Center for Wearable Sensors

UC San Diego
Jacobs School of Engineering

Additive Printing of Flexible Electronics for Sensing

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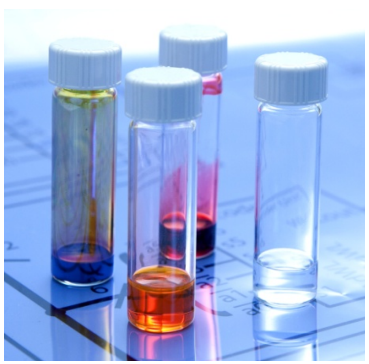
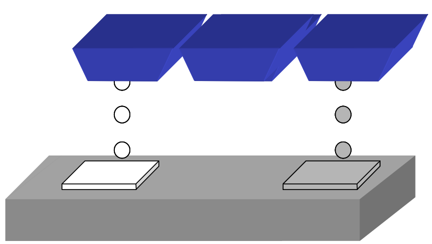
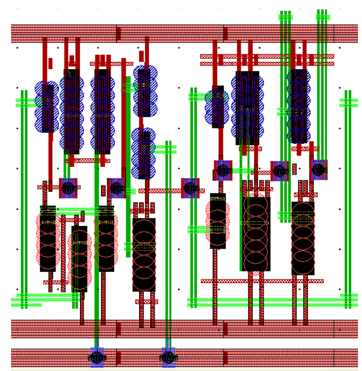
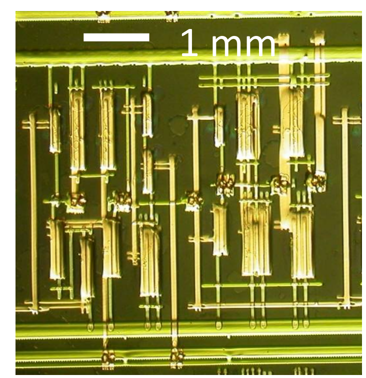
Electrical and Computer Engineering Dept., UC San Diego

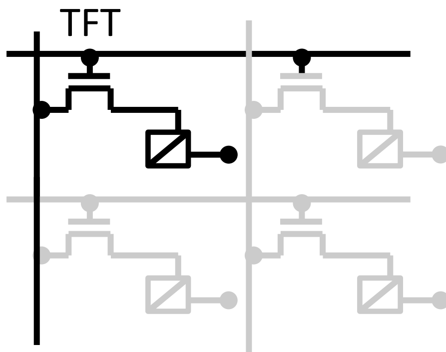


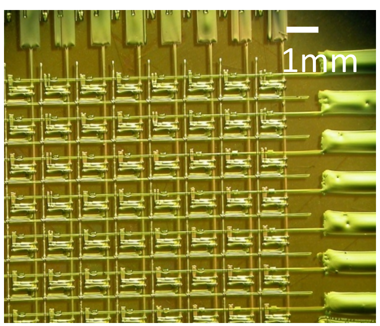




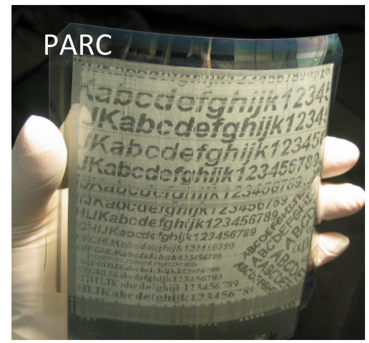


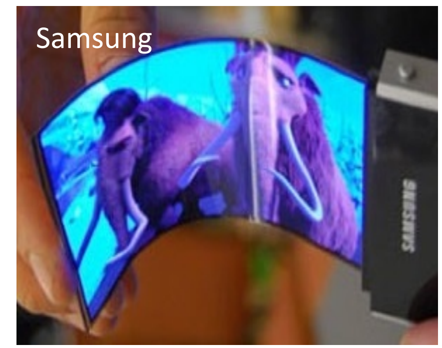
Large-area active matrix array for flexible electronics





Output- display

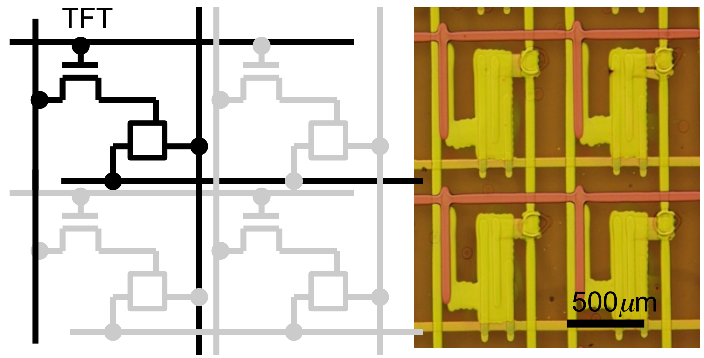




Input -imager, pressure sensors

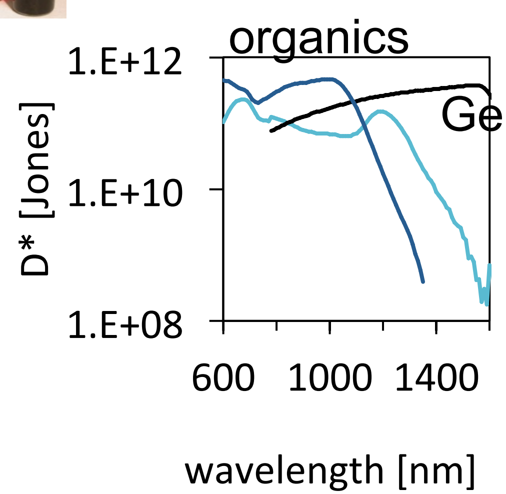
Appl. Phys. Lett. **92** (2008) 213303
Adv. Mater. **21** *(*2009) 1855

Sensing elements for matrix arrays



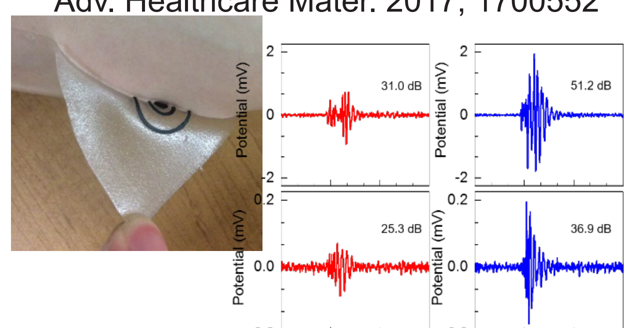
Input devices

infrared photosensor



Bioimpedance electrodes

Adv. Healthcare Mater. 2017, 1700552

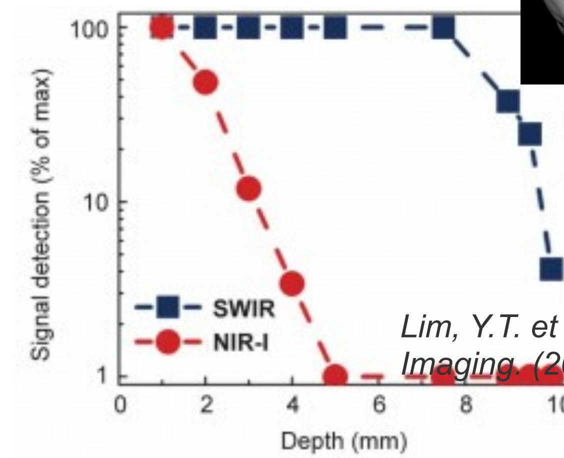


Glove pressure sensor

Imaging beyond visible wavelength

Argos Inc

Figure 5 Test at indoor of visibility in artificial fog

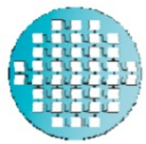
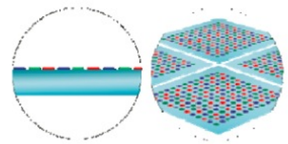
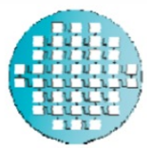
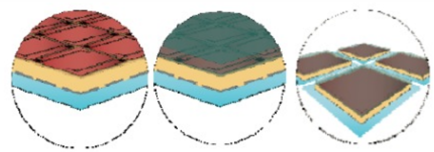
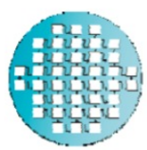
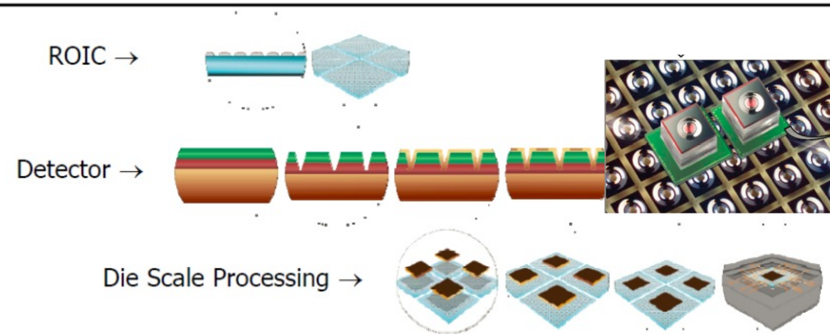


Lim, Y.T. et al. Mol Imaging. (2003) 2, 50.

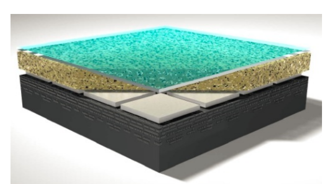
SWIR wavelength can image through fog
-not interfere with human vision
-deeper penetration depth into tissues

Different from thermal imaging, higher resolution for biometrics

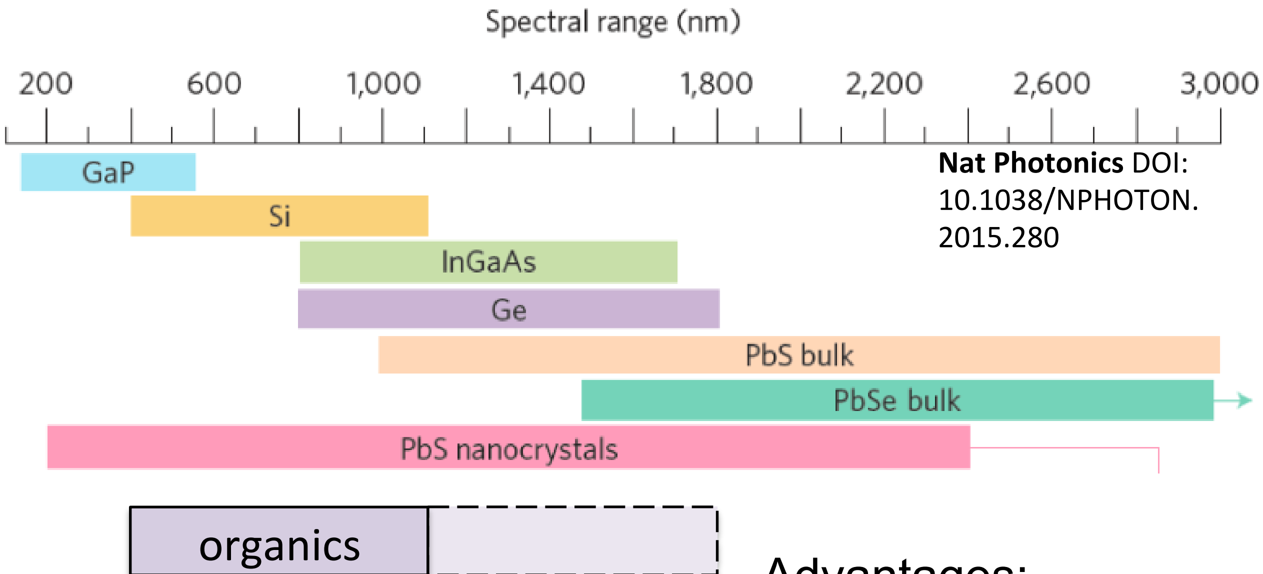
Complex processing of IR photosensor

Technology	Process	Standard Foundry Processes	Non-Standard Processes	Cost
Visible CMOS	Wafer Scale			**$15**
LWIR Micro-bolometer	Wafer Scale			**$1,000**
SWIR IRFPA 1-3um	Die Scale			**> $25,000**
MWIR IRFPA 3-5um	Die Scale			**> $40,000**

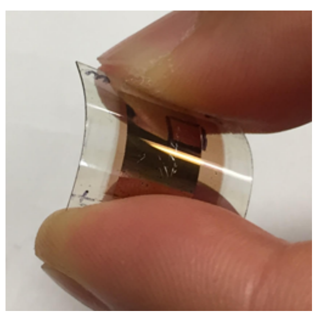
Source: Dr. J. Lewis, DARPA



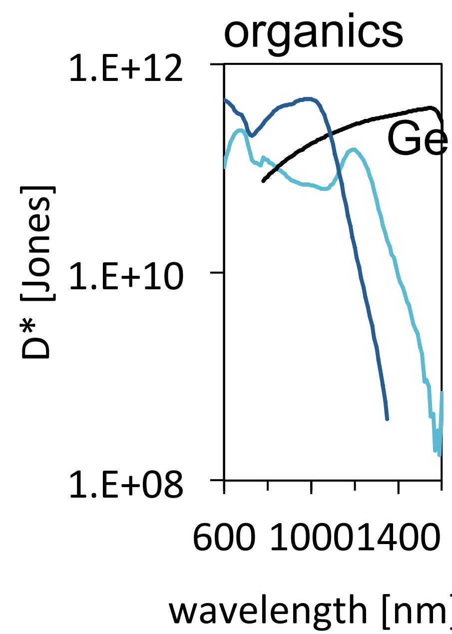
Materials for SWIR spectral region



Nat Photonics DOI: 10.1038/NPHOTON. 2015.280

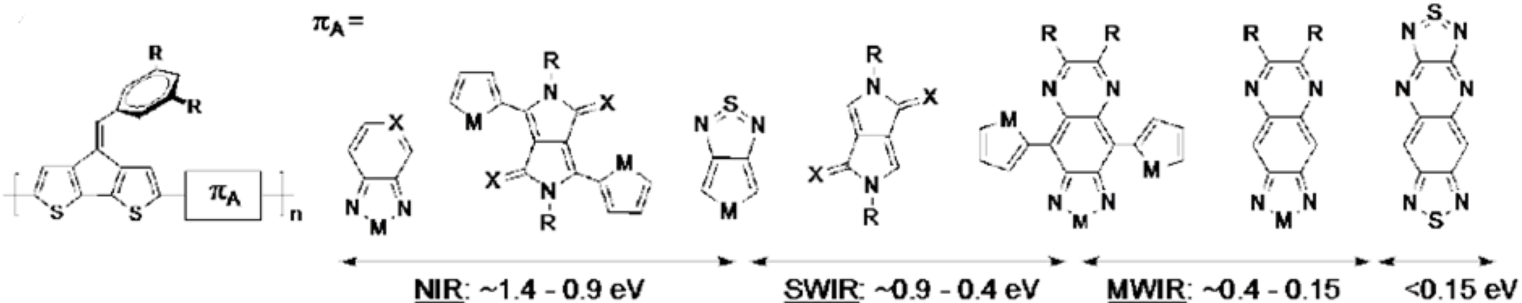


Advantages: scalable, flexible



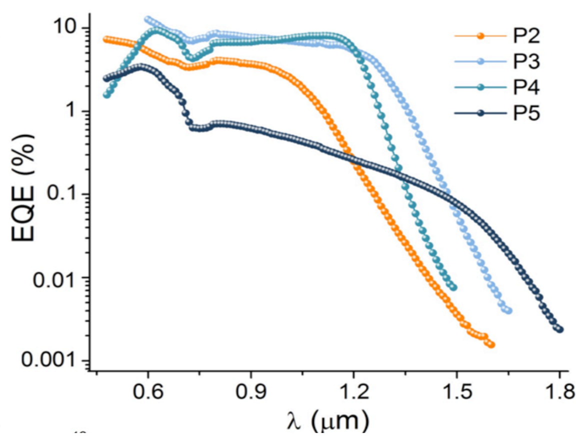
Collaborator: Jason Azoulay, U Southern Mississippi

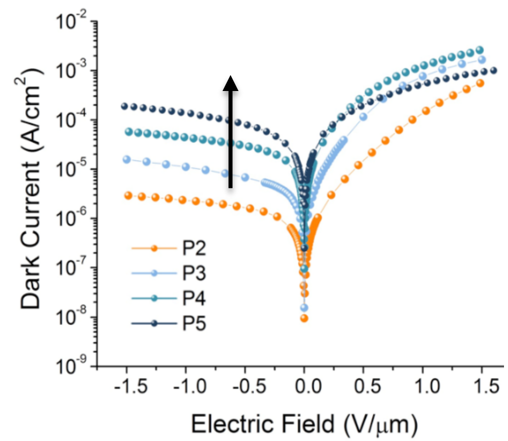
Challenges for SWIR polymer photodiodes



- Signal: low EQE in photodiodes using bulk heterojunction (BHJ):
  - visible: EQE >80%
  - SWIR: EQE <10%

- Noise: increasing dark current with narrowing bandgap





Polymer Chemistry, (2017) **8,** 2922.

ACS Appl. Mater. Interfaces 2017, 9, 1654–1660

Photocurrent generation in BHJ

$$EQE = \eta_{absorb} \cdot \eta_{dissociate} \cdot \eta_{collect}$$

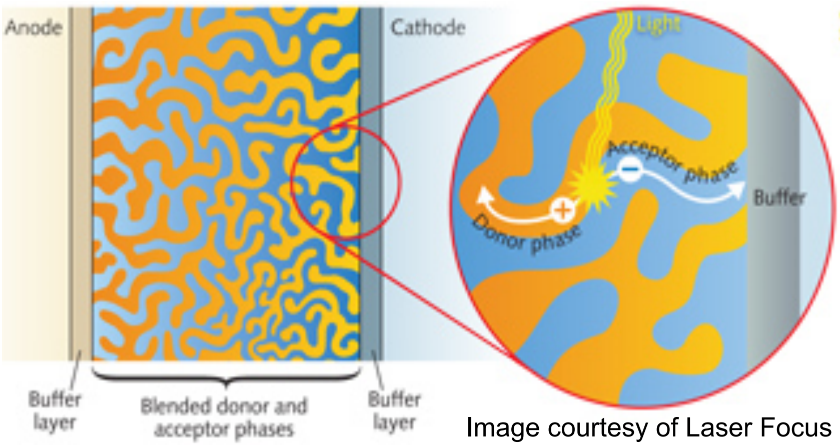


Image courtesy of Laser Focus World

Limiting factor: dissociation or collection?

$$EQE = \eta_{absorb} \cdot \eta_{dissociate} \cdot \eta_{collect}$$

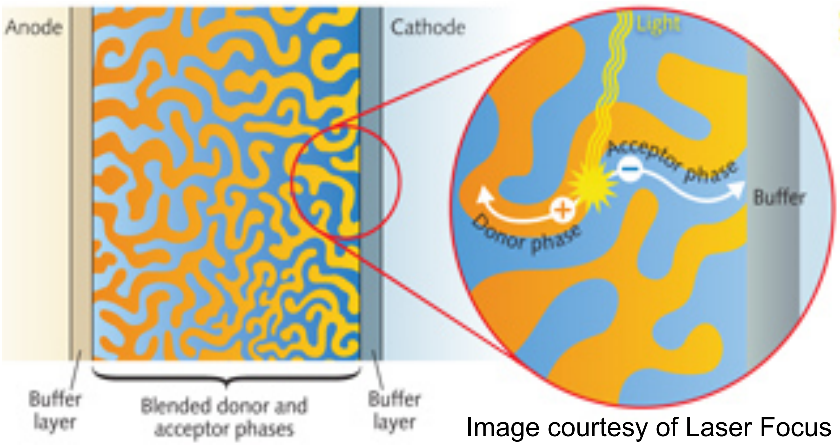
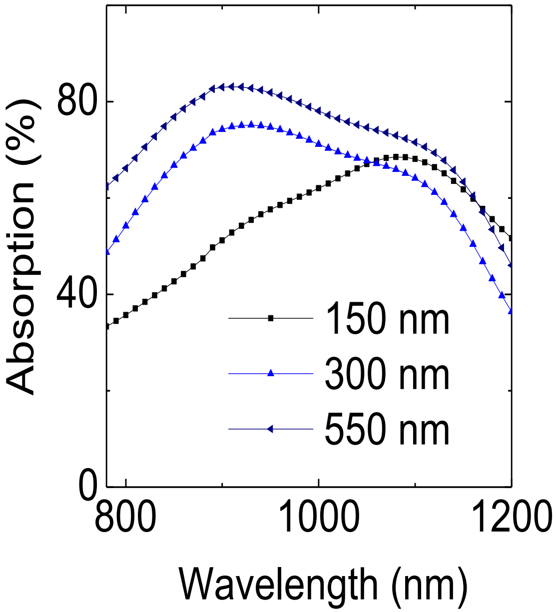
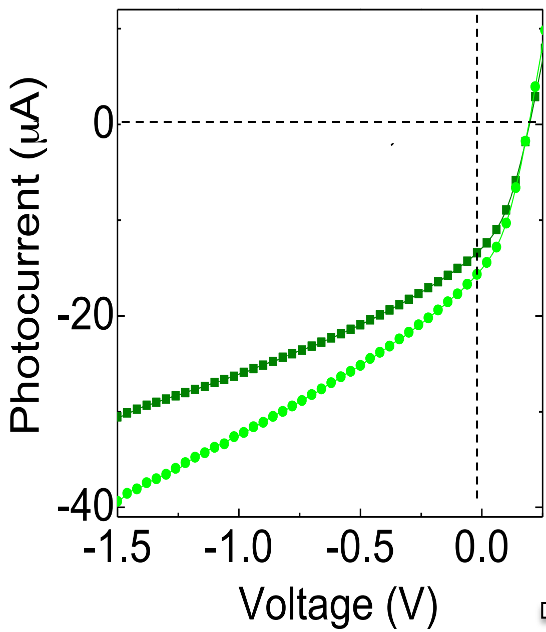


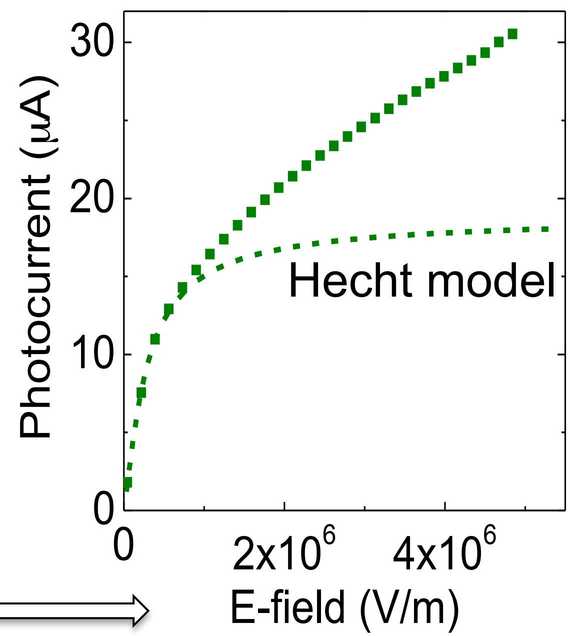
Image courtesy of Laser Focus World



Absorption up to 80% -> OK
EQE ~10%; then accounted for η_{absorb}, IQE~12-20%
is loss mainly due to inefficient dissociation or collection?

