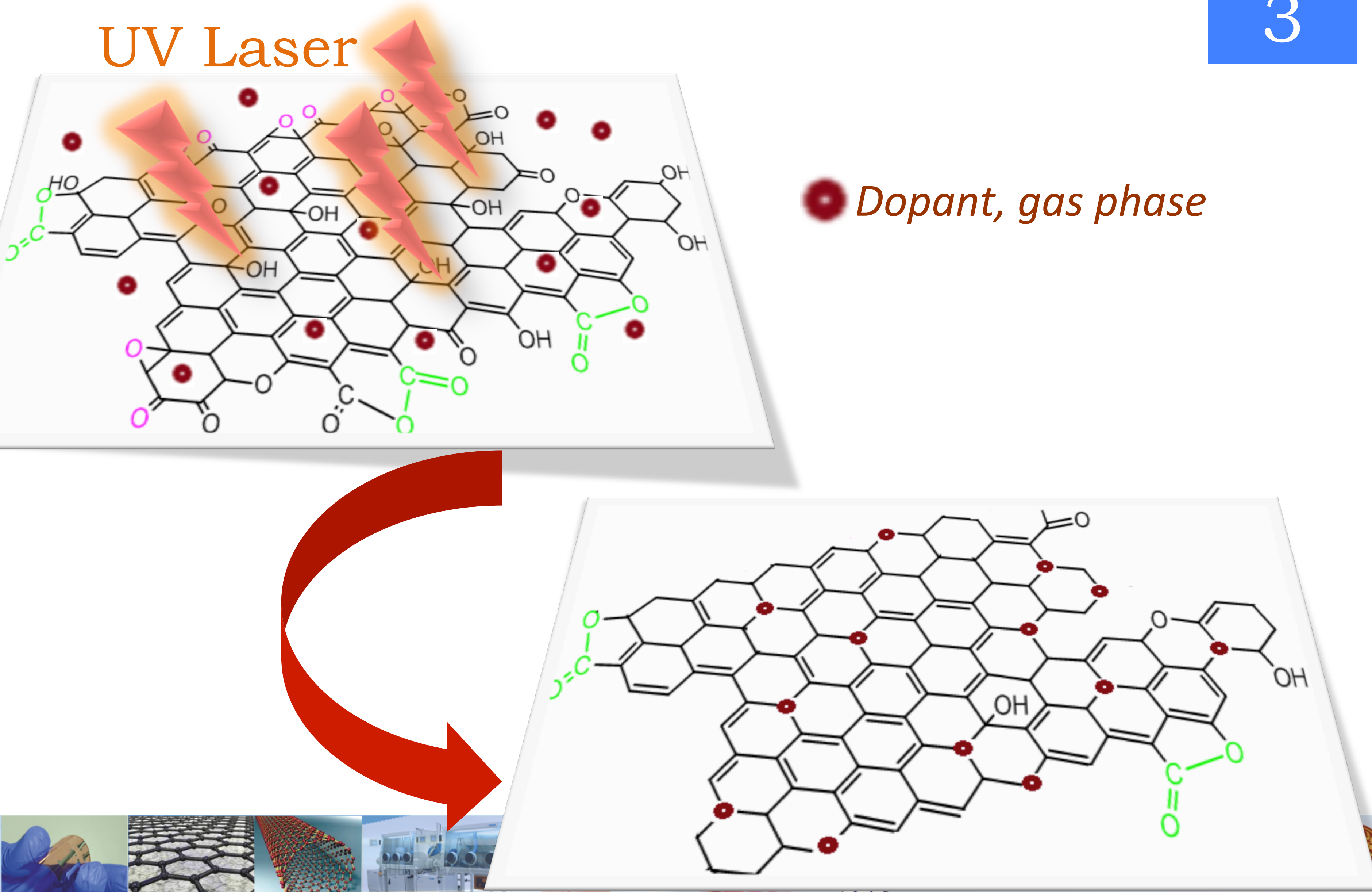


Functional unit,
liquid phase

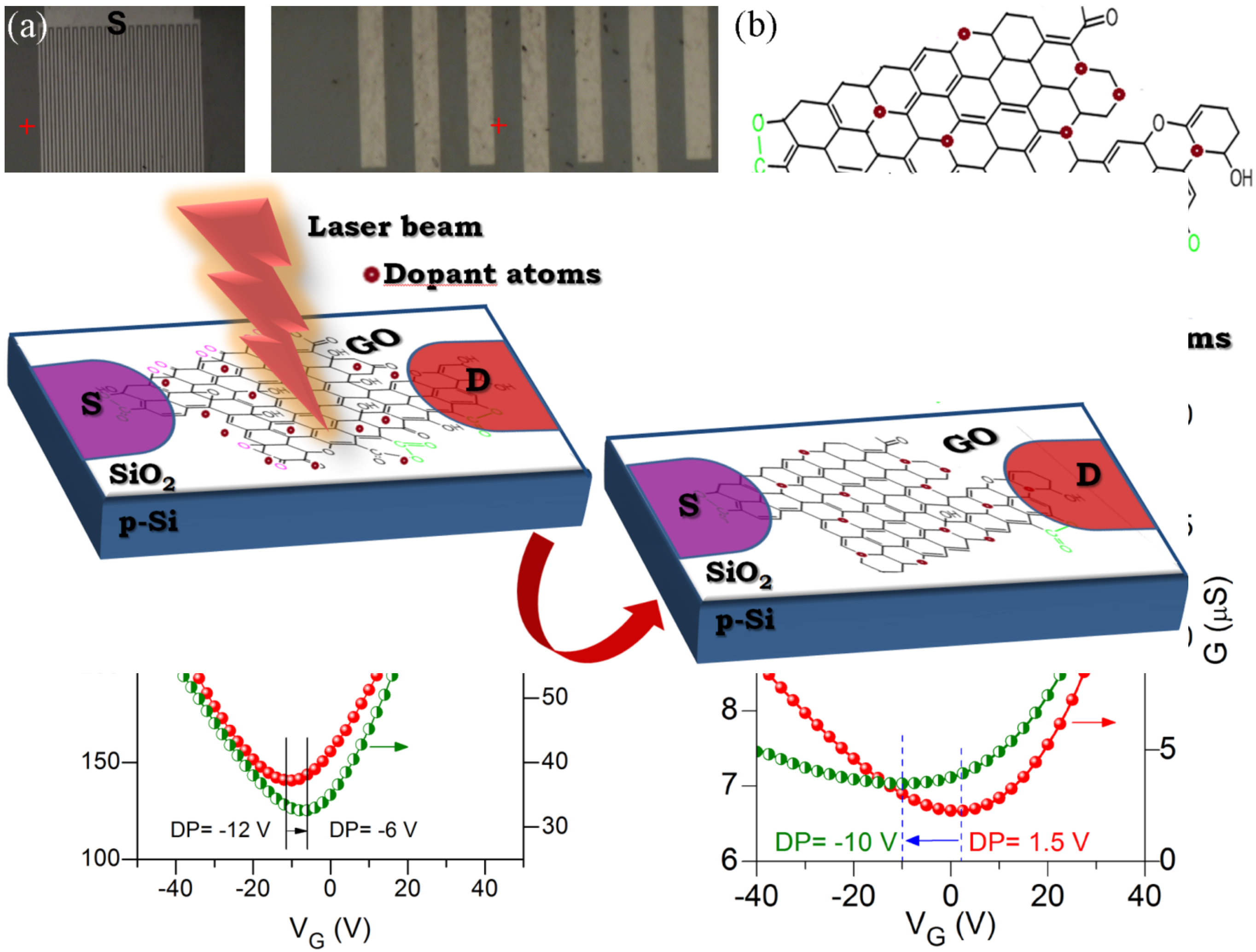


Photochemical doping of GO from gas phase

