Metal Oxide Based Silicon Heterojunctions

Martin Bivour / Workshop on Passivating Contacts 2018

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Fraunhofer Institute for Solar Energy Systems ISE

Advancing PV: from passivation to contacts – A passivating contact workshop

Eindhoven, 31 January 2018

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AGENDA

- Introduction: Passivating (AND carrier-selective) contacts based on high / low work function thin films
- Material screening: Importance of induced c-Si junction
- MoO_x based hole contacts: Simulation, fabrication and selectivity issues

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Passivating (AND Carrier-Selective) Contacts

Optically Transparent / Passivating / Selective for e or h

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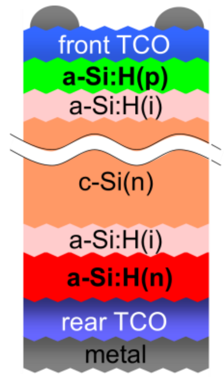
- a-Si:H / c-Si heterojunction is prime example
  - Champion efficiencies for c-Si solar cells
- Challenges: doped a-Si:H
  - High parasitic absorption (J_{sc})
  - Poor doping efficiency ($\rho_{contact}$)
  - PECVD, toxic gases (costs)

→ Alternatives to doped a-Si:H?



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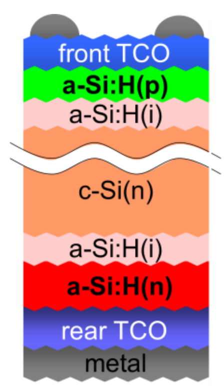

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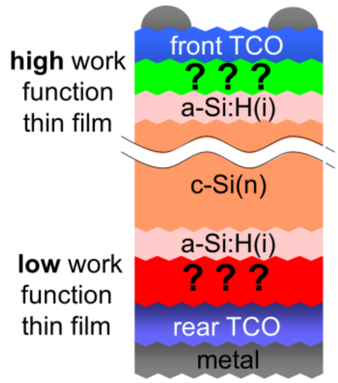

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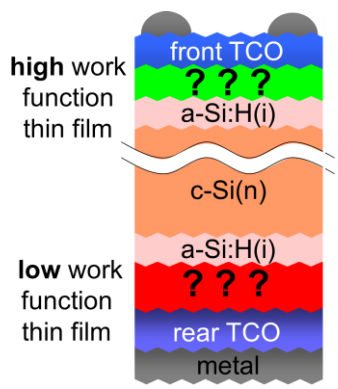
  - Metals: only moderate WF range and optics
  - Oxides of some metal are interesting candidates



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"Contac Layers" Featuring High / Low Work Functions

Organic Absorber Devices (OLED, OPV)

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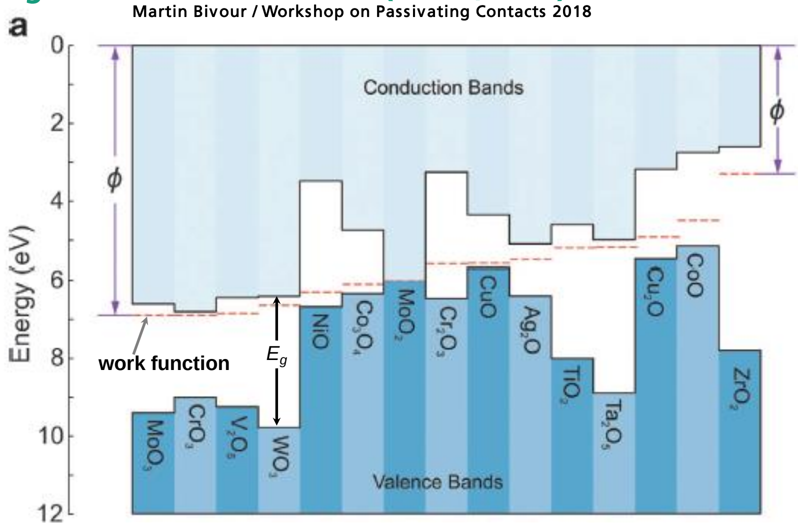

Greiner et al., "Thin-film metal oxides in organic semiconductor devices: their electronic structures, work functions and interfaces" NPG Asia Mater 5, 2013

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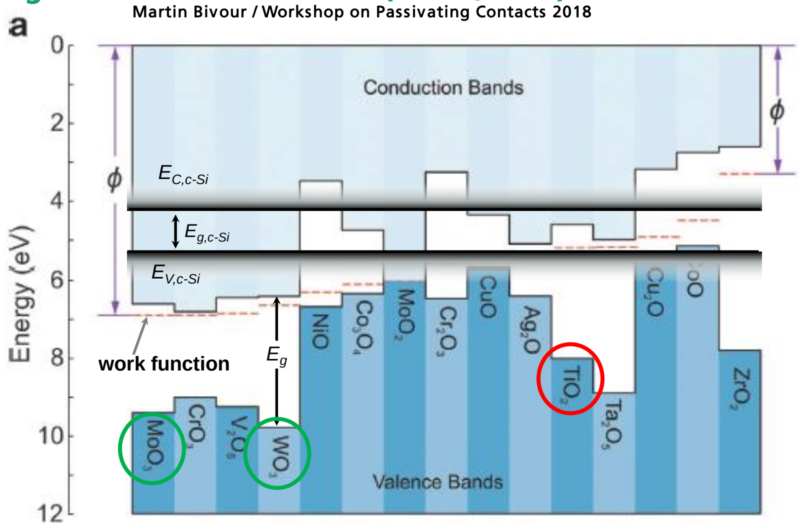