Data do not follow Hecht charge collection model





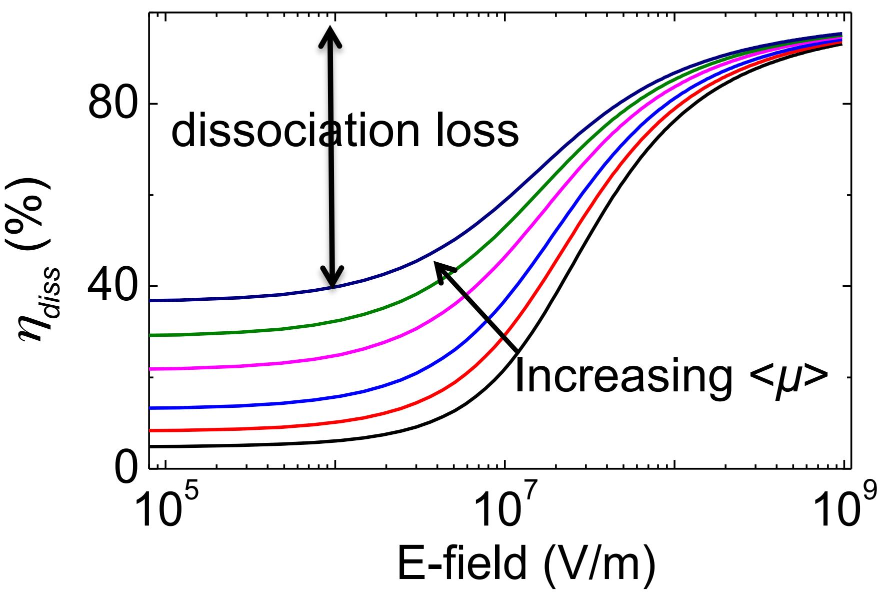
$$J_{ph} = J_{sat} \cdot \frac{2\mu\tau E}{d}\left[1 - \exp\left(-\frac{d}{2\mu\tau E}\right)\right]$$

$$= J_{sat} \cdot \eta_{collect}(E) \cdot \eta_{dissociate}$$

Hecht model assumed this term to be 1

Need to account for $\eta_{dissociate}(E)$ with electric-field dependence

Braun model of exciton dissociation under E-field



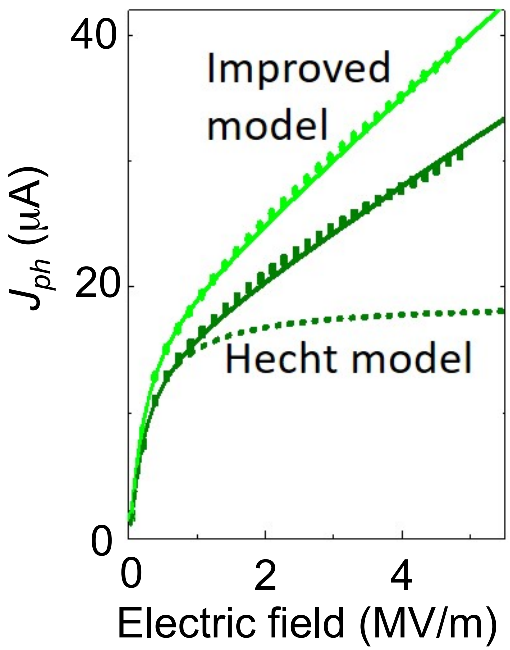
$$k_D(E) = \frac{3e\langle\mu\rangle}{4\pi\varepsilon_0\varepsilon_r a^3}\exp(-\frac{E_B}{kT})[1 + b + \frac{b^2}{3} + \frac{b^3}{18} + \ldots]$$

$$\eta_{diss} = k_D/(k_D + k_F)$$

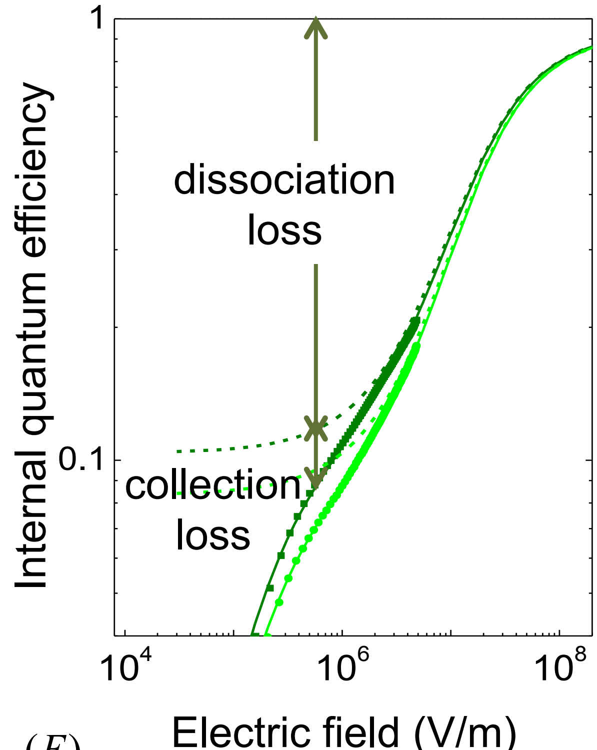
k_D is dissociation rate dependent on E-field
k_F is recombination constant

- Higher dissociation efficiency with larger E-field, towards 100% if field is sufficiently high
- We typically operate around low field of 10^6 to 10^7 V/m; efficiency only between 20-40%

Identify main limiting factor: inefficient dissociation



$$IQE = J_{ph} / J_{sat} = \eta_{collect}(E) \cdot \eta_{dissociate}(E)$$



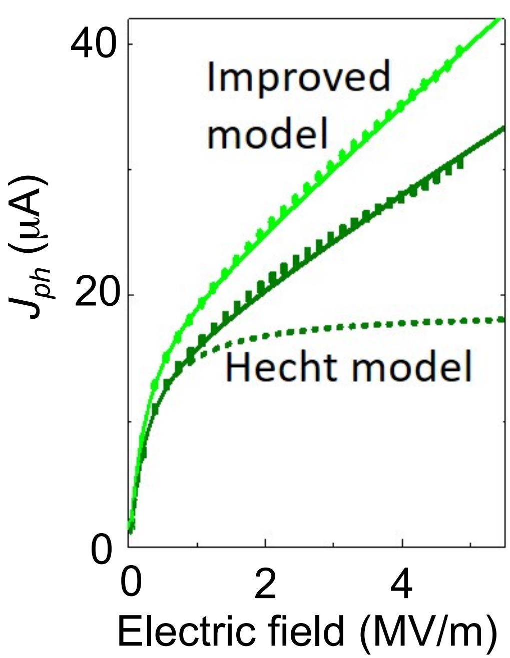
$$EQE = \eta_{absorb} \cdot \eta_{dissociate} \cdot \eta_{collect}$$

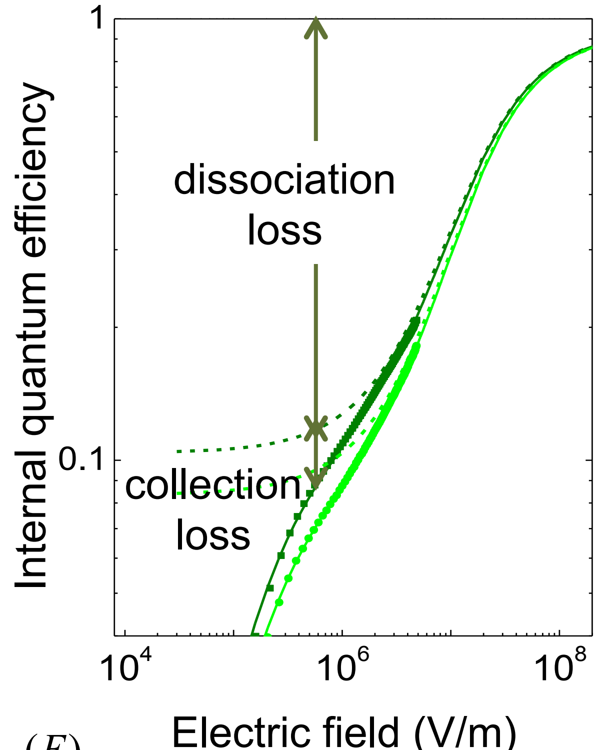
At built-in field ~ 10^6 V/m

EQE (%)	8.0
η_{absorb} (%)	80±5
$\eta_{dissociate}$ (%)	12
$\eta_{collect}$ (%)	75

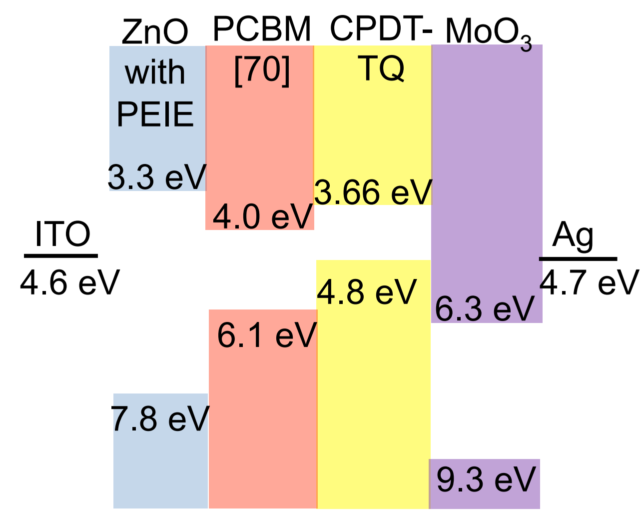
this analysis points out that the most important thing to improve is dissociation.

Identify main limiting factor: inefficient dissociation





$$EQE = \eta_{absorb} \cdot \eta_{dissociate} \cdot \eta_{collect}$$



$$IQE = J_{ph} / J_{sat} = \eta_{collect}(E) \cdot \eta_{dissociate}(E)$$

this analysis points out that the most important thing to improve is dissociation.

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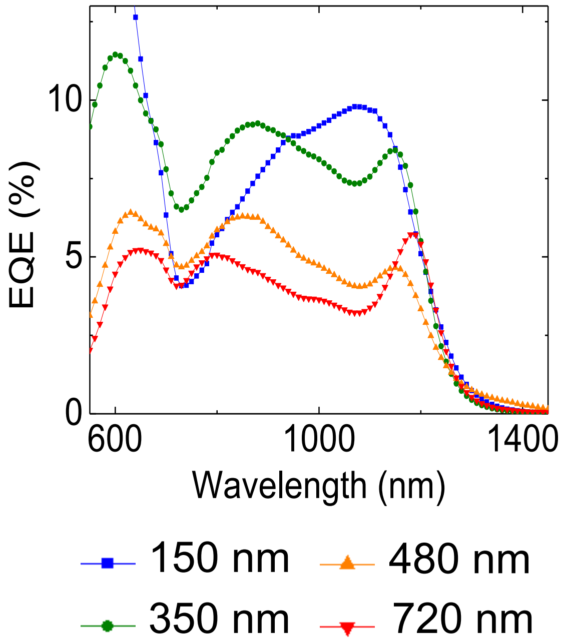
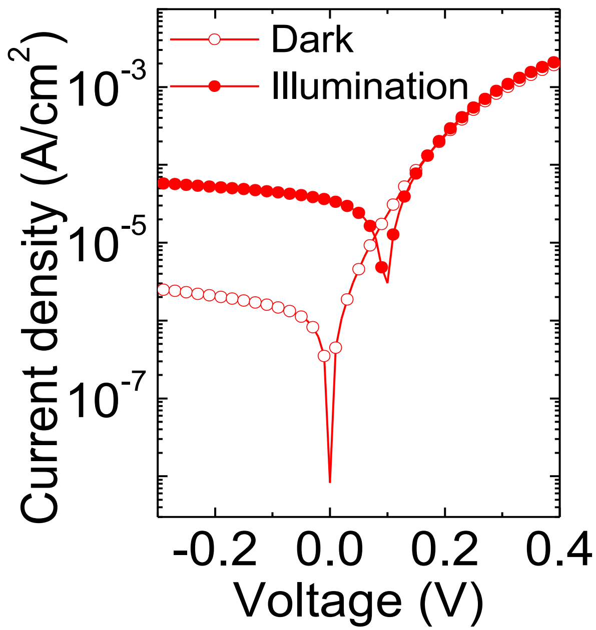
Detectivity is a signal-to-noise ratio

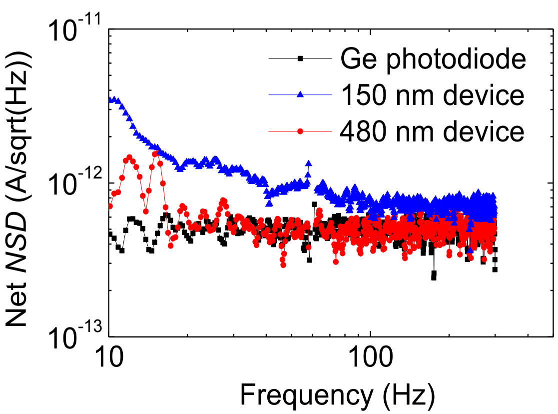
EQE is the signal efficiency



Need to account for noise in detectivity:

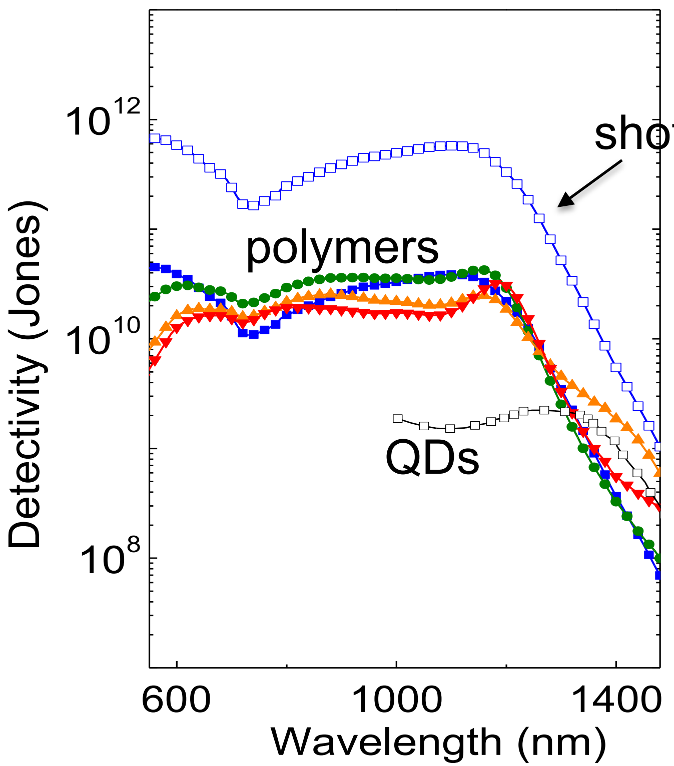


$$D^* = \frac{EQE}{i_{noise}} \frac{\lambda q \sqrt{Af}}{hc}$$



Accurate detectivity calculation

$$D^* = \frac{EQE}{i_{noise}} \frac{\lambda q \sqrt{Af}}{hc}$$



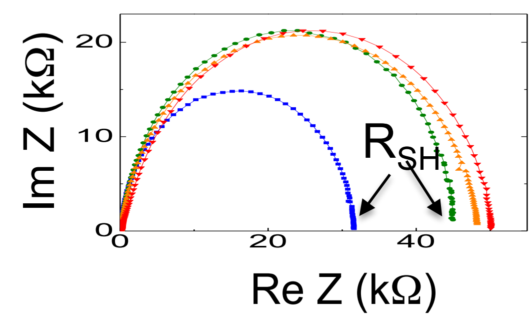
shot noise assumption

$$I_{\text{shot noise}} = \sqrt{2qI_D \Delta f}$$

Measurements that account for thermal noise

$$I_{\text{thermal noise}} = \sqrt{\frac{4kT\Delta f}{R_{SH}}}$$

If shunt resistance is on the order of 1 Mohm, then shot noise ~thermal noise



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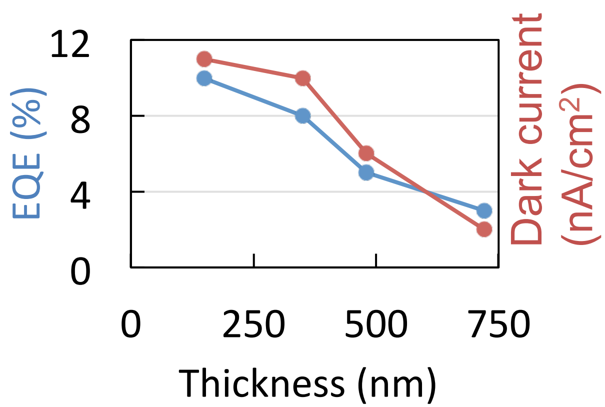
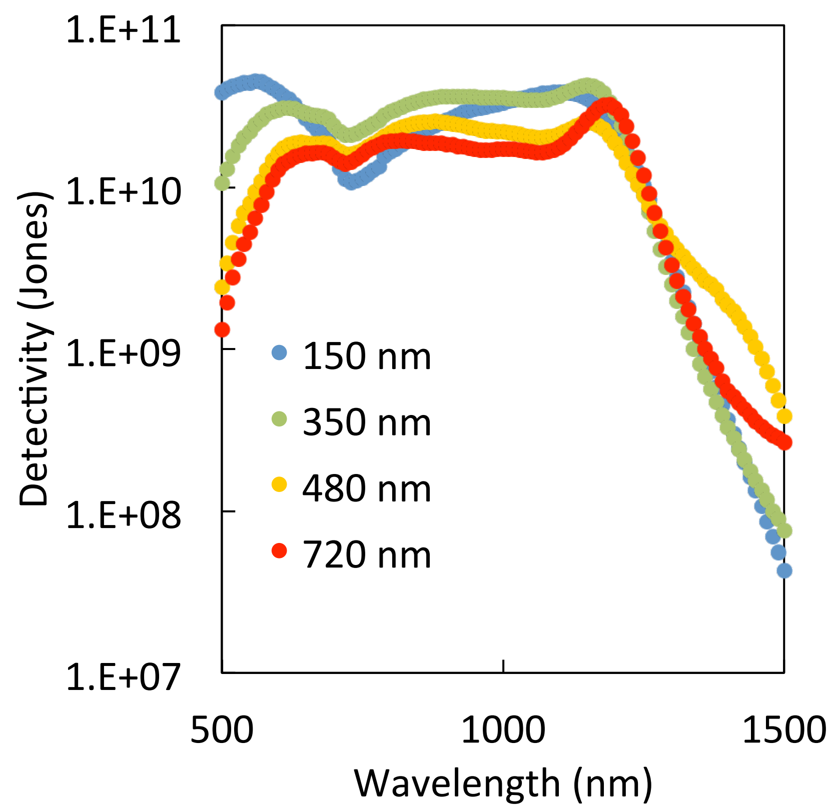
Detectivity upon varying device thickness

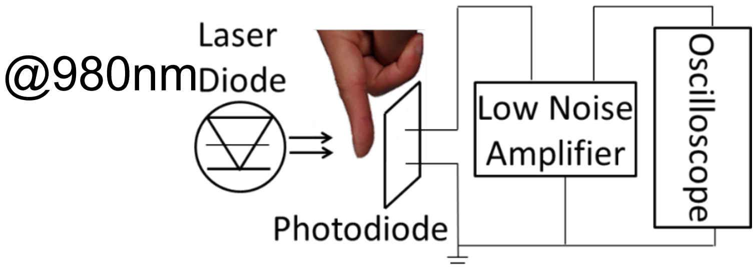
$$D^* = \frac{EQE}{i_{noise}} \frac{\lambda q \sqrt{Af}}{hc}$$

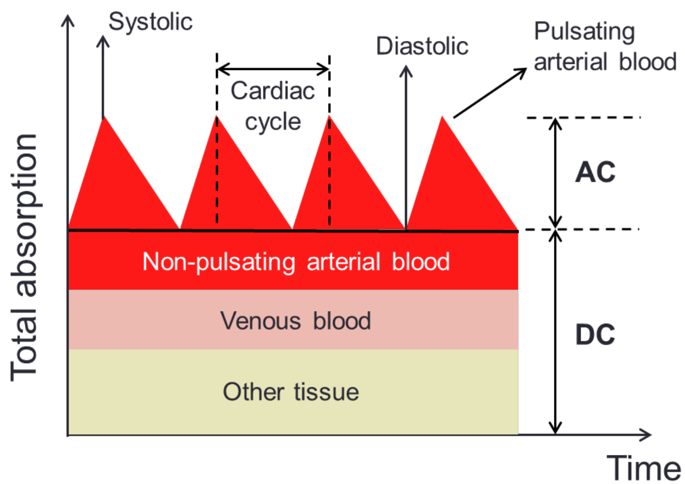




Optimal thickness:
~150 nm (0V, increase optical path)

Example: Measurement of heart pulse rate





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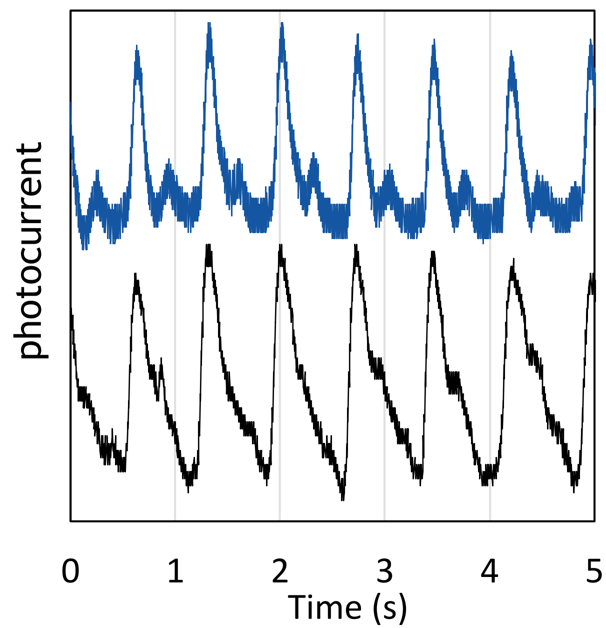
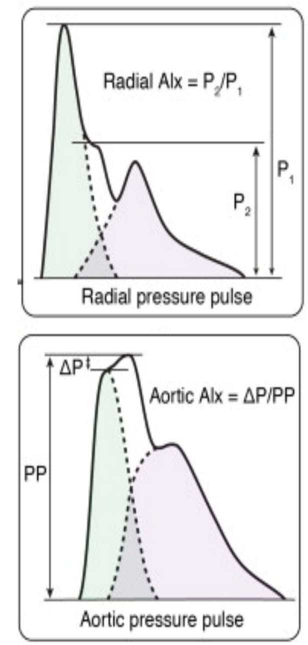
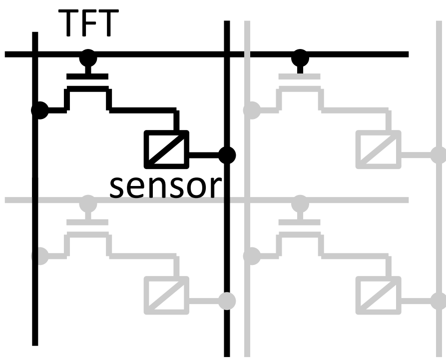