3



In situ laser doping of GO-FETs

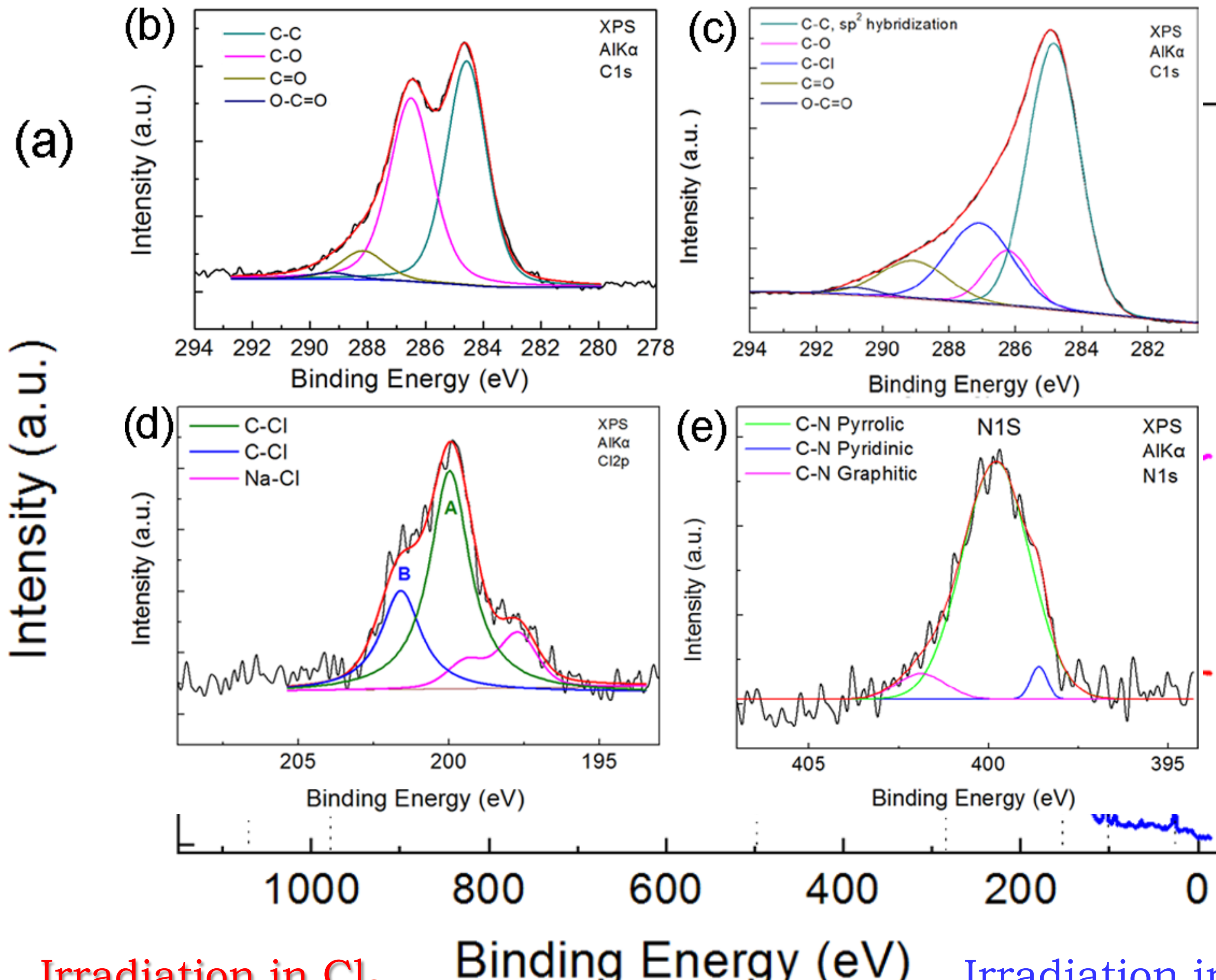


Cl-doping: p-type

N-doping: n-type