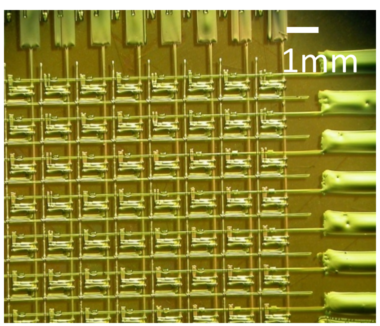


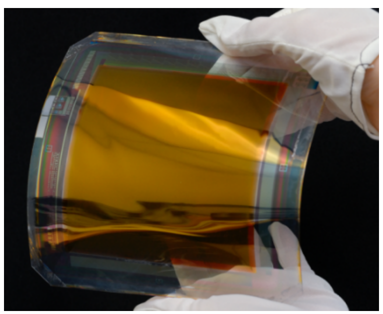
Augmentation index: indicate arterial stiffness, higher index is higher risk

Integration prototype examples

Active-matrix array sensor







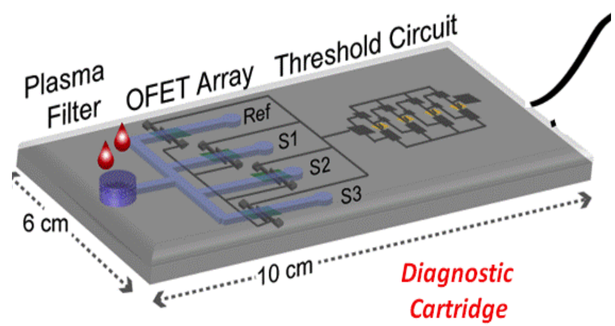
Integrated sensor circuit tags

Sensors

TFT processing circuits

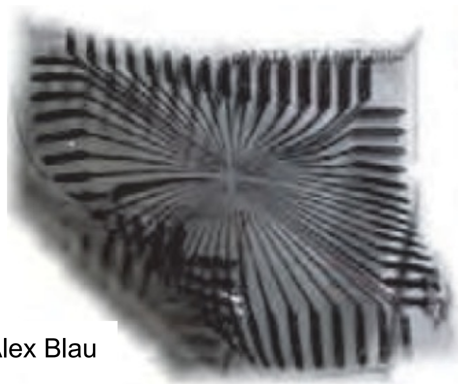
Memory

Display



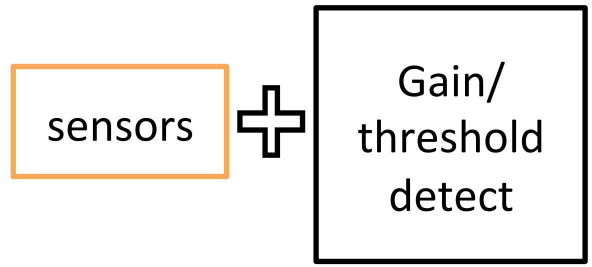
Printed TFTs for local sensor control

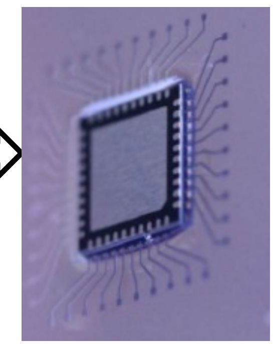
M x N lines, interconnect takes more space than sensors



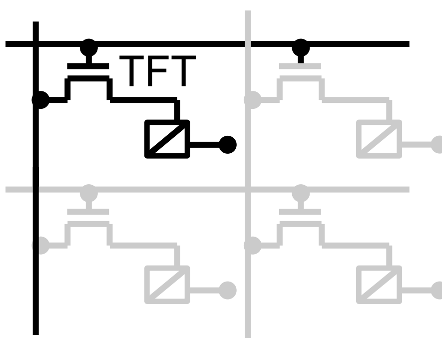
Alex Blau

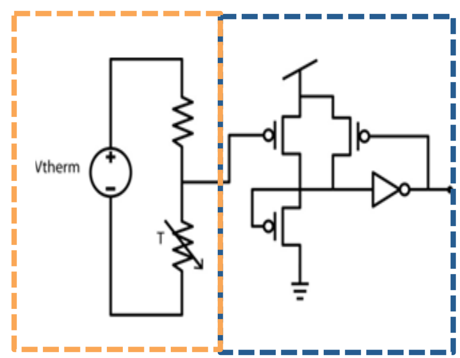
Simple signal conditioning/processing





With TFTs, M + N lines only

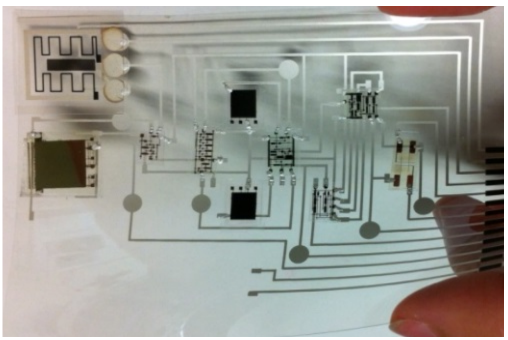




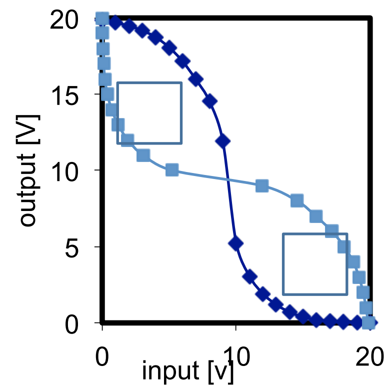
TFT integrated circuits provides signal conditioning before Si chip

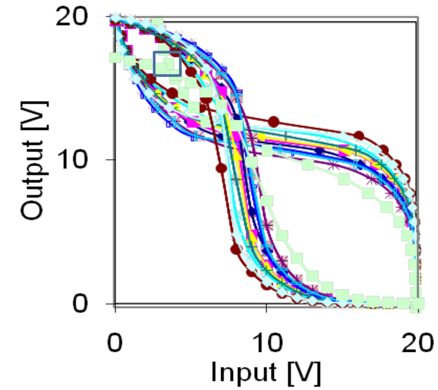
Key challenge for integrated TFT circuits

Challenge for implementation: Designs that tolerate variations in OTFTs



Sci. Rep (2015) 5, 13457.
Proc. IEEE (2015) 103, 607





- Variation leads to circuit error
- Controlling variation is key to practical yield

TFT variation (1s std dev)	Yield
25%	50%
10%	80%
5%	98%

Monte Carlo Simulation for 100 samples
-for a gain + latch circuit with 7 TFTs

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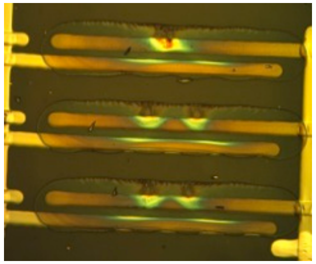
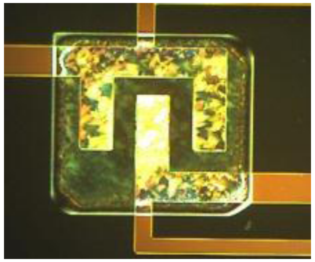
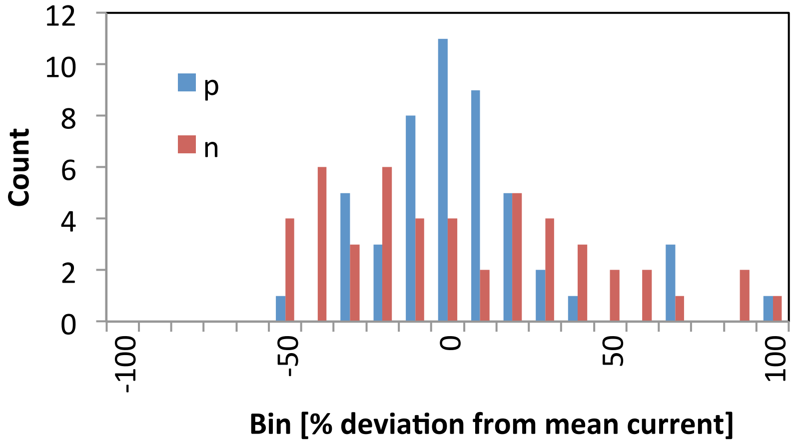
Printed vs photolithographic OTFTs

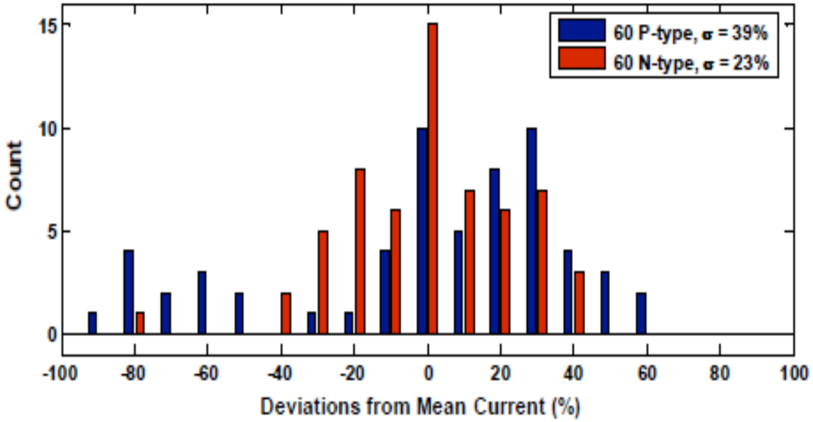


All-printed OTFTs



Photolithographically patterned OTFTs





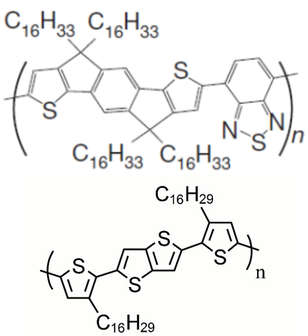
Courtesy of Murmann group at Stanford

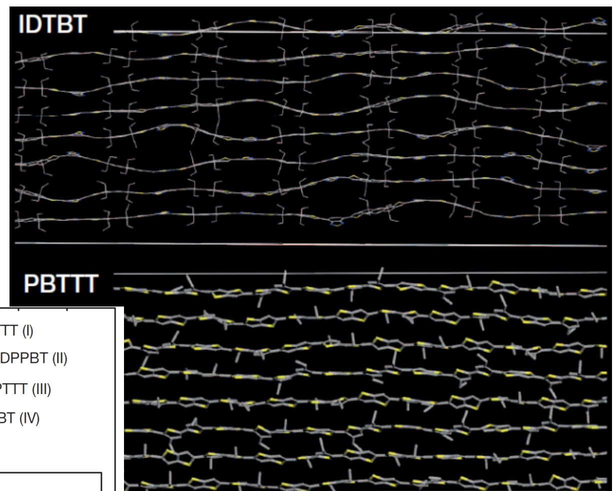
Sci. Rep (2012) 2, 585.

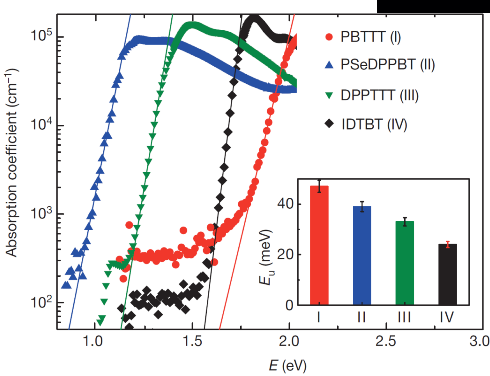
Similar level of variations: main source of variation is semiconductor, less impact from channel W/L

Material structures that reduce disorder

Polymers:
Reduce tail states by rigid backbone that reduces torsion

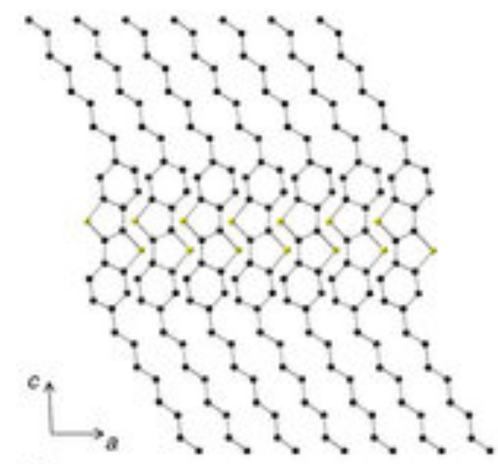


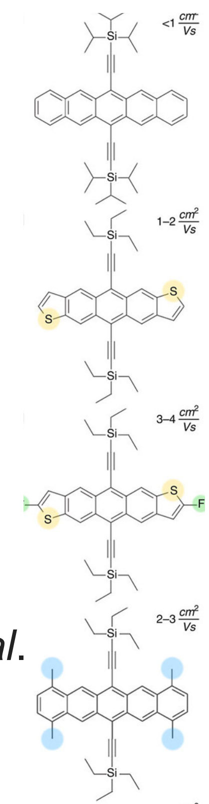




McCoullouch, *et al.*
Nature **515**, (2014) 384–388

Small molecules:
Suppressing thermal disorder by side chain location

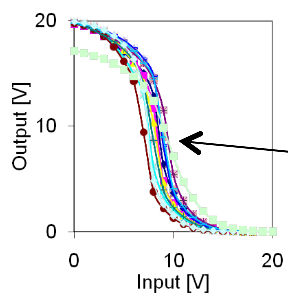




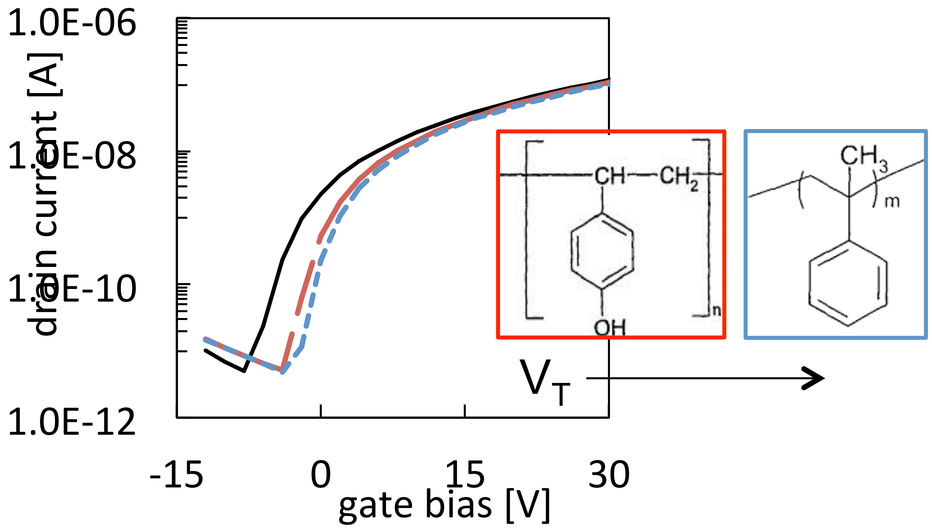
J. Anthony, Sirringhaus, *et al.*
Nature Communications **7**, 10736 (2016)

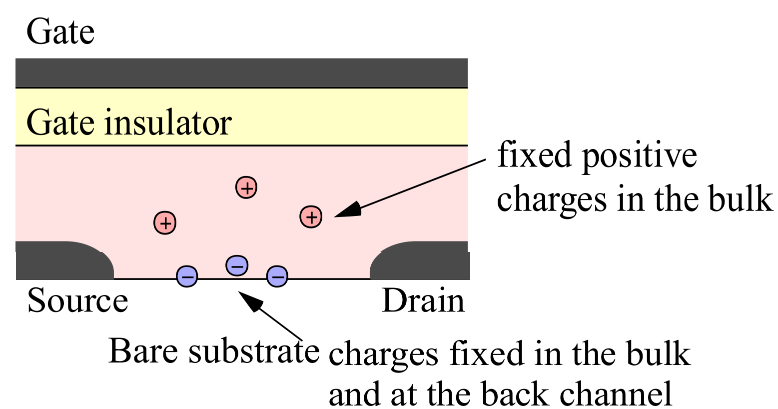
Modify channel surface to adjust V_T

- Important to control threshold voltage V_T
- Back-channel interface affects V_T: electronic dipole, film morphology, etc.



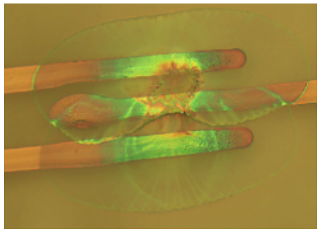
$$V_{inv} = \frac{V_{DD} + V_T^p + V_T^n \sqrt{\beta^n / \beta^p}}{1 + \sqrt{\beta^n / \beta^p}}, \beta = \frac{W}{L}\mu$$

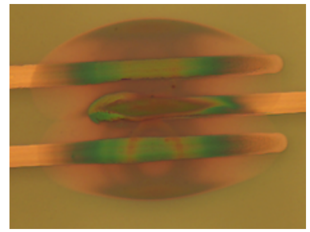


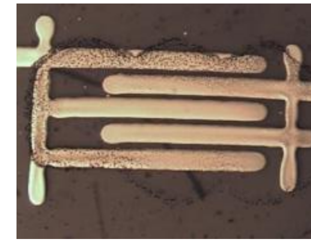


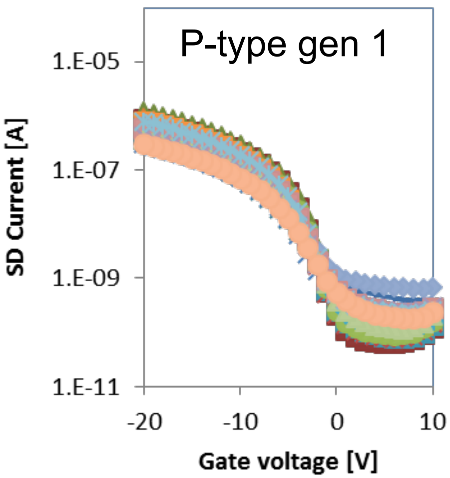
J. Appl. Phys. **113** (2013) 094506

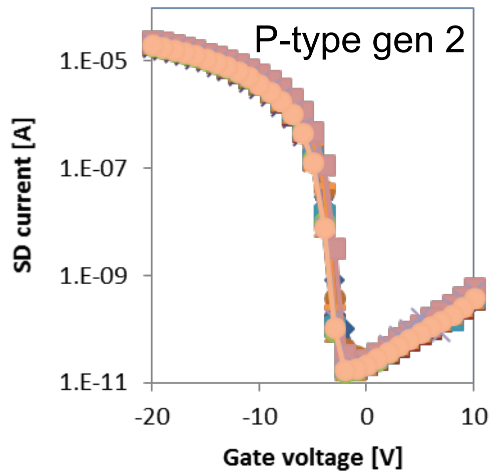
Reduced variations in printed OTFTs

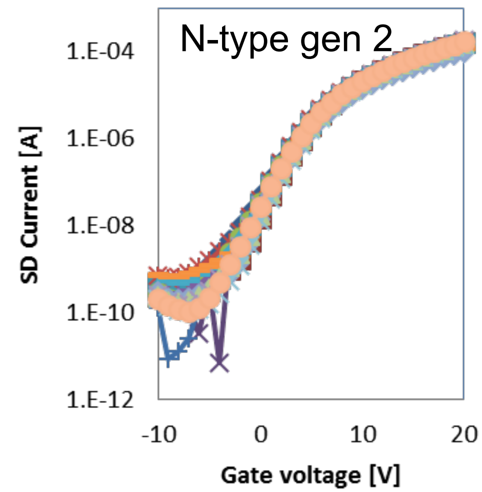












1σ =40%
Mobility =0.12cm^2/Vs

1σ =8-10%
Mobility =0.6cm^2/Vs

1σ =12%
Mobility =0.7cm^2/Vs

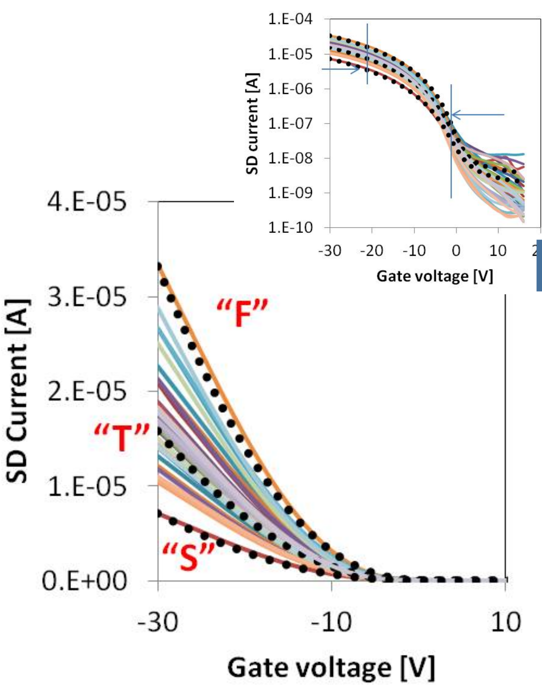
Uniformity can be improved in both polymer and small molecules

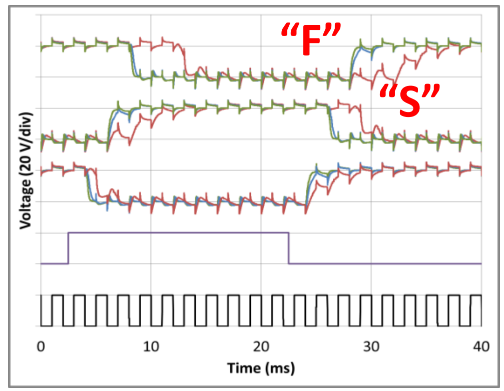
From materials to circuit fabrication

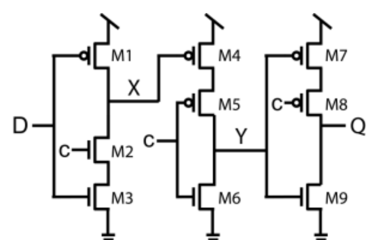
Develop ink & devices
→ Build device models

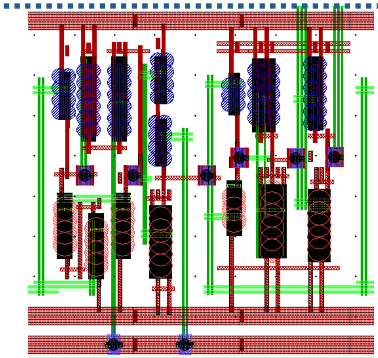
Design, simulate circuits

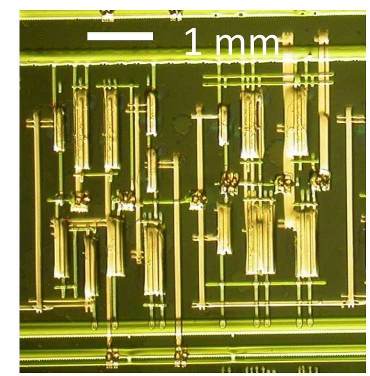
Print and test







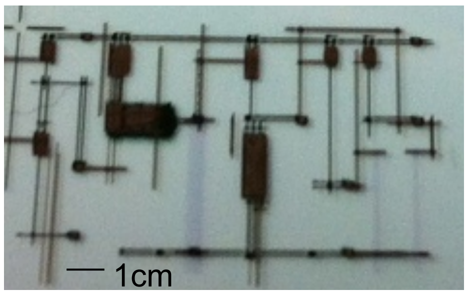


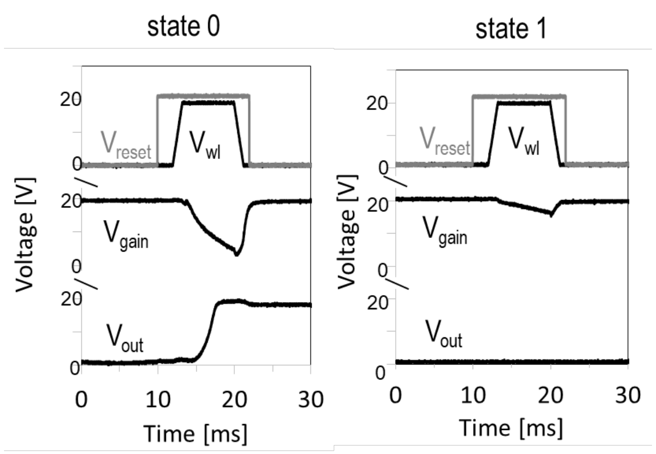


IEEE Elec. Dev. Lett. (2013) 34, 271.

Examples of printed circuits

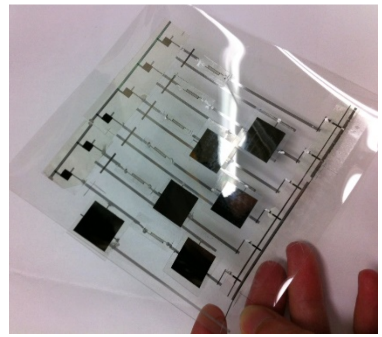
Memory readout

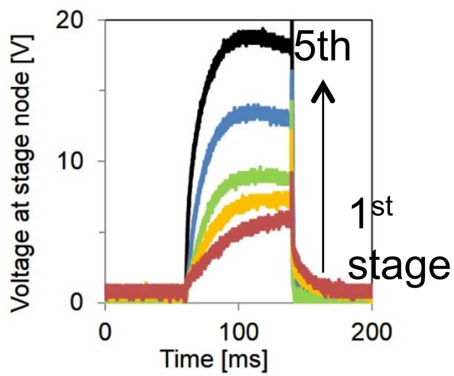




IEEE Transactions on Electron Devices (2017) 64, 1981-1984

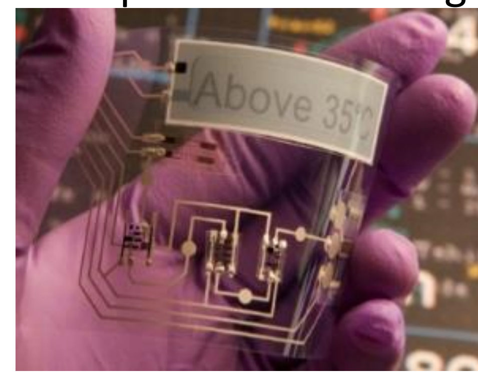
Voltage multiplier

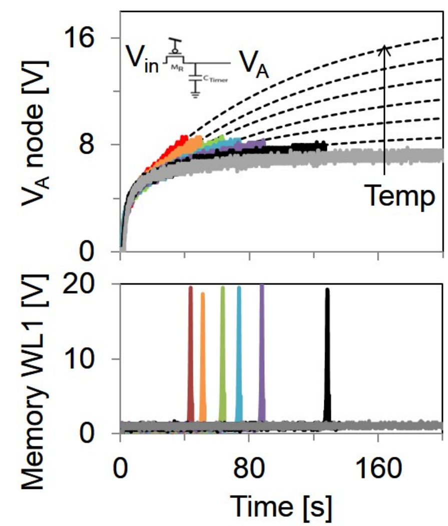




Flexible Printed Electronics (2016) 1, 015002.

Temperature dose tag





Scientific Reports (2015), 5, 13457.

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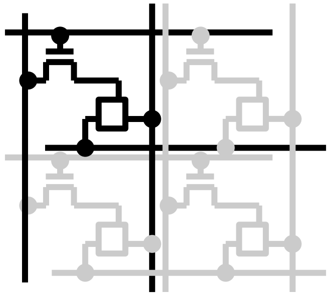
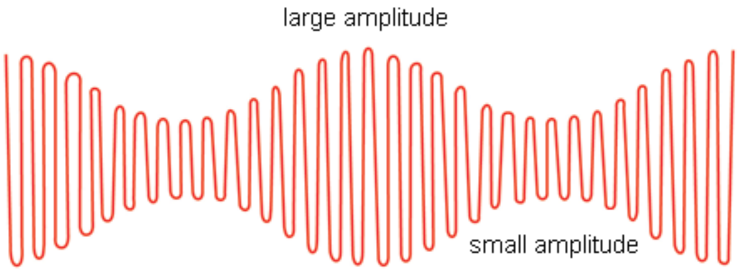
Desirable to digitize signal near sensor

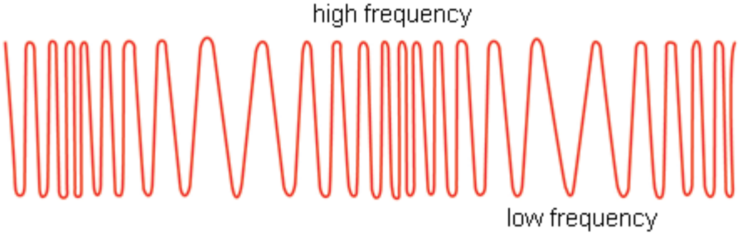
Amplitude signal prone to attenuation error; frequency signal more reliable

Need to add digitizing circuit near sensor



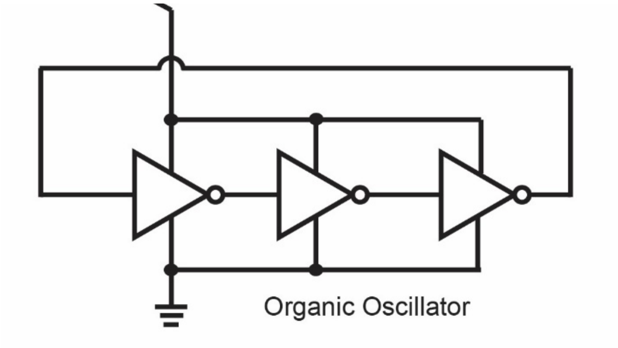


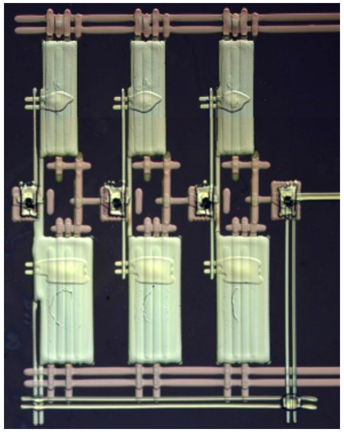
Attenuation affects amplitude measurement

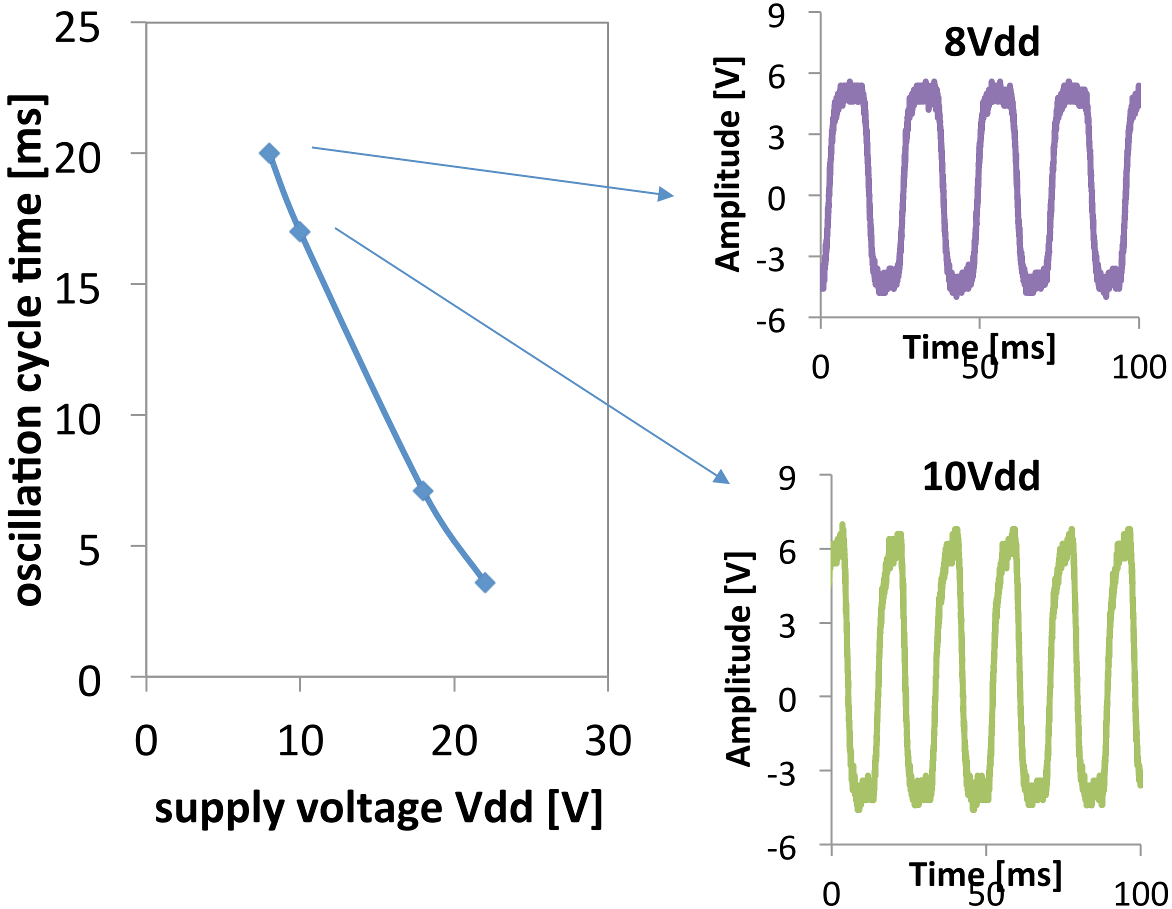


Same freq as before, will get same readout

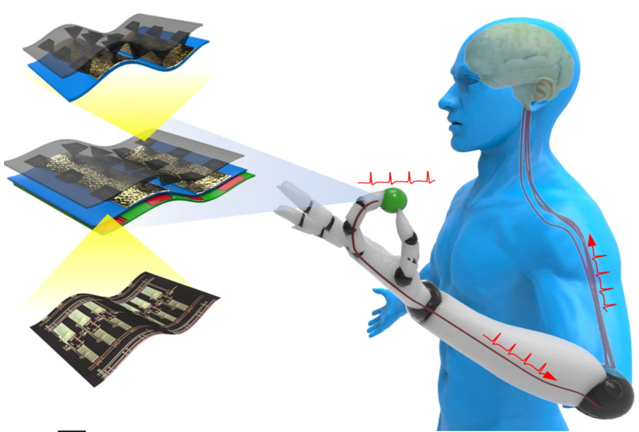
Voltage-controlled oscillator

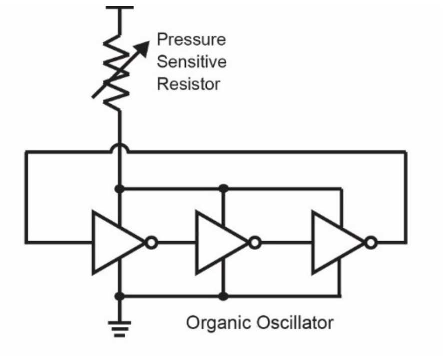


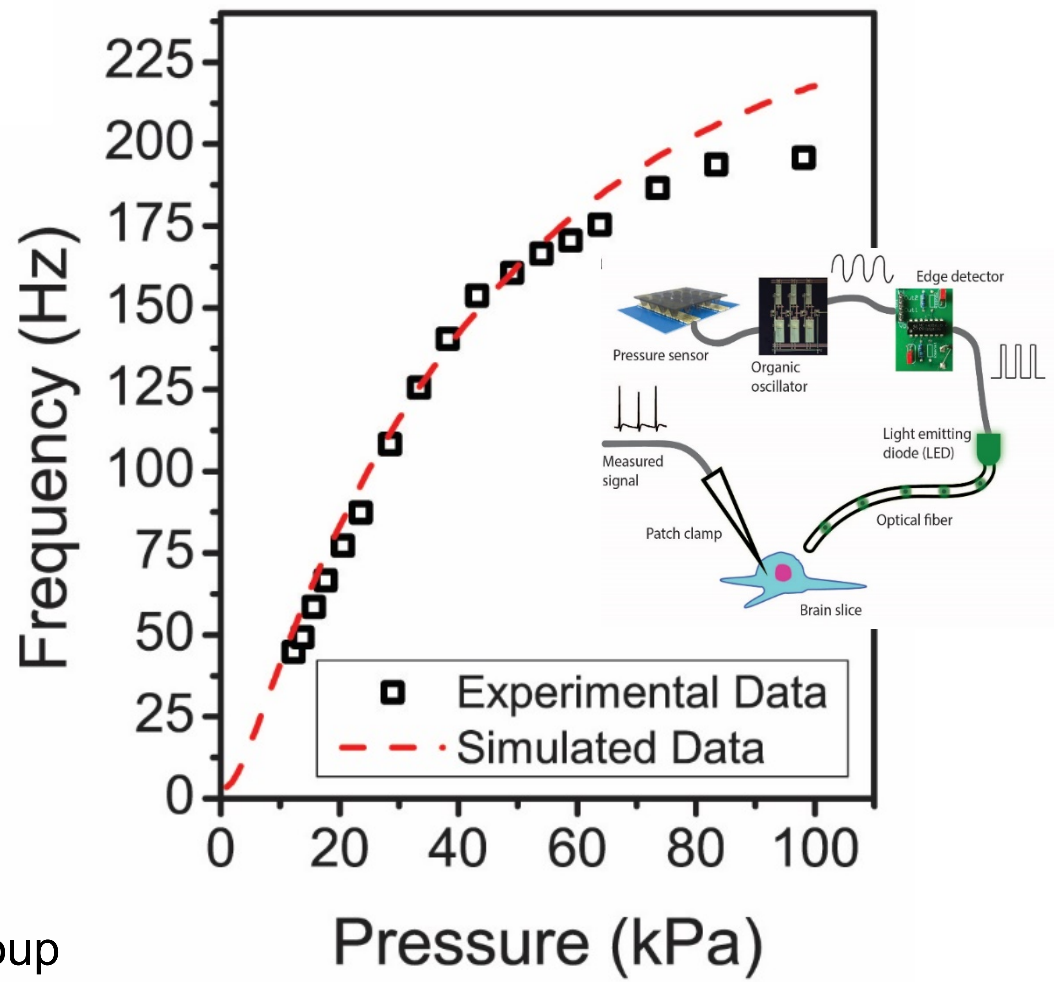




Using printed components to mimic skin mechanoreceptor





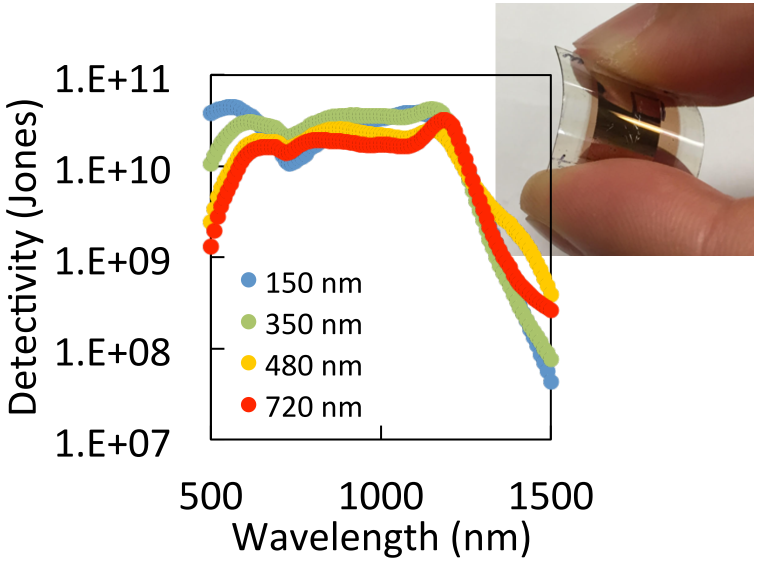


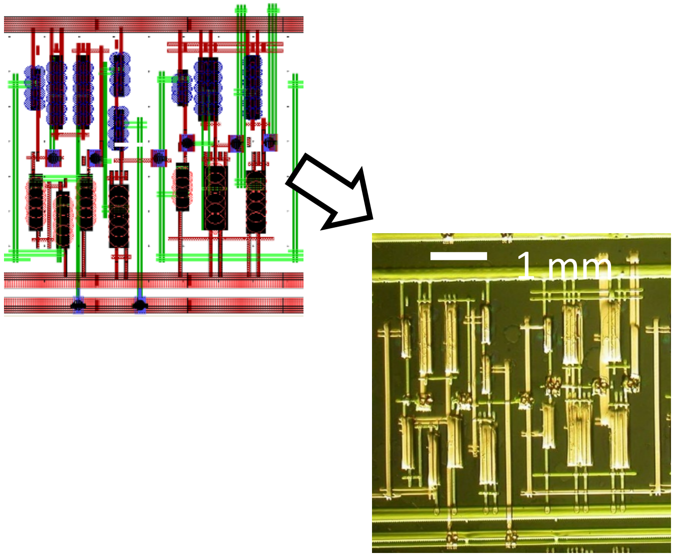
Science (2015) 350, 313.
In collaboration with Zhenan Bao group

Summary

- Develop short-wave infrared sensors for flexible matrix arrays

- Apply additive printing to demonstrate organic TFT circuits
    1. increase tolerance to device variation issues
    2. integrated local digitizing circuits near sensors





Acknowledgment

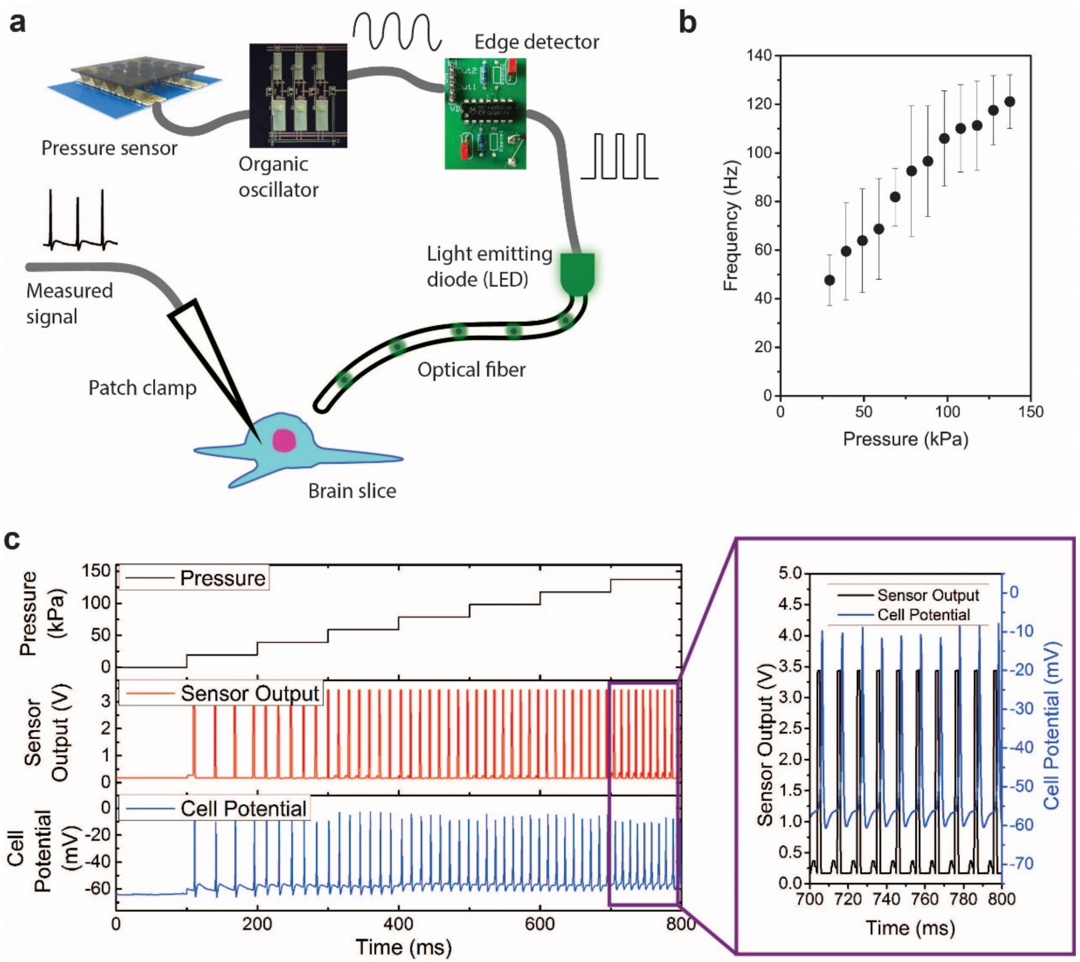
UCSD colleagues:
Leanne Chukoskie
Harinath Garudadri
Andrew Skalsky
Michael Yip

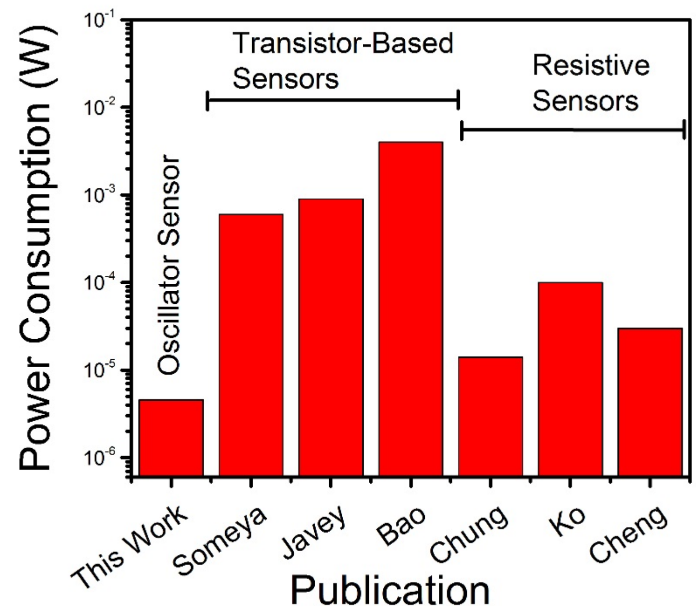
Students:
Fei Deng, Padmaja Jonnalagedda,
Zhenghui Wu, Weichuan Yao,
Hyunwoong Kim, Kaiping Wang,
Udit Parekh, Moran Amit