Cl and N Doping: Pulsed laser irradiation in precursor gas

3



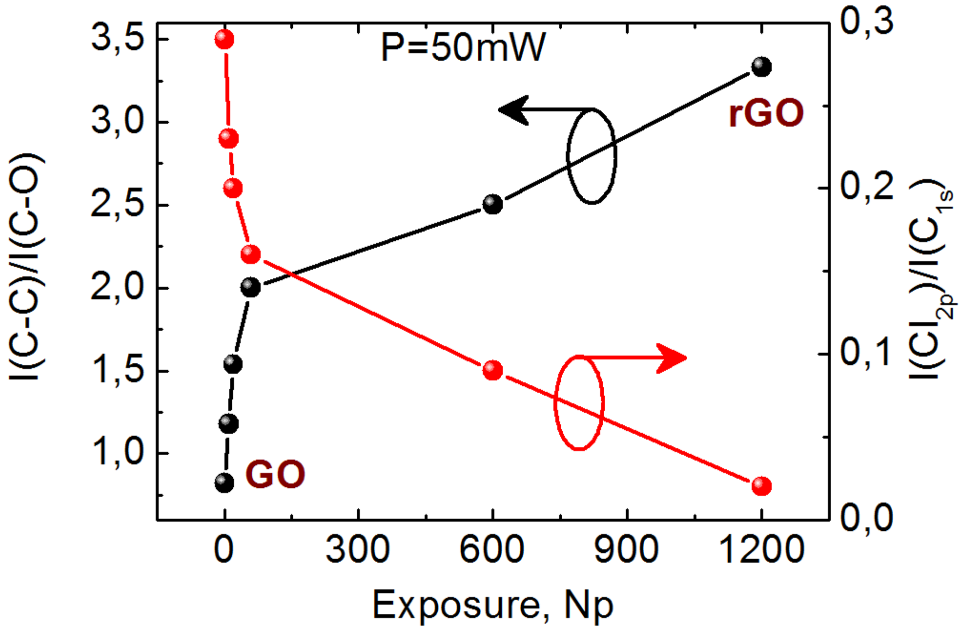
Irradiation in Cl_2

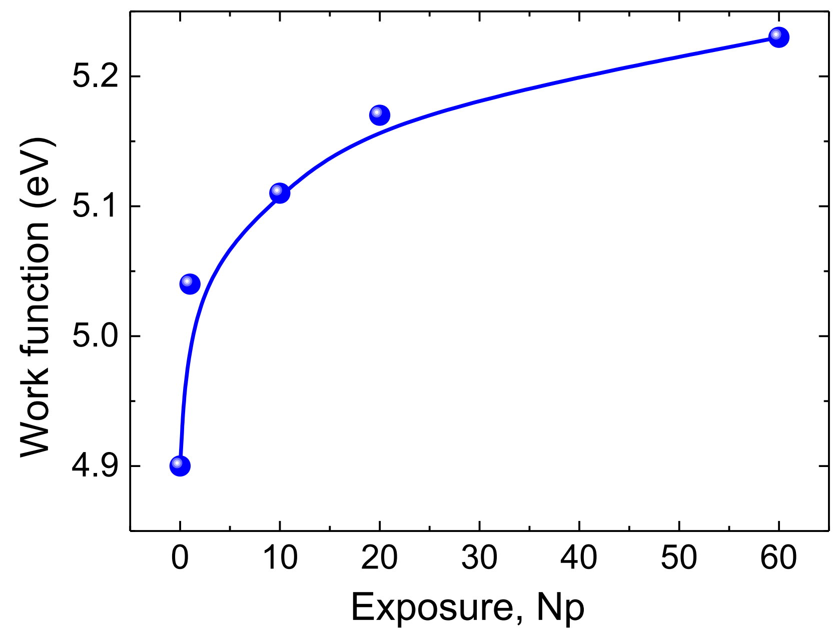
Irradiation in NH_3

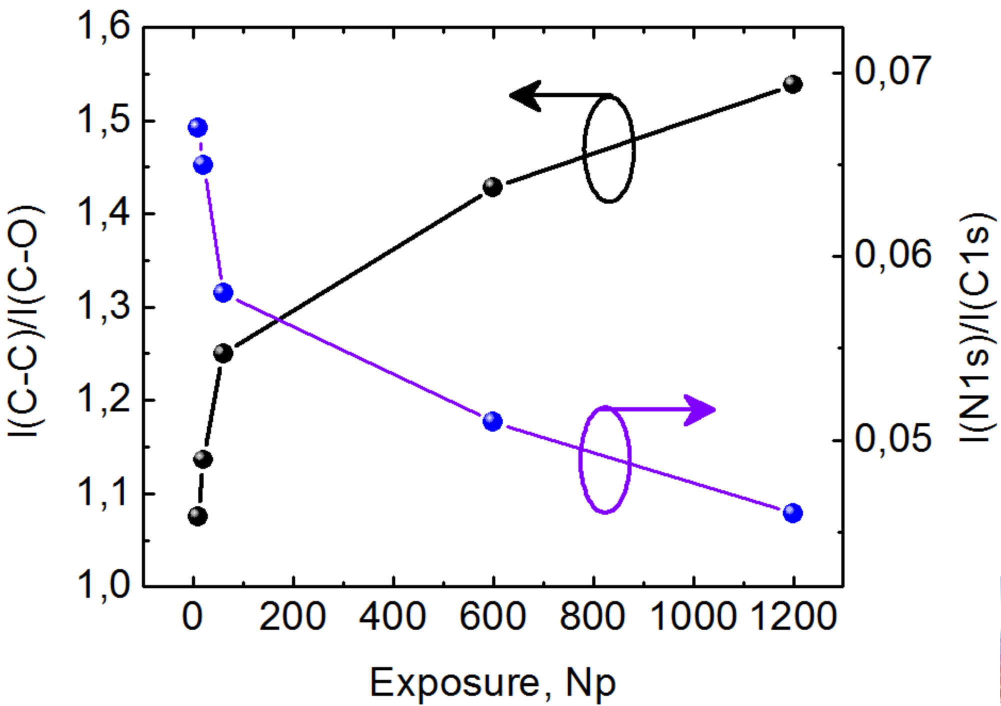
E. Stratakis et al. Nanoscale, (2014), DOI: 10.1039/C4NR01539H