Collaborators:
PARC colleagues
Jason Azoulay, U Southern Mississippi
Zhenan Bao, Stanford

Funding:

Low-power circuit that stimulates neuron





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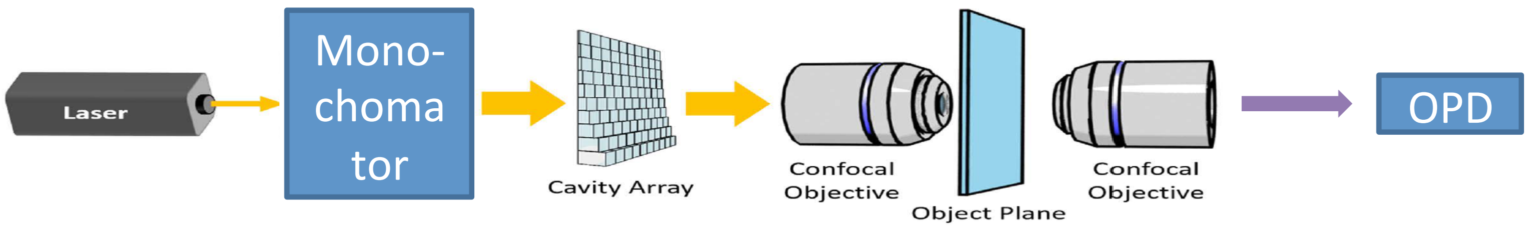
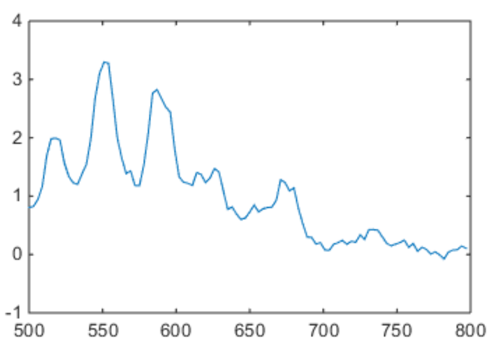
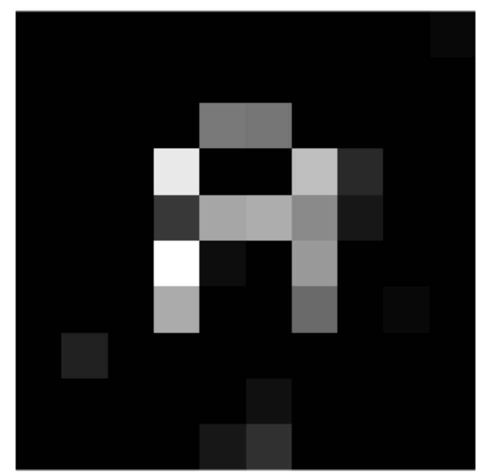
Reconstructive imaging



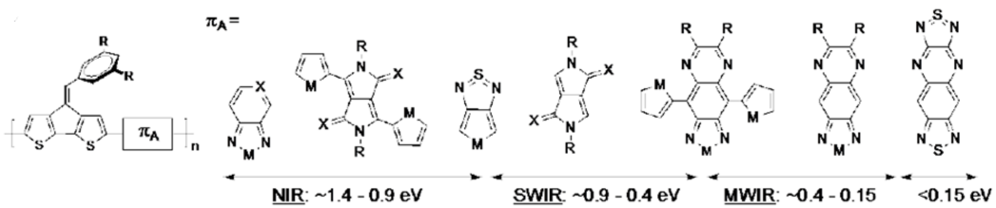
$$y(\lambda)=\int T(x,y,\lambda)\times O(x,y)\,dx\,dy$$

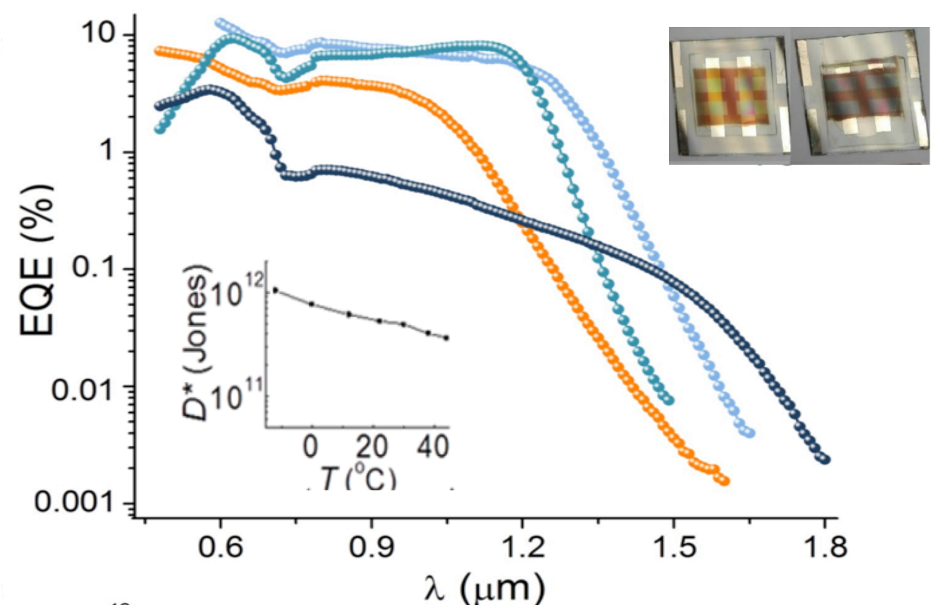
$y(\lambda)$=sensor measurement
$T(x,y,\lambda)$=etalon transmission
$O(x,y)$ = object image



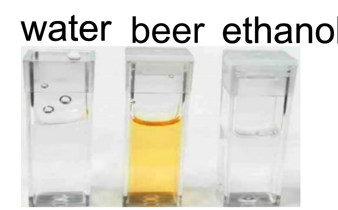


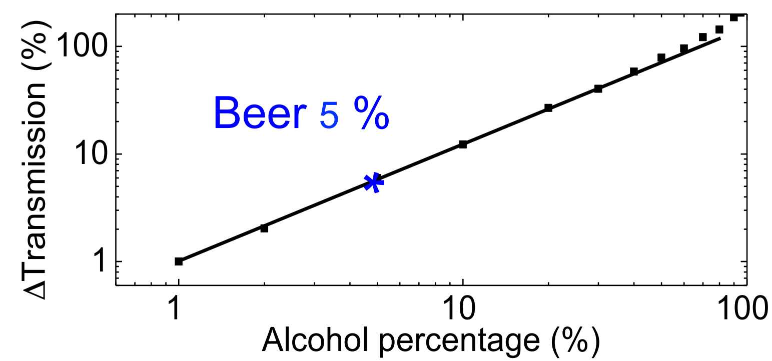
Spectral Range to 1.8 um



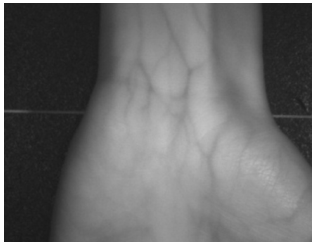


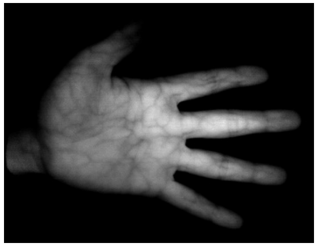
- State-of-the-art spectral range for organics
- Detectivity changes <50% with increasing temperature, great for on-skin wearables





Flexible Organic Shortwave Infrared Camera



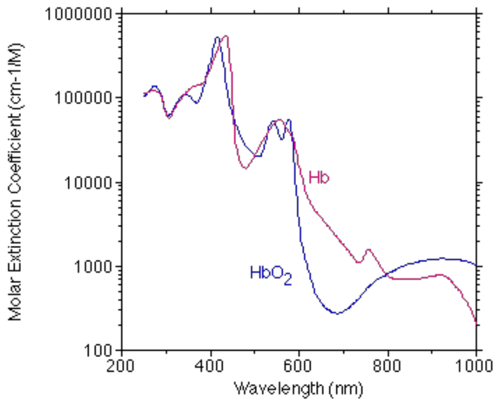


- Biomedical monitoring: identify pulse, vein recognition, blood flow volume monitoring

Requirements:
1. Spectral Range: 1-3 um

2. Flexible thin film sensor that allows skin contact

3. Low cost dynamic imager for 24/7 monitoring



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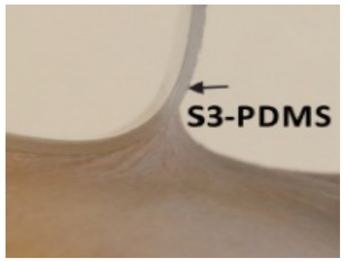
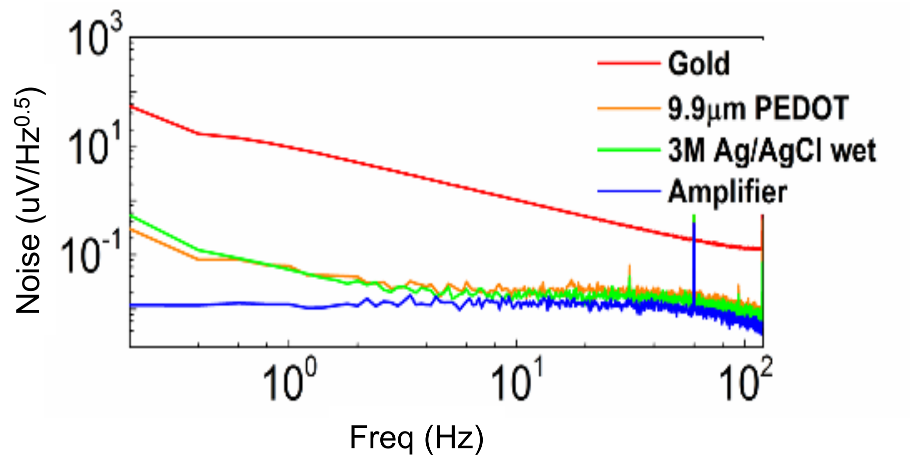
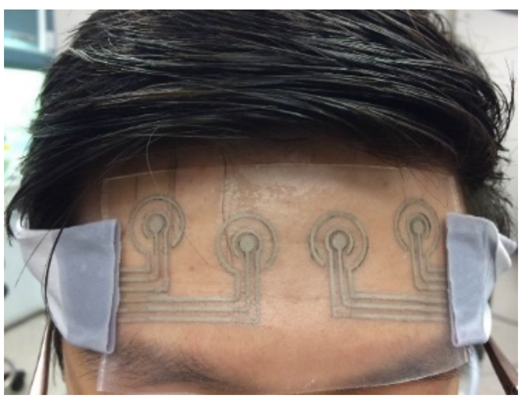
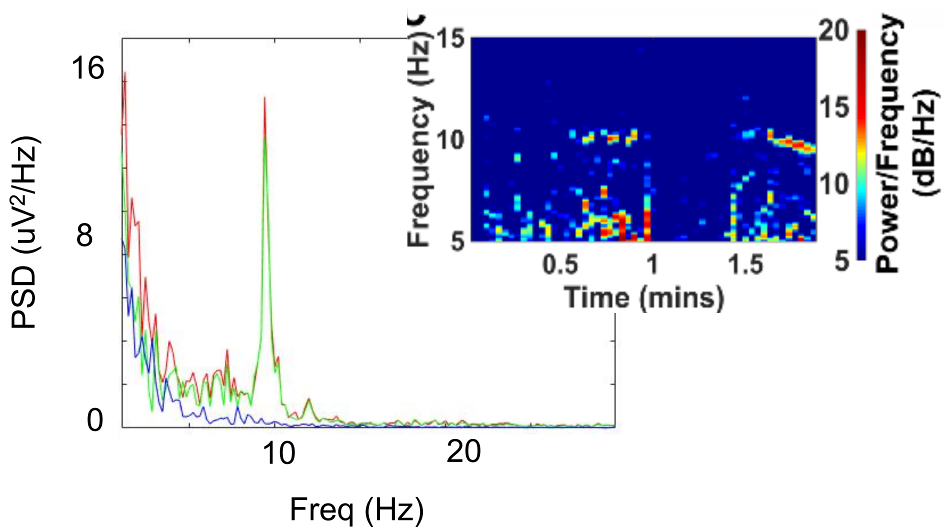
Printed dry electrodes for electrophysiology measurement

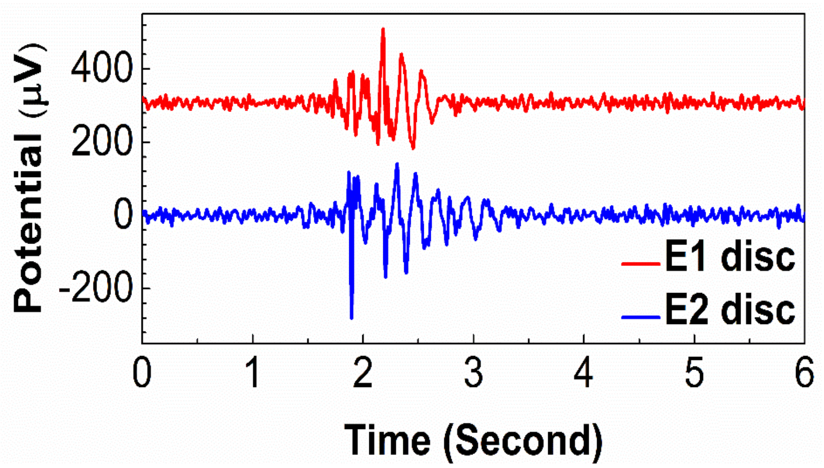


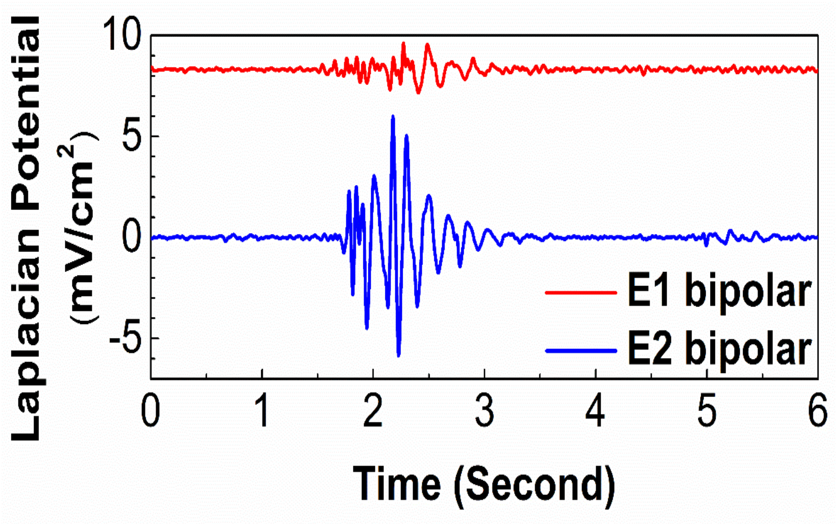






Concentric rings to increase spatial resolution





Adv Healthcare Mater, 2017, in press

Acknowledgment

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Collaborators:
PARC colleagues
Antonio Facchetti, Northwestern
Zhenan Bao, Stanford
Iain McCollouch, Imperial/KAUST

Funding:

NSF

ACS PRF

FlexTech Alliance for Displays & Flexible, Printed Electronics

Need to augment spasticity diagnosis

UI Chicago

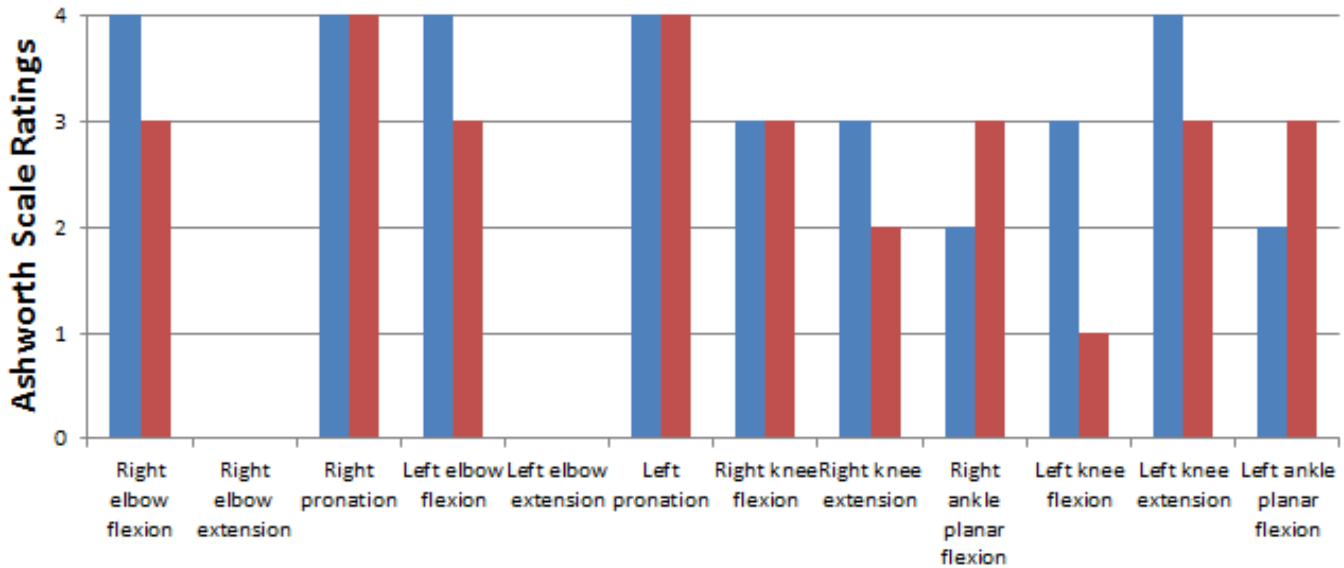
Score	Modified Ashworth Scale (MAS)
0	No increase in muscle tone
1	Slight increase in muscle tone, with a catch and release at the end of the range of motion (ROM)
1+	Slight increase in muscle tone, followed by minimal resistance throughout the remainder of ROM
2	More marked increase in muscle tone through most of the ROM, but affected parts easily moved
3	Considerable increase in muscle tone, passive movement difficult
4	Affected part is rigid in both flexion and extension

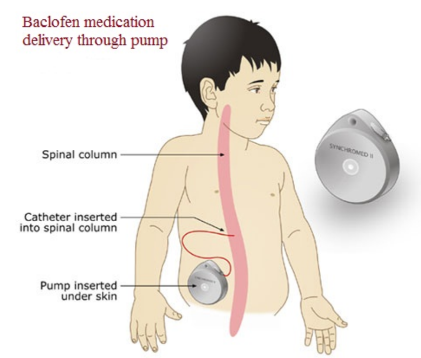
Spatiscity -involuntary activation of muscle, very common in patient with neurological disorders such as stroke, traumatic brain injury, cerebral palsy, etc. affect 764K in US; 17M world wide

Issue with reliability in MAS ratings

- 5 patients and 12 tasks: each doctor gave 60 MAS ratings
- Only 27% of the ratings were the same; poor inter-rating reliability, yet dosage is based on this rating

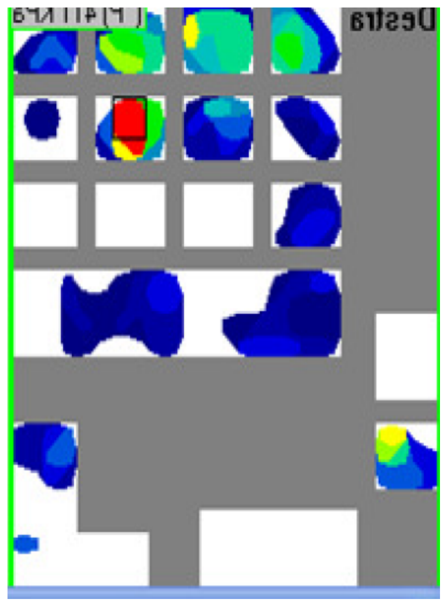
Two doctors' MAS ratings on the same patients

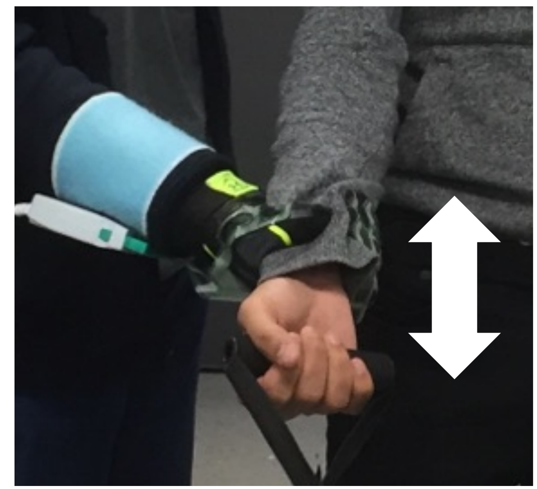


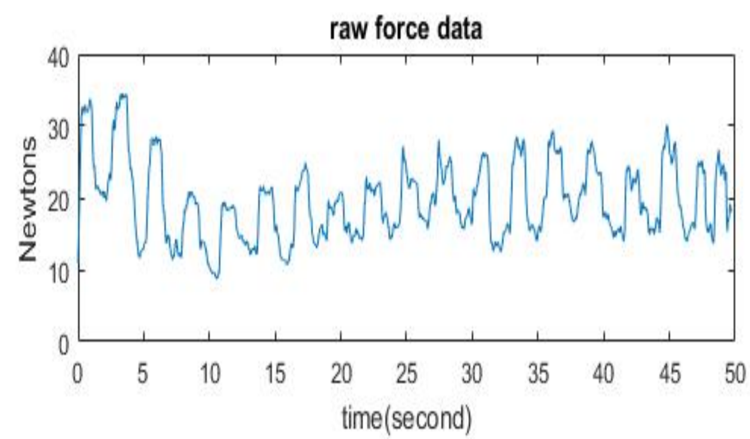


Lead by Dr. Garudadri (Calit2) and Dr. Skalsky at UCSD School of Medicine

Prototype glove to quantify spasticity

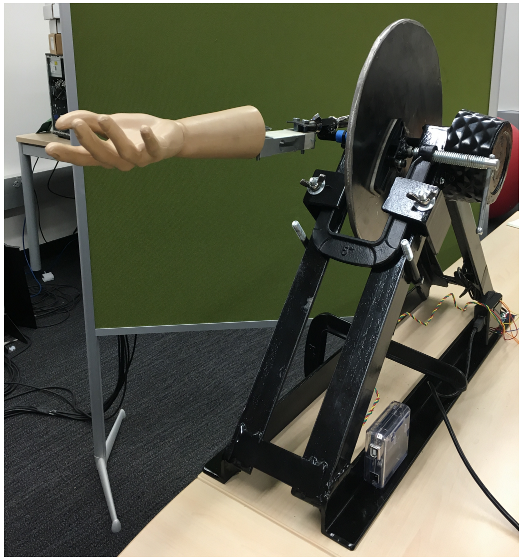


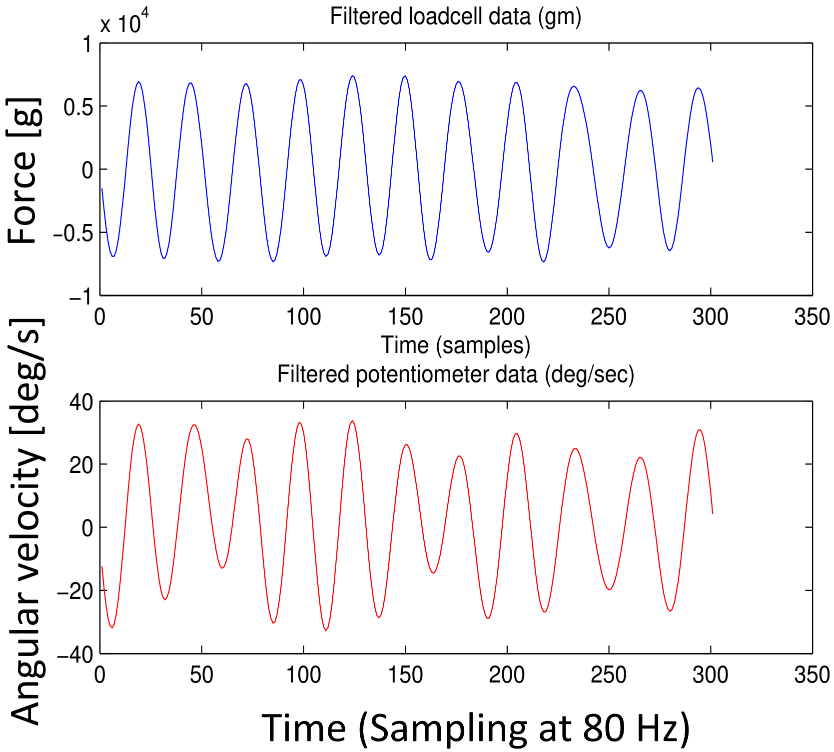




Glove worn by the doctor during assessment:
-measure force (printed pressure sensor by Tekscan) and angular velocity (gyroscope)
-Power to move a limb P=F*v