K. Savva et.al. Journal of Materials Chemistry C (2014)

Cl and N Doping: Pulsed laser irradiation in precursor gas



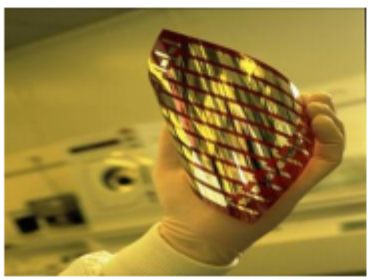


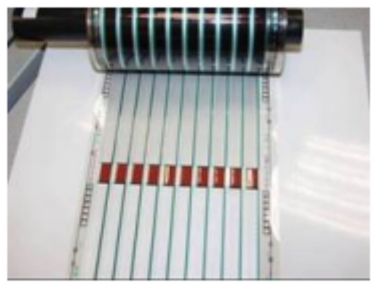


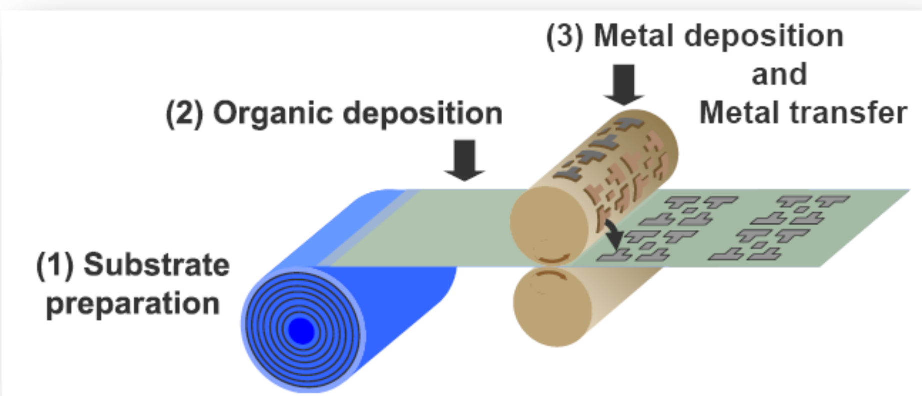
Work Function Tuning !!!

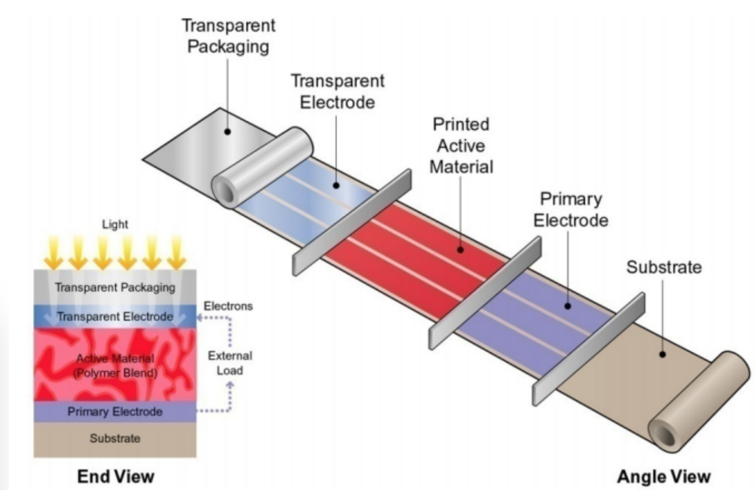
From lab-cells to mass production

- A proposed means to lower the cost of producing flexible displays in a high-volume manufacturing environment by taking advantage of a unique attribute of flexible substrates relative to the traditional glass substrate





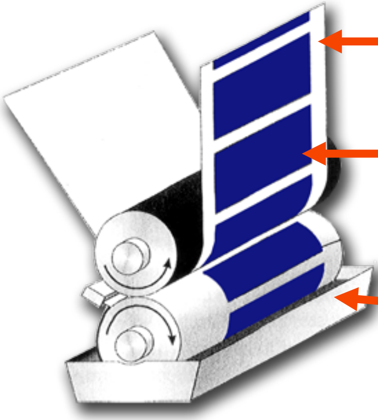




Printing solar cells

"inks" ---- with electronic functionality!

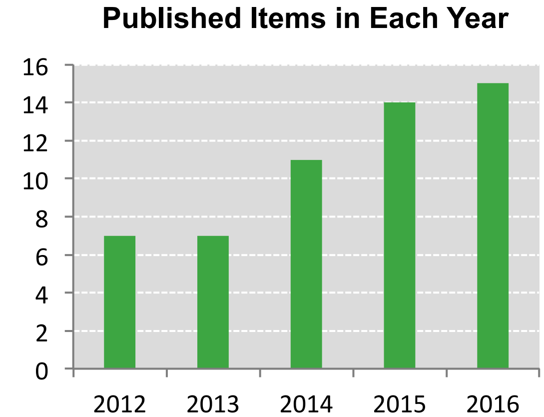
The Dream →



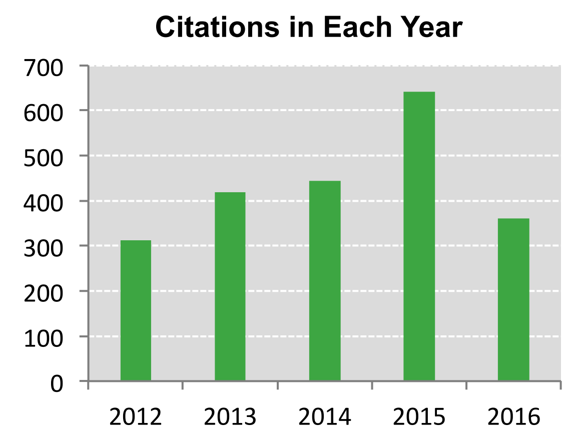
→

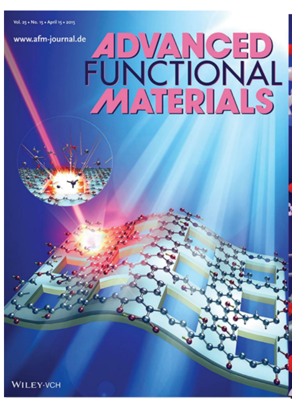
Recent research output

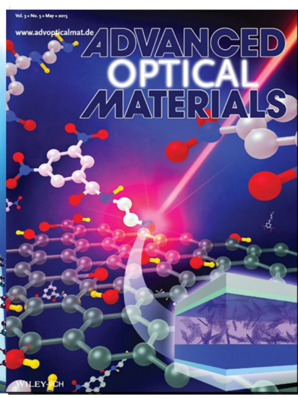
- In situ laser photochemical reduction of graphene oxide for use as the transparent electrode in flexible OPVs
- Chemical functionalization of graphene oxide for use as the electron acceptor or as additives in OPVs
- Combination of graphene oxide with metal nanoparticles for use as the buffer layer in OPVs
- Field emission cathodes based on polymer-graphene and polymer-WS_2 nanotubes electrodes
- Post fabrication laser assisted reduction of GO based FETs

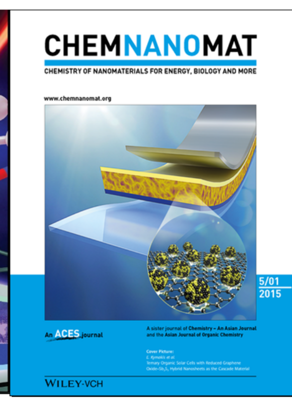


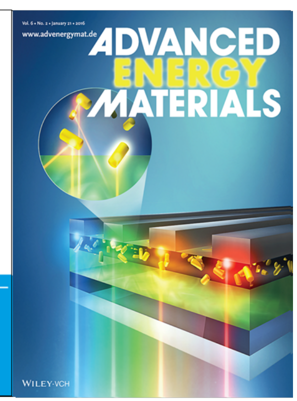
83 publications
4.100 citations
475,18 Impact points





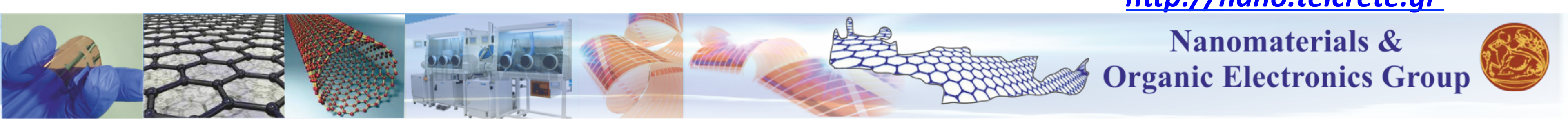






Center of Advanced Materials & Photonics

http://nano.teicrete.gr



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Flexible Organic Photovoltaic Cells with In Situ Nonthermal Photoreduction of Spin-Coated Graphene Oxide Electrodes

Emmanuel Kymakis, Kyriaki Savva, Minas M. Stylianakis, Costas Fotakis, and Emmanuel Stratakis**

The first reduction methodology, compatible with flexible, temperature-sensitive substrates, for the production of reduced spin-coated graphene oxide (GO) electrodes is reported. It is based on the use of a laser beam for the in situ, non-thermal, reduction of spin-coated GO films on flexible substrates over a large area. The photoreduction process is one-step, facile, and is rapidly carried out at room temperature in air without affecting the integrity of the graphene lattice or the flexibility of the underlying substrate. Conductive graphene films with a sheet resistance of as low as 700 Ω sq^{-1} and transmittance of 44% can be obtained, much higher than can be achieved for flexible layers reduced by chemical means. As a proof of concept of our technique, laser-reduced GO (LrGO) films are utilized as transparent electrodes in flexible, bulk heterojunction, organic photovoltaic (OPV) devices, replacing the traditional ITO. The devices displayed a power-conversion efficiency of 1.1%, which is the highest reported so far for OPV device incorporating reduced GO as the transparent electrode. The in situ non-thermal photoreduction of spin-coated GO films creates a new way to produce flexible functional graphene electrodes for a variety of electronic applications in a process that carries substantial promise for industrial implementation.

conductive and optically transparent, electrode materials of next-generation flexible electronic devices are required to be robust under extensive bending and compatible with large-scale manufacturing.[2] Indium tin oxide (ITO) is the currently leading transparent conductive electrode in rigid optoelectronic devices.[3] However, ITO cannot fulfi ll such requirements, since it is brittle and cracks under bending or stretching,[4] and on top of that it is practically expensive, since it suffers from both the indium scarcity and the sputter deposition line expenses.[5] Furthermore, device issues like instability of ITO toward acidic or basic conditions[6] and mechanical brittleness[7] limit the applicability of ITO in flexible electronics. A flexible substitutive material for ITO with a similar performance but lower cost is evidently needed. Recent research has focused on the development of thin layers of highly