Mock patient to calibrate sensor glove

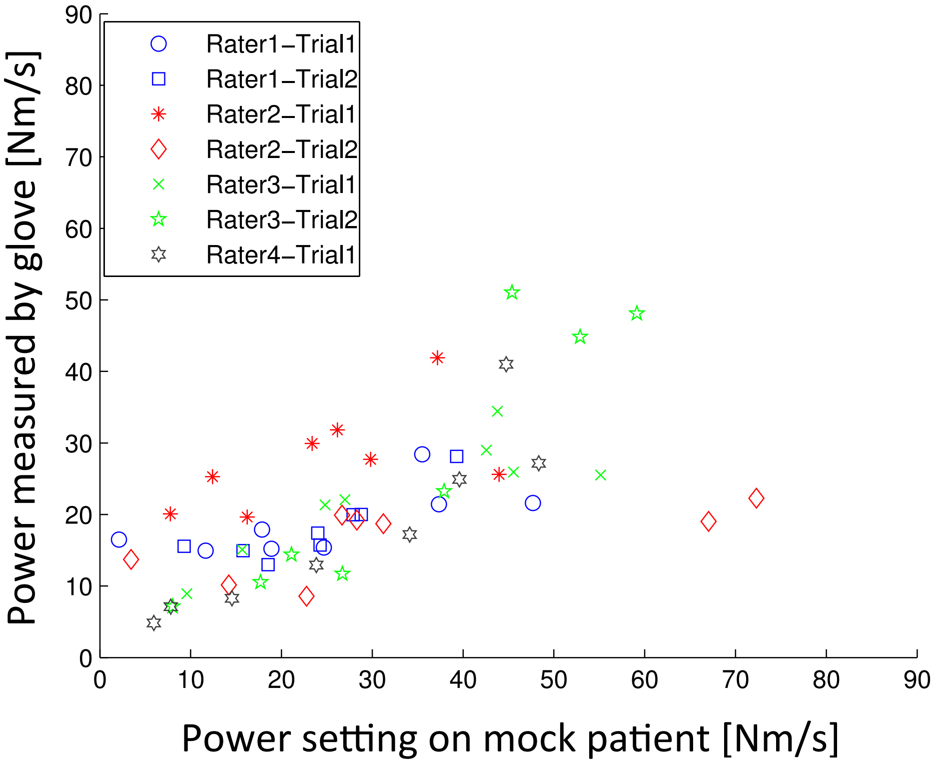


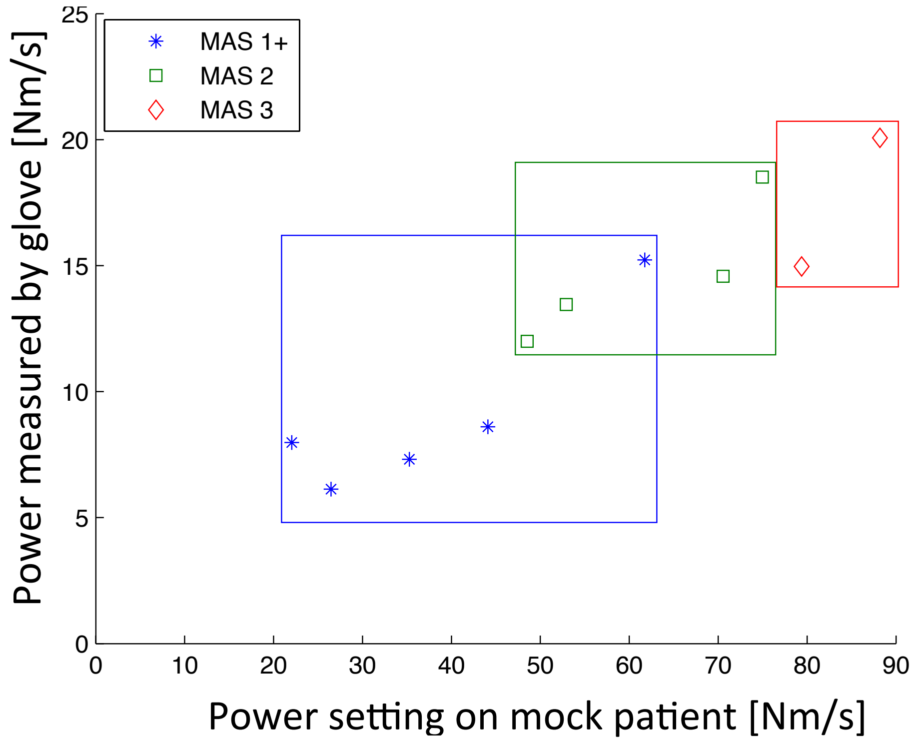


-calibrate sensor glove with a mock patient with changeable resistance (2-20kg)

- load cell to measure force
- potentiometer to measure angular velocity
- the power P=F*v to move the mock limb is recorded

Better resolution than MAS scale





In press, IEEE-NIH 2016 HI-POCT Proceeding

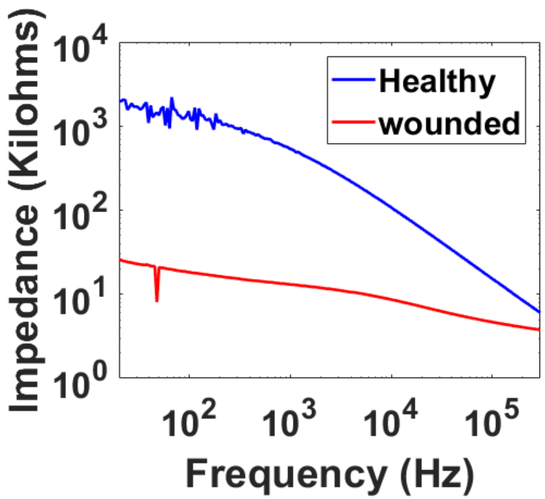
- Quantitative glove measurement allows comparison between rating trials, less dependence on rater perception
- Glove sensor improves the resolution of the spasticity assessment

UC San Diego
Jacobs School of Engineering

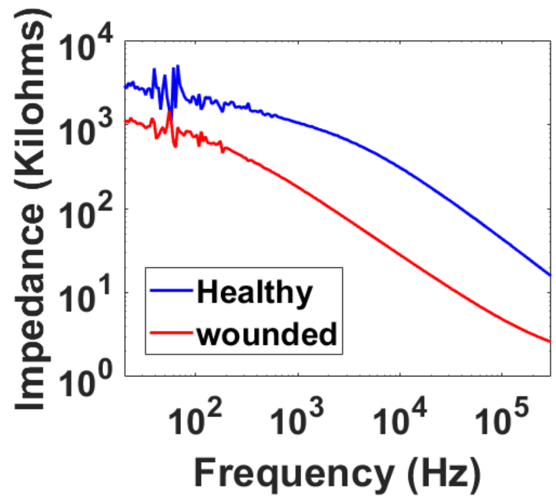
Monitoring wound impedance and electrical stimulation

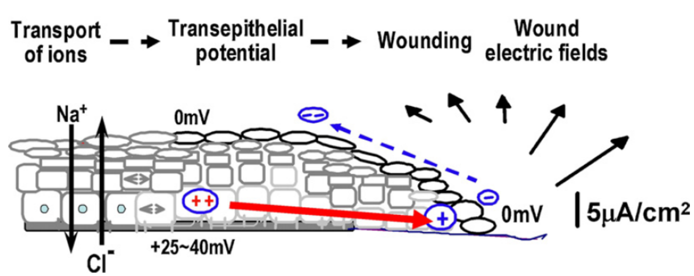
- The impedance between two electrodes was measured before and after wound; the stimulation test is scheduled for Aug.

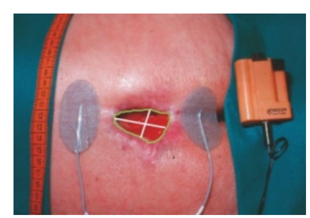
Electrode 1



Electrode 2

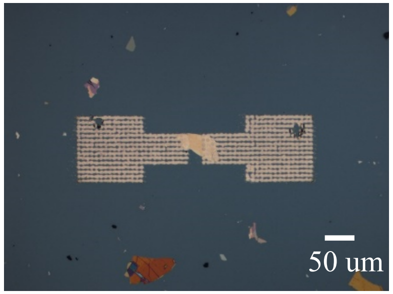


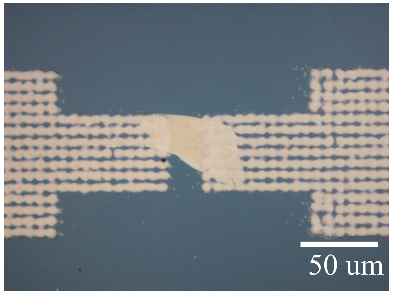




Electrohydrodynamic printing on MoS2 flakes

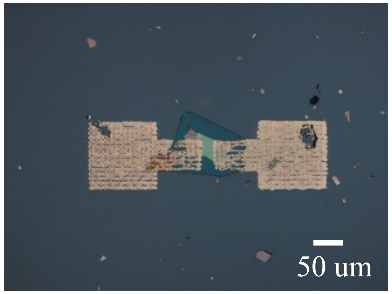
1. MoS2 flake on bare SiO2

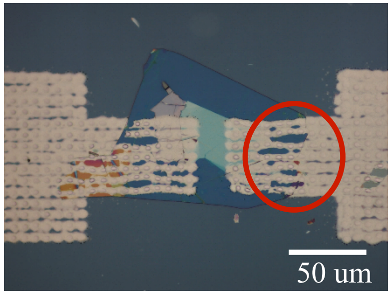




W: (24.0 +32.4)/2=21.3 um
L: 18.5 um
W/L=1.15

2. MoS2 flake on h-BN layered SiO2



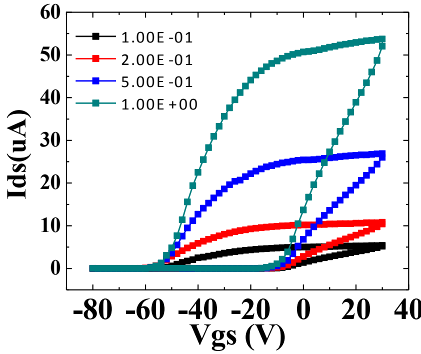


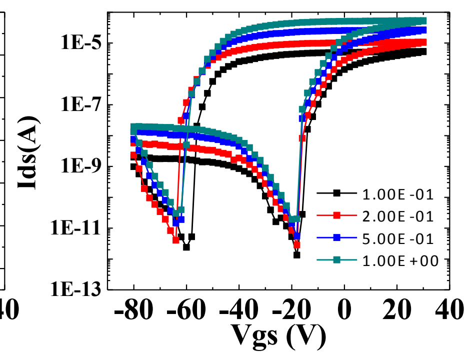
W: (27.8 +32.4)/2=23.1 um
L: 18.5 um
W/L=1.25

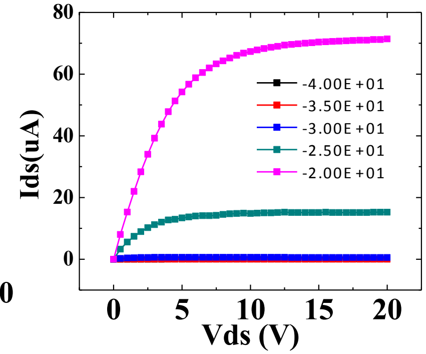
Technically, smaller gate length is possible
Different surface properties causes disconnection in some part (red circle)
Needs information on transferred MoS2 and h-BN layer (thickness=number of layers, cap of h-BN)

MoS2 on h-BN layered SiO2 wafer

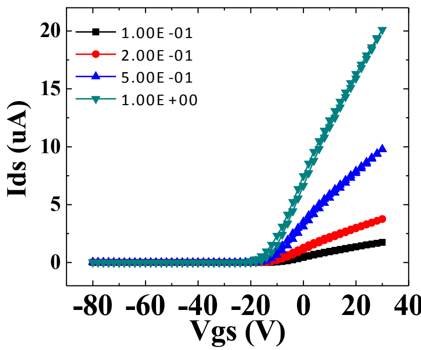
1. On bare SiO2/ Si

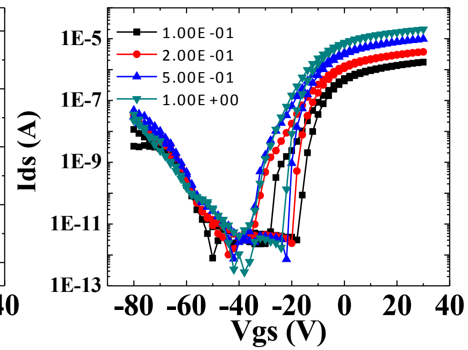


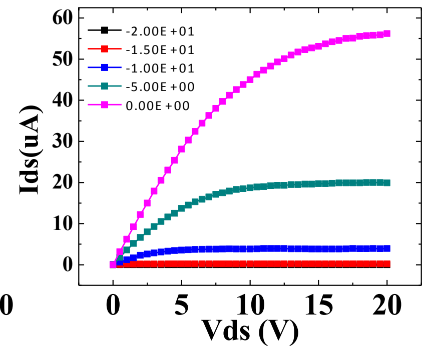




2. On h-BN layered SiO2/Si







- Hysteresis was not observed in the device on h-BN layered Si wafer
- Coulomb scattering originating from SiO2 might have been suppressed by inserted h-BN layer
- More positive threshold in h-BN layered device

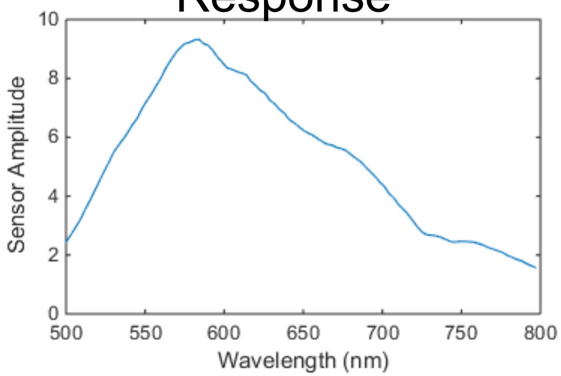
Calibration

$$T(x,y,\lambda)=S(\lambda)\times U(x,y,\lambda)/C(\lambda)$$

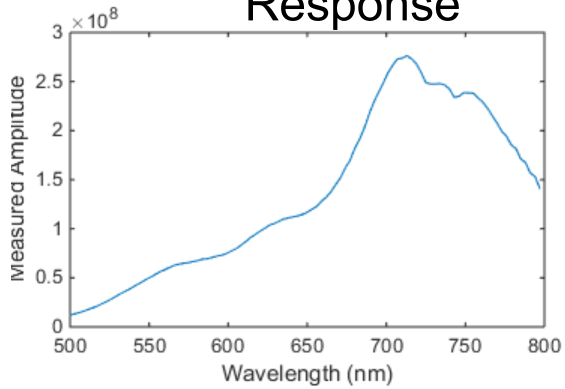
S(λ)=Sensor wavelength response
C(λ)=Camera wavelength response
U(x,y,λ)=camera transmission image of etalon

Etalon Transmission Images

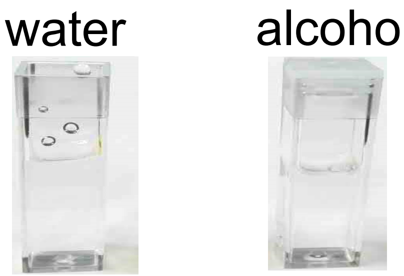
Sensor Response



Camera Response

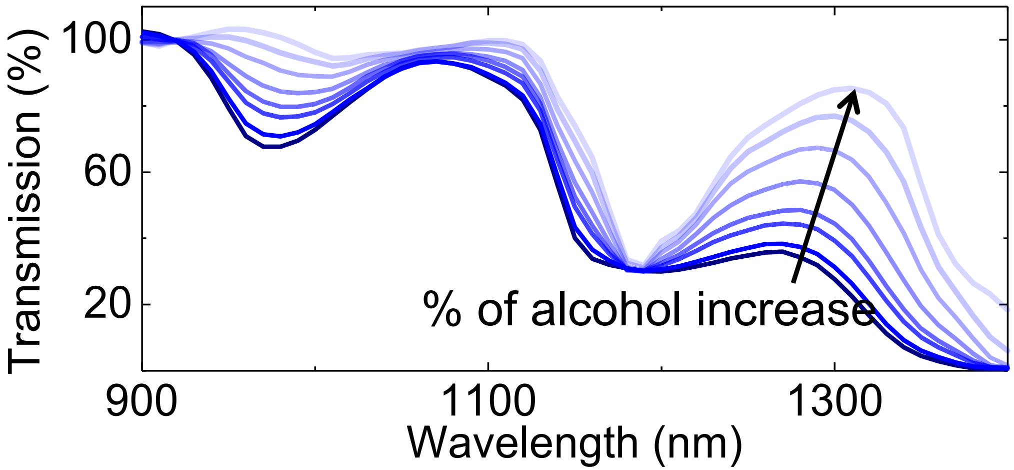


Application example to detect alcohol concentration

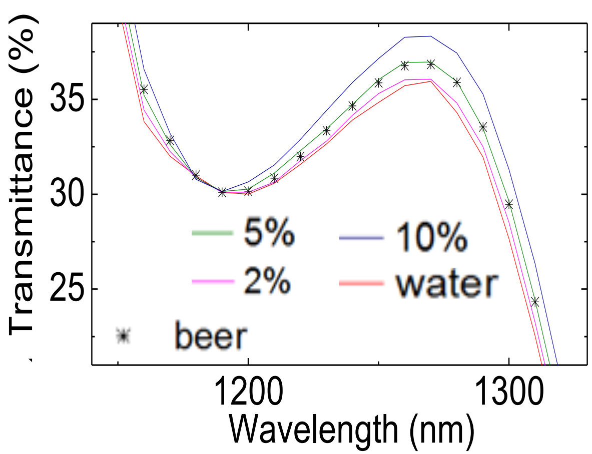


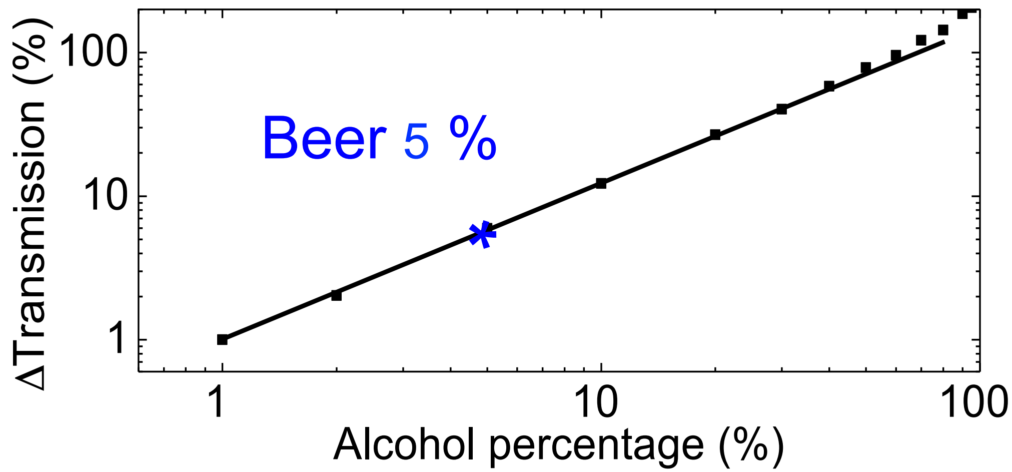
In visible spectrum, both water and alcohol are transparent;
For example want to detect water in gasoline

In SWIR spectrum, water absorption peak responsive to %change near 1300 nm



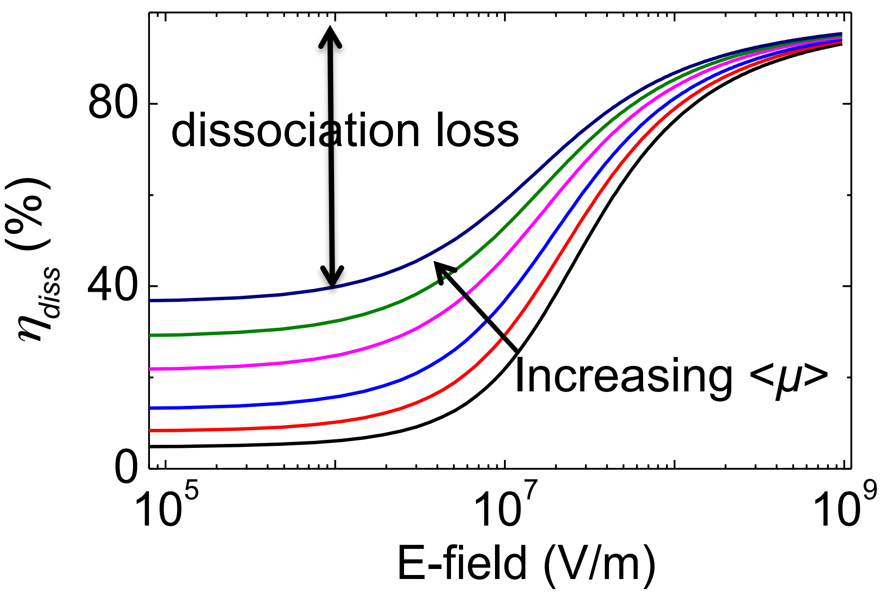
Application example to detect alcohol concentration





Successful determination of chemical composition in SWIR spectrum; to expand into medical sensors