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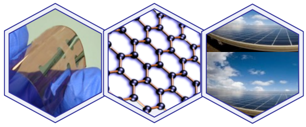

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In situ photo-induced chemical doping of solution-processed graphene oxide for electronic applications†

K. Savva,[a] Y.-H. Lin,[b] C. Petridis,[cd] E. Kymakis,[d] T. D. Anthopoulos*[b] and E. Stratakis*[ae]

We developed a photochemical method for the simultaneous reduction and doping of graphene oxide (GO) layers through ultraviolet laser irradiation in the presence of a dopant precursor gas. It is shown that a few seconds of irradiation is sufficient to dope the GO lattice, while the doping and reduction levels can be readily controlled upon variation of the irradiation time. Using this method, the simultaneous reduction and doping of GO with chlorine or nitrogen atoms is achieved and confirmed by Raman, FTIR and X-ray photoelectron (XPS) spectroscopy measurements. To demonstrate the potential of the approach for practical applications, the photochemical method was successfully employed for the in situ laser

tions in a process that carries substantial promise for industrial implementation.

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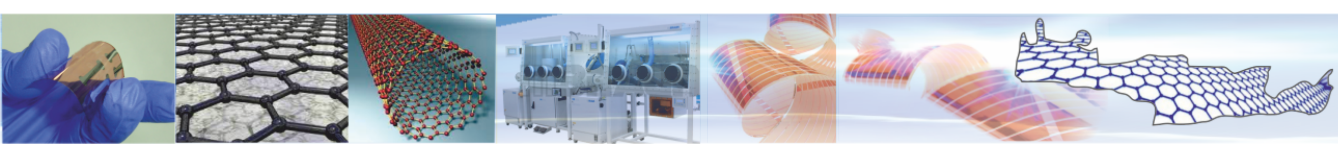

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Reduced Graphene Oxide Micromesh Electrodes for Large Area, Flexible, Organic Photovoltaic Devices

Dimitrios Konios, Constantinos Petridis, George Kakavelakis, Maria Sygletou, Kyriaki Savva, Emmanuel Stratakis, and Emmanuel Kymakis**

A laser-based patterning technique—compatible with flexible, temperature-sensitive substrates—for the production of large area reduced graphene oxide micromesh (rGOMM) electrodes is presented. The mesh patterning can be accurately controlled in order to significantly enhance the electrode transparency, with a subsequent slight increase in the sheet resistance, and therefore improve the tradeoff between transparency and conductivity of reduced graphene oxide (rGO) layers. In particular, rGO films with an initial transparency of ≈20% are patterned, resulting in rGOMMs films with a ≈59% transmittance and a sheet resistance of ≈565 Ω sq^{-1}, that is significantly lower than the resistance of ≈780 Ω sq^{-1}, exhibited by the pristine rGO films at the same transparency. As a proof-of-concept application, rGOMMs are used as the transparent electrodes in flexible organic photovoltaic (OPV) devices, achieving power conversion efficiency of 3.05%, the highest ever reported for flexible OPV devices incorporating solution-processed graphene-based electrodes. The controllable and highly reproducible laser-induced patterning of rGO hold enormous promise for both rigid and flexible large-scale organic electronic devices, eliminating the lag between graphene-based and indium–tin oxide electrodes, while providing conductivity and transparency tunability for next generation flexible electronics.

Being the first layer of an OPV device that comes in contact with the light, the transparent conductive electrode (TCE) is a vital determining factor to the device power conversion efficiency (PCE). Recently, OPV devices with efficiencies above 10% were certified, with their operation lifetimes exceeding industrially interesting levels.[4,5] Indium–tin oxide (ITO) is currently the dominant material used as TCE in rigid optoelectronic devices owing to its high transparency, T_r, in the visible spectrum and its good conductivity.[6] However, considering the employment of OPVs in everyday applications, the electrodes should be inexpensive, lightweight, and highly elastic in order to conserve their electrical properties under high stresses. In this context, ITO suffers from considerable limitations. First, it is expensive due to both the scarcity of indium reserves and the sputter deposition line expenses and second is not flexible, since its polycrystalline microstructure is brittle and cracks

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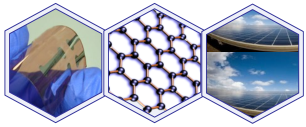

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Laser-Assisted Reduction of Graphene Oxide for Flexible, Large-Area Optoelectronics

Emmanuel Kymakis, *Member, IEEE*, Constantinos Petridis, Thomas D. Anthopoulos, and Emmanuel Stratakis