Braun model of exciton dissociation under E-field



$$k_D(E) = \frac{3e\langle\mu\rangle}{4\pi\varepsilon_0\varepsilon_r a^3}\exp(-\frac{E_B}{kT})[1 + b + \frac{b^2}{3} + \frac{b^3}{18} + \dots]$$

$$\eta_{diss} = k_D/(k_D + k_F)$$

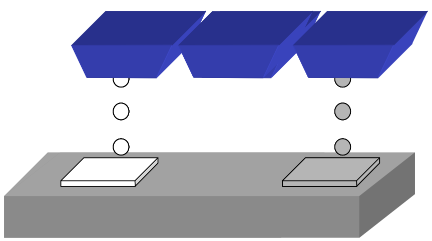
k_D is dissociation rate dependent on E-field
k_F is recombination constant

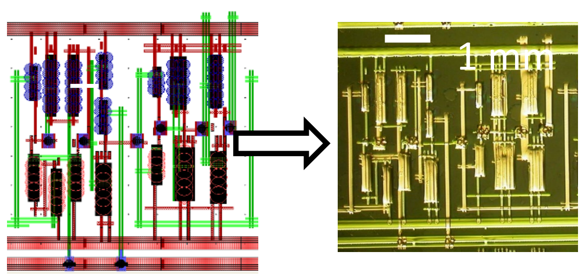
- Higher dissociation efficiency with larger E-field, towards 100% if field is sufficiently high
- We typically operate around low field of 10^6 to 10^7 V/m; efficiency only between 20-40%

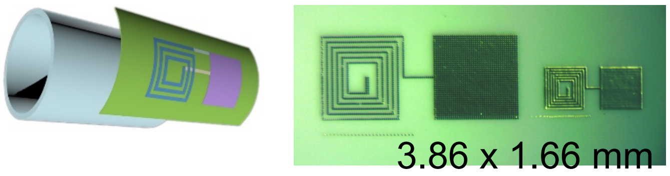
Summary

Examples of printed sensor:

- Dry electrodes for electrophysiology measurement
- Printed transistor circuits for signal modulation
- Quantitative assessment of force in spasticity diagnosis or for robotics







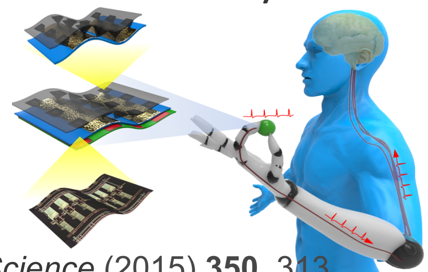
UC San Diego
Electrical and Computer Engineering

Flexible Printed Electronics in the Ng Lab

PI: Tina Tse Nga Ng, Assoc. Prof. in ECE Dept. http://flexible-electronics.ucsd.edu/

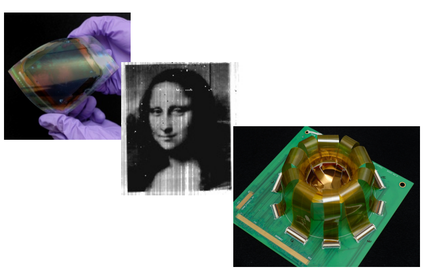
Vision: to achieve *on-demand* digital fabrication, integration of electronics on any surface

Customizable, flexible, biocompatible

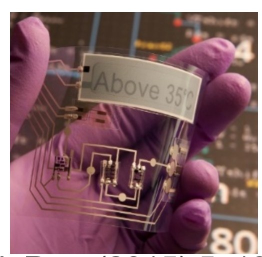


Science (2015) **350**, 313.

Impact: novel electronic materials and devices with new form factor



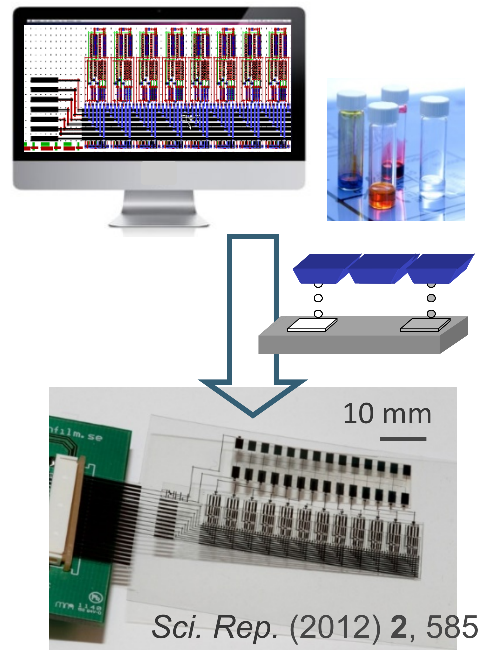
Flexible imager
APL (2008) **92**, 213303.



Sci. Rep. (2015) **5**, 13457.
Low-cost tag to track vaccine temp

Approach: digital fabrication

Direct printing –maskless, low temp

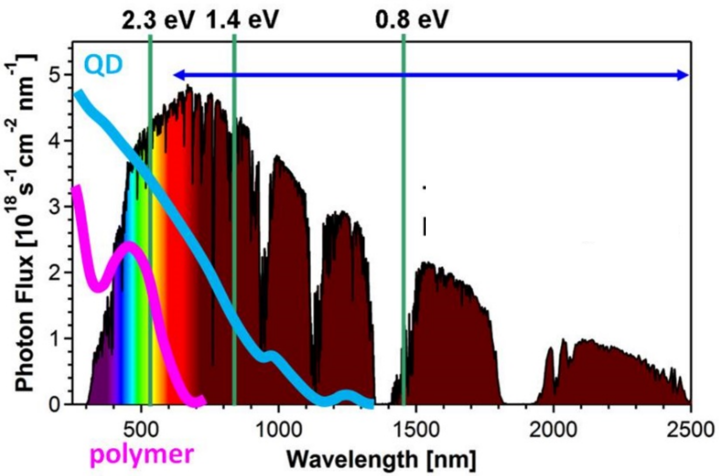


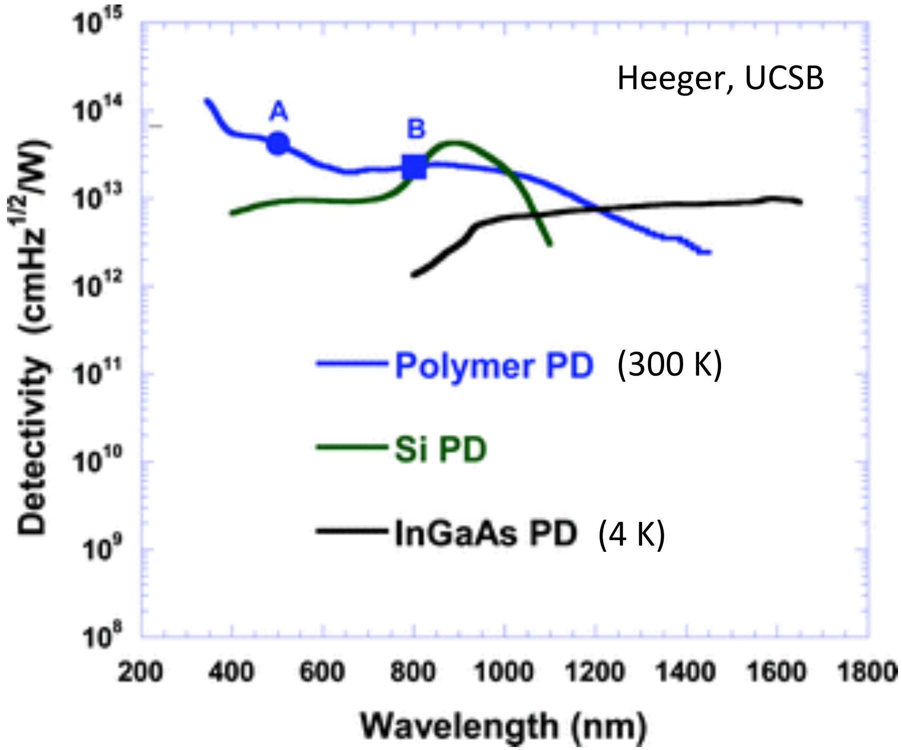
Sci. Rep. (2012) **2**, 585.

Develop design rules, new processes

Infrared imaging applications for organics

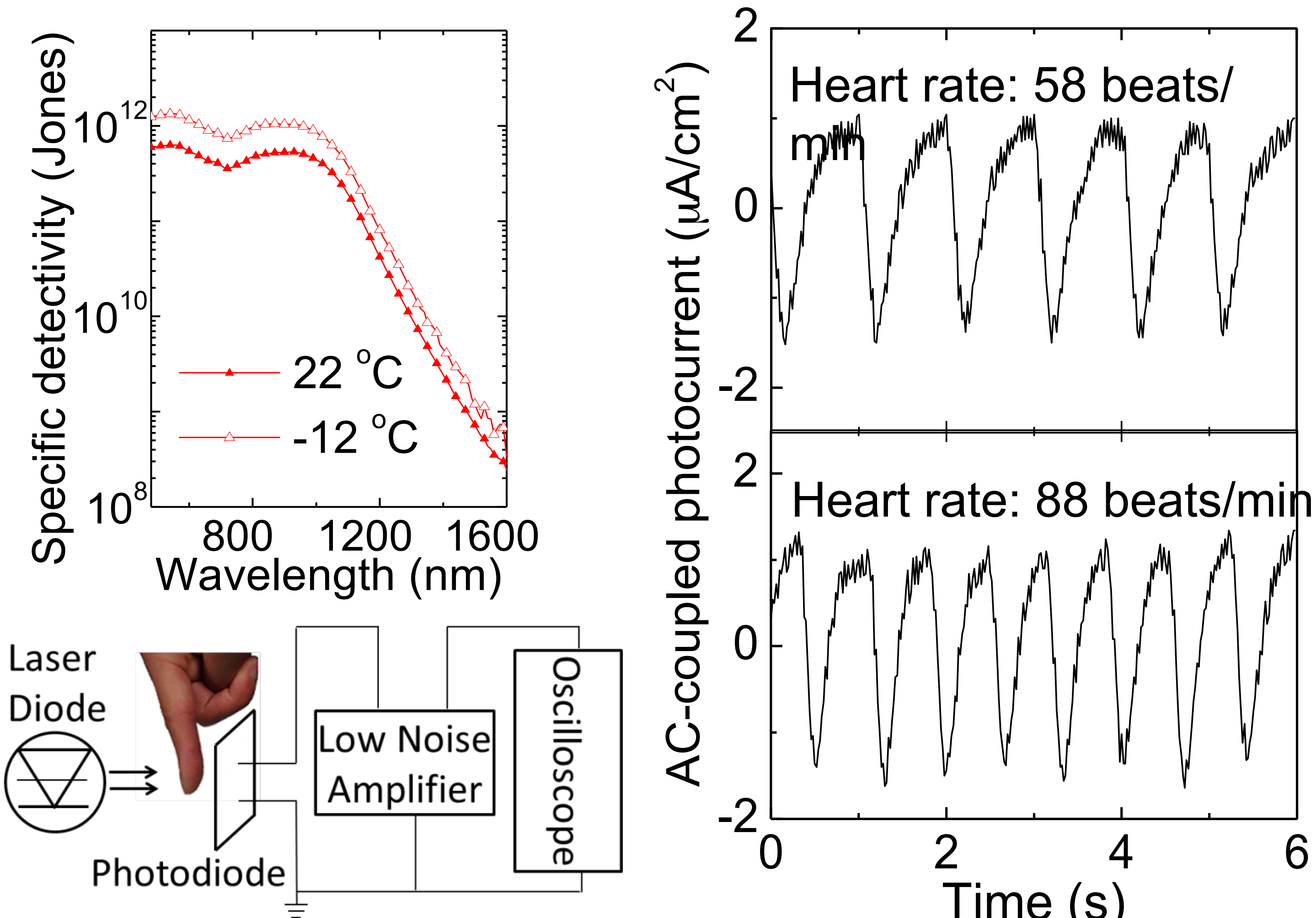
Imaging at different spectral region





At room temp, organic materials has detectivity exceeding conventional III-V compound semiconductors in NIR

TOC graphic

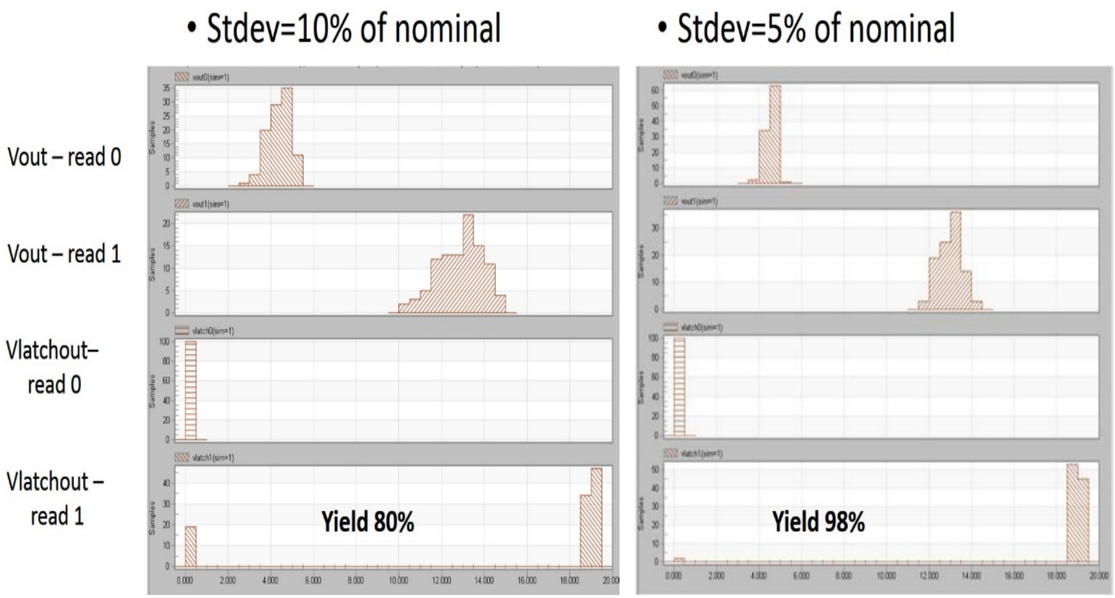


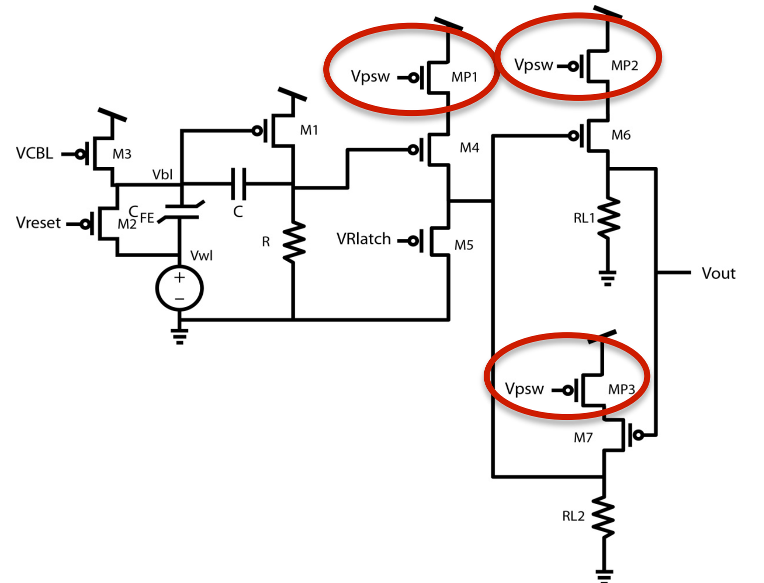
Specific detectivity (Jones)
10^{12}
10^{10}
10^{8}
800
1200
1600
Wavelength (nm)
22 °C
-12 °C
Laser
Diode
Photodiode
Low Noise
Amplifier
Oscilloscope
AC-coupled photocurrent (μA/cm^2)
Heart rate: 58 beats/min
Heart rate: 88 beats/min
2
0
-2
0
2
4
6
Time (s)

Acknowledgment

Grateful to our partners and materials suppliers

Predicted yield of the readout circuit

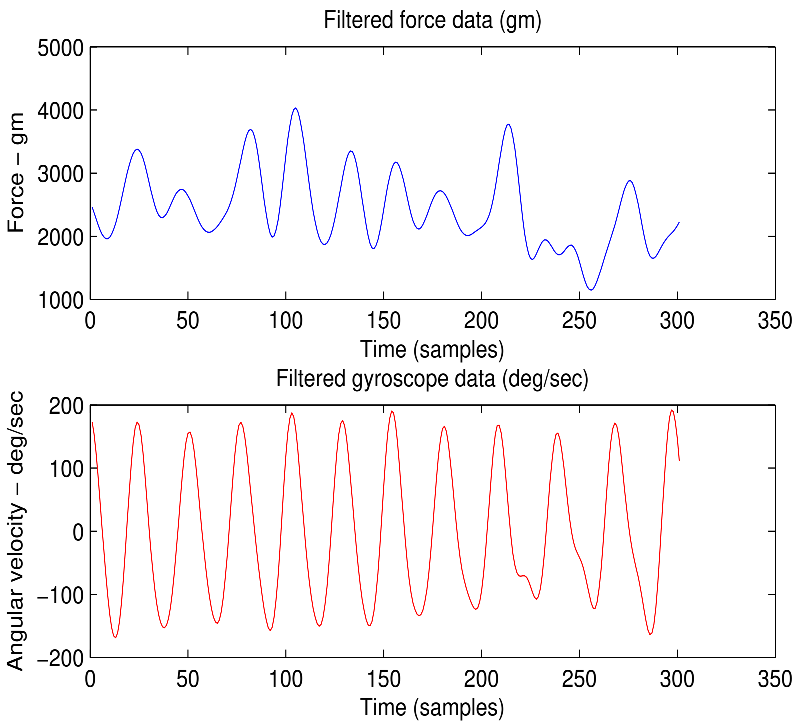




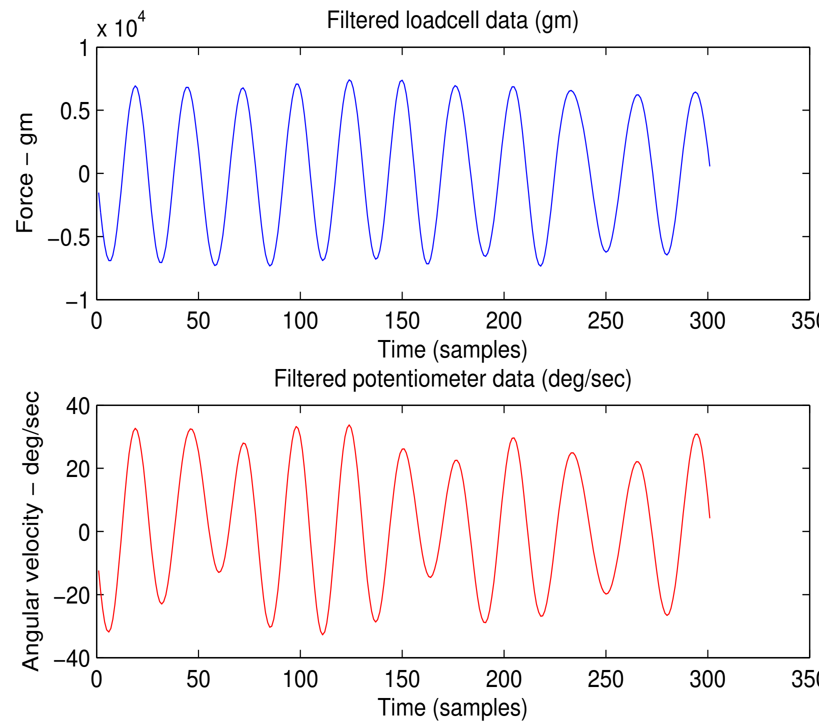
- Predicted yield from Monte Carlo points to 5% std dev
- Add power switches to keep circuit off and minimize V_T shift

Sensor Data

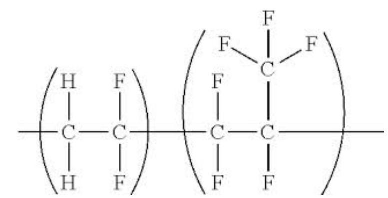
Glove Data
Force and Angular velocity (Gyro)



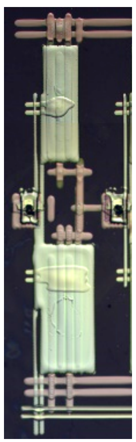
Mock Patient Data
Load cell and Potentiometer