(Invited Paper)

***Abstract*—This paper reviews recent work on the development and use of a low-temperature, laser-based method for the efficient reduction of graphene oxide (GO) films. The method utilizes a laser beam for the *in-situ* and nonthermal reduction of solution-processed GO layers onto arbitrary substrates. Compared to other reduction techniques, it is single-step, facile, and can be performed at room temperature in ambient atmosphere without affecting the integrity of the either the graphene lattice or the physical properties of the underling substrate. Using this method, conductive layers of reduced GO with a sheet resistance down to ∼700 Ω/sq, are obtained. This is much lower than sheet resistance values reported previously for GO layers reduced by chemical means. As a proof of concept, laser-reduced GO layers were successfully utilized as the transparent anode electrodes in flexible bulk-heterojunction OPVs and as the channel material in field-effect transistors. To the best of our knowledge, this is the only example of an *in-situ*, postfabrication method for the reduction of GO and its implementation in fully functional opto/electronic devices. The nonthermal nature of the method combined with its simplicity and scalability, makes it very**

pensive flexible substrate materials, as well as control of their electrical, magnetic, and optical properties [2], [3].

Although the family of solution-processable semiconductors includes both organic as well as inorganic materials, the branch of electronics based on them is often called organic/plastic electronics [4]. Flexible electronics [5] have been a growing part of research and development in organic electronics due to their expanding applications, including touch screens, optical displays, photovoltaics, lighting devices, and sensors [6]. This technology is based on the controlled deposition and/or printing of different solution-processed layers that form the various device components, onto mechanically flexible substrates [5]. A critical requirement for this technology is that the fabrication processes must be compatible with the nominally low-temperature plastic materials that are being considered for the substrates. In addition to the electrical and optical properties, the ideal solution-

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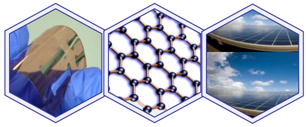

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Solution processed reduced graphene oxide electrodes for organic photovoltaics

Constantinos Petridis,*[ab] Dimitrios Konios,[ac] Minas M. Stylianakis,[a] George Kakavelakis,[a] Maria Sygletou,[d] Kyriaki Savva,[d] Pavlos Tzourmpakis,[a] Miron Krassas,[a] Naoum Vaenas,[a] Emmanuel Stratakis[d] and Emmanuel Kymakis[a]

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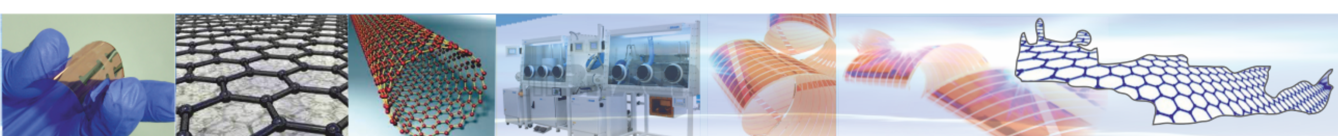
Since the isolation of free standing graphene in 2004, graphene research has experienced a phenomenal growth. Due to its exceptional electronic, optical and mechanical properties, graphene is believed to be the next wonder material for optoelectronics. The enhanced electrical conductivity, combined with its high transparency in the visible and near-infrared regions of the spectrum, enabled graphene to be an ideal low cost indium-tin oxide (ITO) substitute. Solution-processed reduced graphene oxide combines the unique optoelectrical properties of graphene with large area deposition and flexible substrates rendering it compatible with roll-to-roll manufacturing methods. This paper provides an overview of recent research progress in the application and consequent physical–chemical properties of solution-processed reduced graphene oxide-based films as transparent conductive electrodes (TCEs) in organic photovoltaic (OPV) cells. Reduced graphene oxide (rGO) can be effectively utilized as the TCE in flexible OPVs, where the brittle and expensive ITO is incompatible. The prospects and future research trends in graphene-based TCEs are also discussed.



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Laser generated nanoparticles based photovoltaics

C. Petridis[a, b], K. Savva[c, d], E. Kymakis[b, e], E. Stratakis[c]

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http://dx.doi.org/10.1016/j.jcis.2016.09.065

Abstract

The exploitation of nanoparticles (NP), synthesized via laser ablation in liquids, in photovoltaic devices is reviewed. In particular, the impact of NPs' incorporation into various building blocks within the solar cell architecture on the photovoltaic performance and stability is presented and analysed for the current state of the art photovoltaic technologies.

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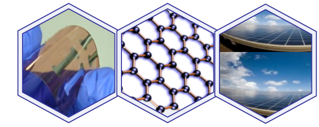

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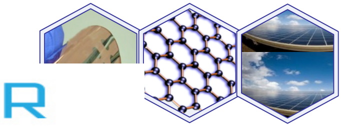

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