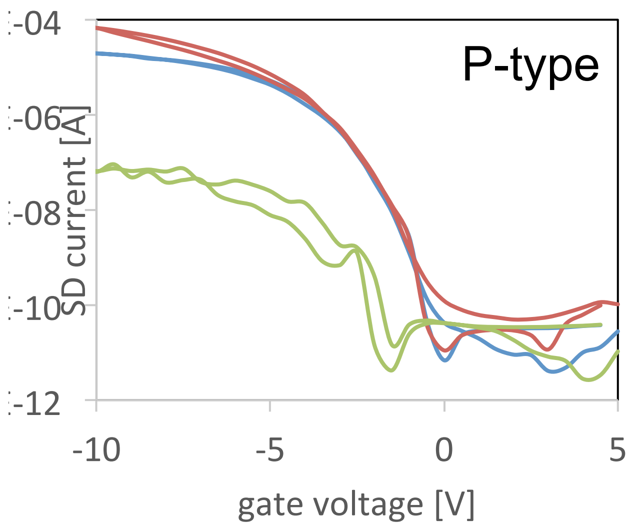


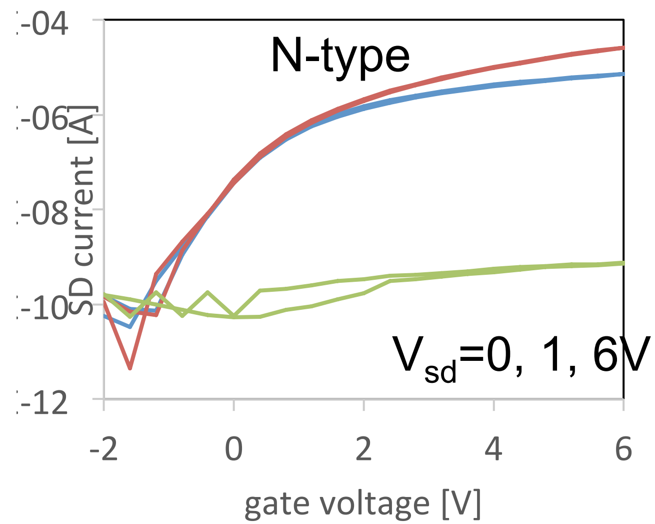
Printed OTFTs operating at ≤5V



High k
up to ~40

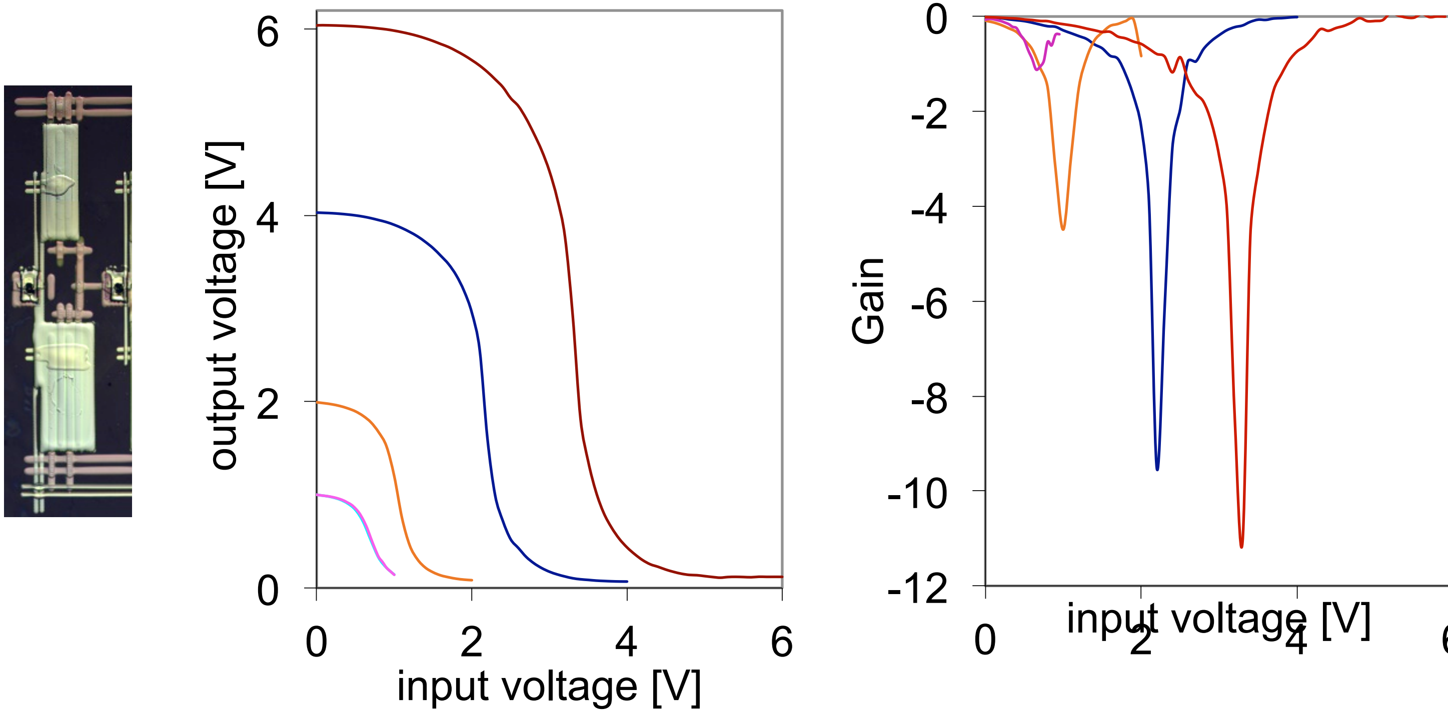






- Dielectric bilayer: Teflon and PVDF-TrFE-CTFE polymer with surfactant
  - -gate capacitance =20nF/cm^2 for 60nm Teflon/600nm PVDF terpolymer
  - -mobility ~0.6 cm^2/Vs for complementary semiconductors

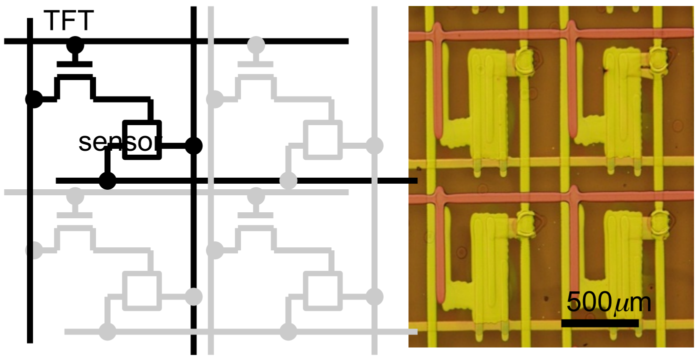
Inverter shows gain of -4 at 2V



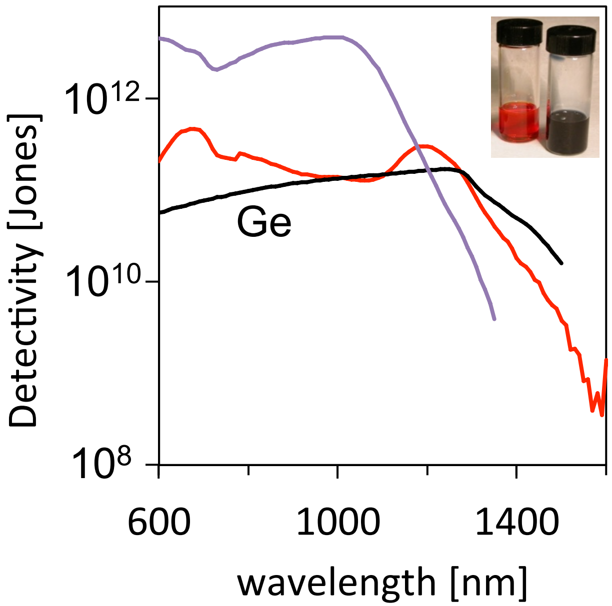
Improved processing shows that the inverters switch properly with gain>1 at Vdd=2V
Ring oscillator works at 3V

Printed sensor systems

Improve TFT and sensors/actuators are foundational

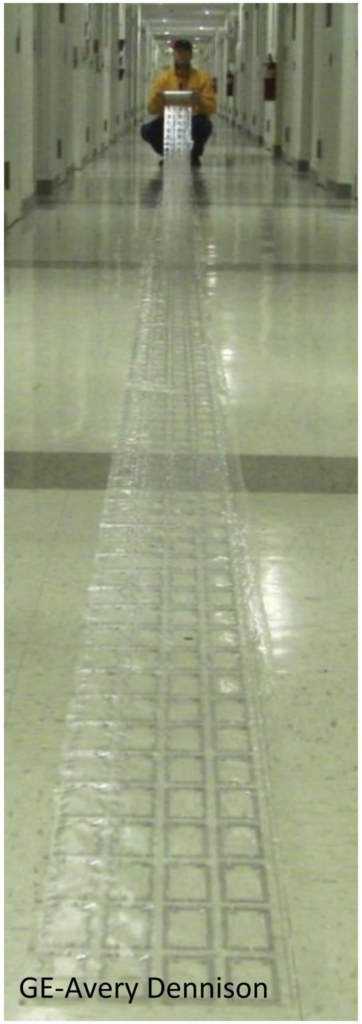


Organic infrared sensors



enable ubiquitous infrared spectroscopy, expand beyond visible wavelength

Applications for printed sensors

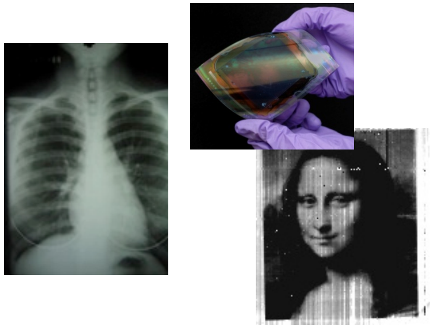


GE-Avery Dennison

- Scalable to large-area, flexible, tunable materials

  Human-computer interface (touch, imager, etc.)
- Multi-component arrays that increase selectivity

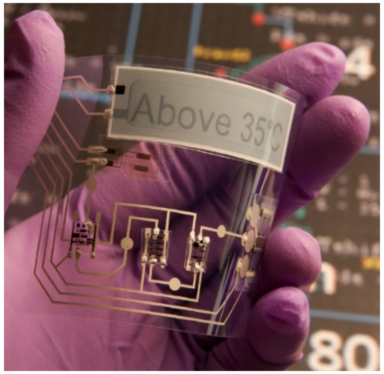
  Low-cost, high-volume for distributed sensing



APL **92** (2008) 213303

Suslick, et al., *Nature Chemistry*, 2009

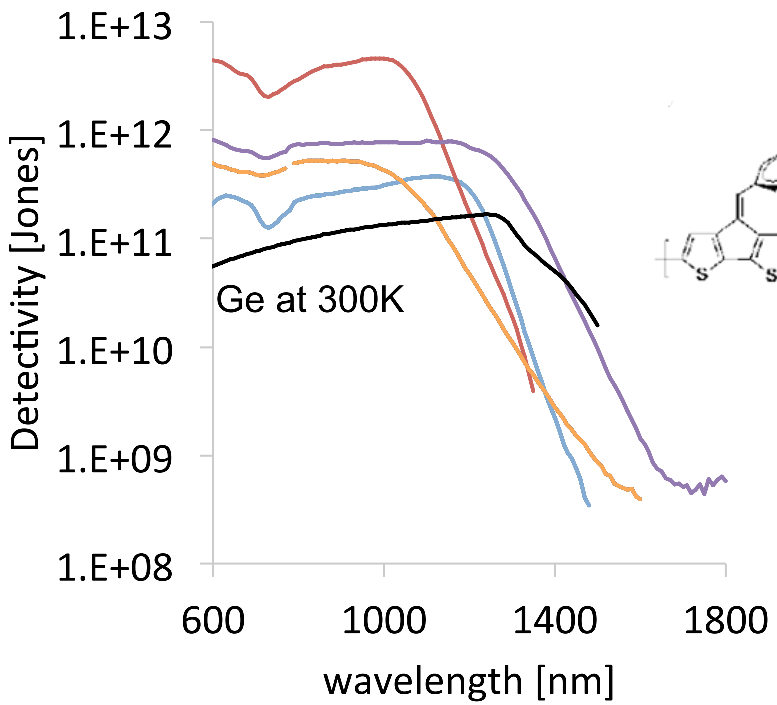
Someya, U Tokyo

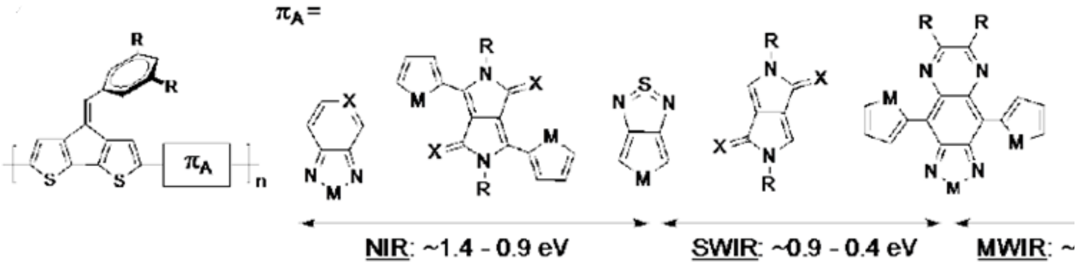


Sci Rep (2015), **5**, 13457.

Solution processed IR sensors exceed conventional Ge

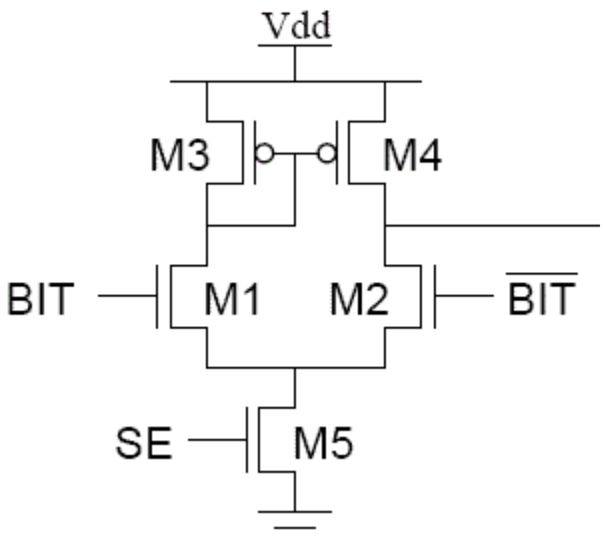
Tunable organic materials with infrared detectivity exceeding commercial Newport Ge diode



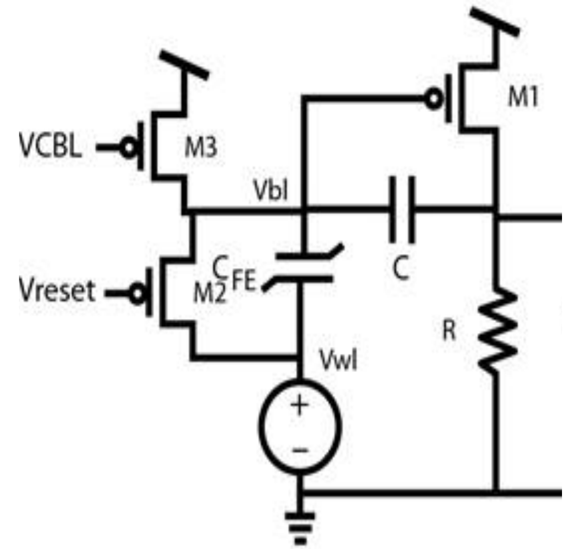


ACS Appl. Mater. Interface 9 (2017) 1654.
Polymer Science, under review.

Low TFT count design



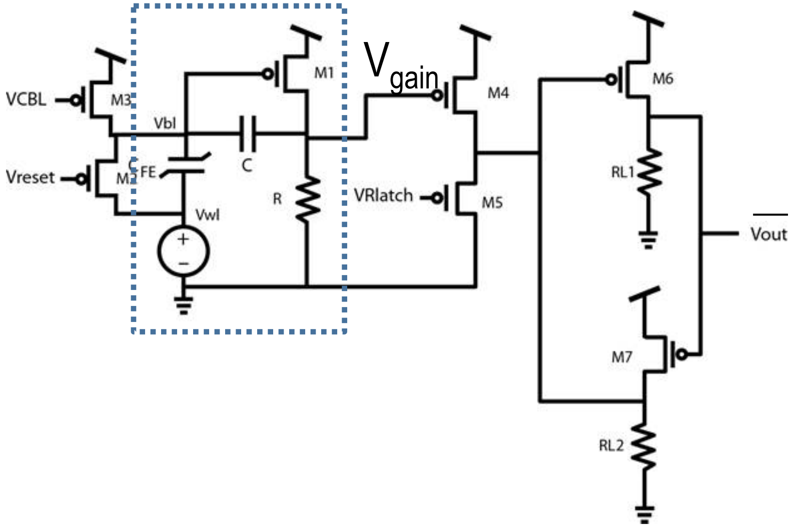
Conventional sense amplifier:
5 TFTs + 2 select TFTs (not shown)

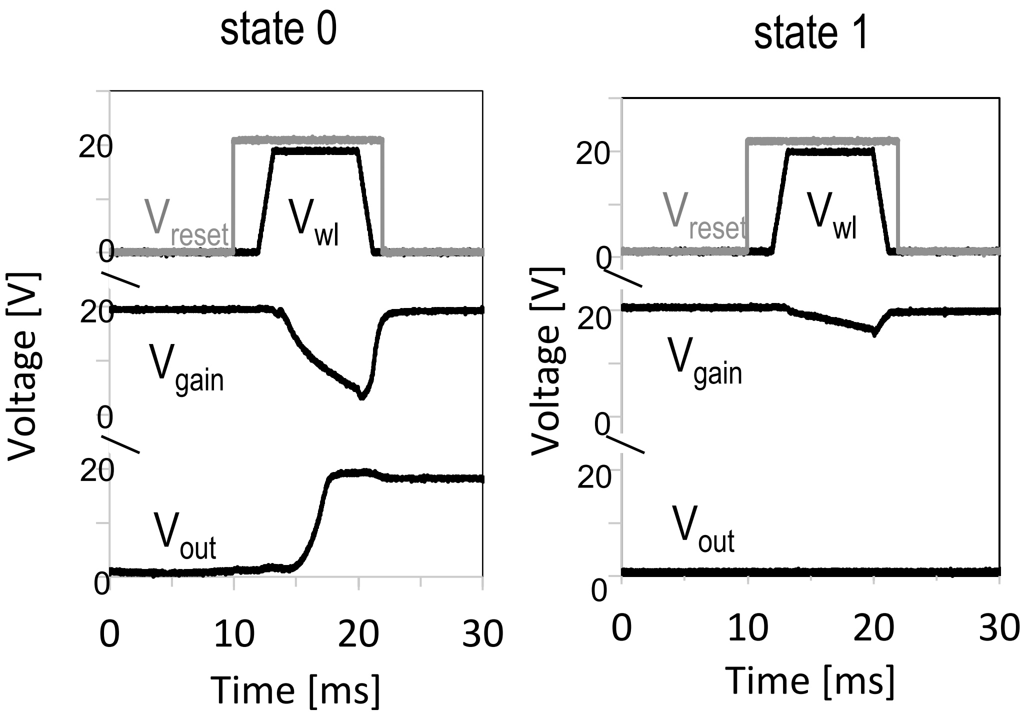


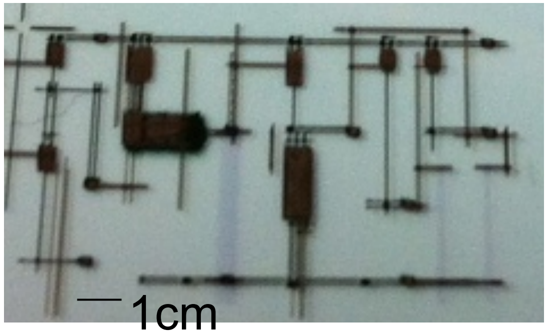
Single TFT gain stage:
1 TFT +2 select

1. Use resistors to avoid bias stress
   ±10 MΩ or ±14% of the nominal value 70 MΩ
2. Single TFT gain stage, reduce TFT count, trade-off with generalizability

Printed gain-stage and latch circuit to read capacitor memory

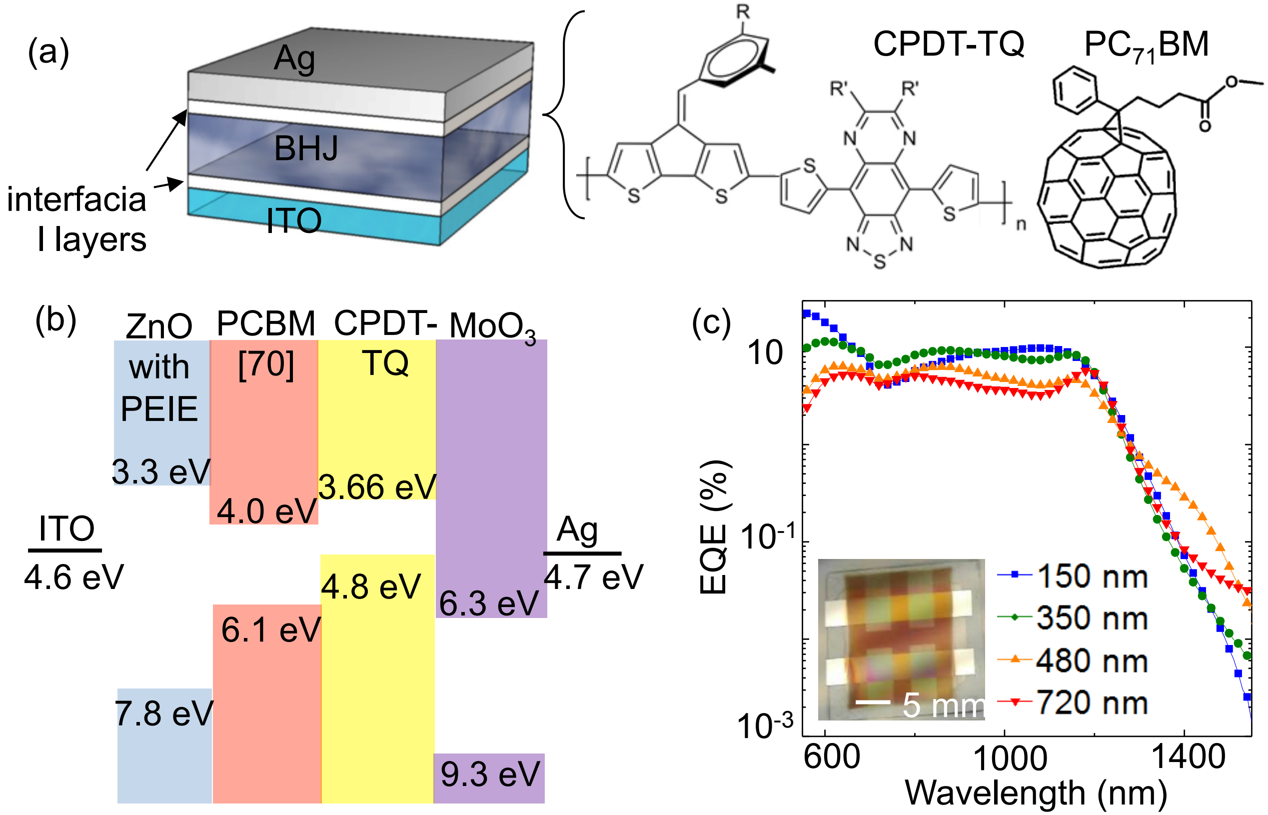






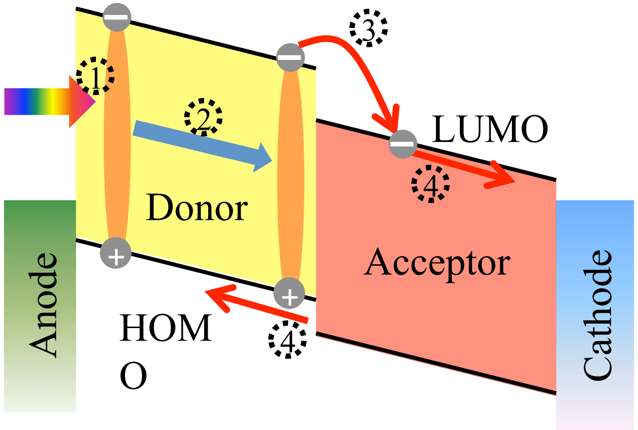
To appear in *IEEE Tran. Elec. Dev.* (special flex elec issue)

Charge in ferroelectric capacitor is amplified and the memory state is distinguished



(a)
Ag
BHJ
ITO
interfacia
l layers
CPDT-TQ
PC71BM
(b)
ZnO with PEIE
PCBM [70]
CPDT-TQ
MoO3
3.3 eV
4.0 eV
3.66 eV
ITO
4.6 eV
Ag
4.7 eV
4.8 eV
6.3 eV
6.1 eV
7.8 eV
9.3 eV
(c)
EQE (%)
Wavelength (nm)
150 nm
350 nm
480 nm
720 nm
5 mm

Device structure and fundamentals

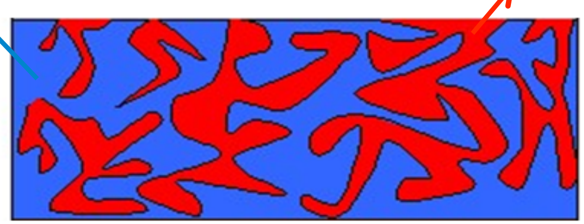


1) Photoexcitation
2) Exciton diffusion
3) Exciton dissociation
4) Carrier transport and collection

Fluorine based derivative **(Acceptor)**

p-type polymers **(Donor)**

BHJ



D phase + A phase

↓

e and *h* transport in two separate channels

↓

Low dark current