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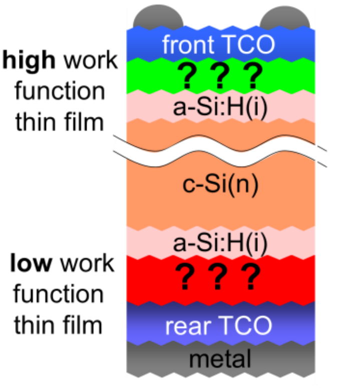
- High / low work function thin film?
- → High band gap transition metal oxides
- → Calls for an efficient work function engineering



Transition Metal Oxides for c-Si Solar Cells
Simulation - TCO / a-Si:H(p) Si Heterojunction

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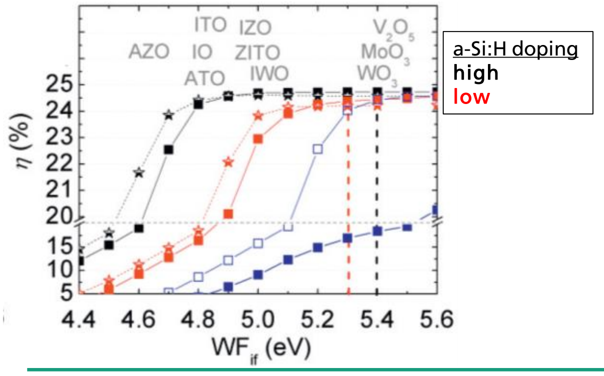
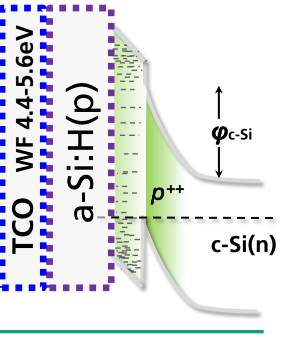
- Metal oxide work function engineering for the hole contact

1) TCO / a-Si:H(p) contact improvement – thinner and less doped films





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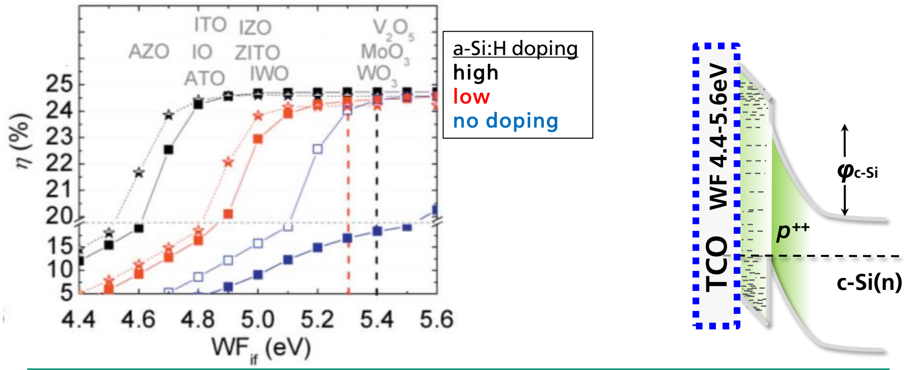
- Metal oxide work function engineering for the hole contact

1) TCO / a-Si:H(p) contact improvement – thinner and less doped films
2) Replacing a-Si:H(p)



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Transition Metal Oxides for c-Si Solar Cells

ITO / MoO_x Front Contact - First Experimental Results

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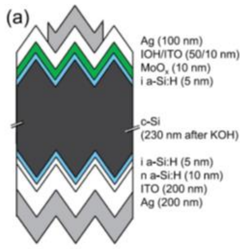
- a-Si:H(p) replaced by MoO_x
  - Encouraging results



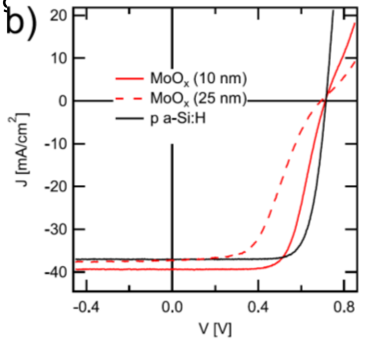


TABLE I. Summary of cell performance parameters.

Hole contact	V_{oc} (mV)	J_{sc} (mA/cm^2)	FF (%)	Efficiency (%)
MoO_x	711	39.4	67.2	18.8
p a-Si:H	716	37.1	75.9	20.2

Battaglia et al., "Silicon heterojunction solar cell with passivated hole selective MoOx contact", APL 104, 2014

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Transition Metal Oxides for c-Si Solar Cells

Scope of Application for MoO_x, WO_x and V_2O_x

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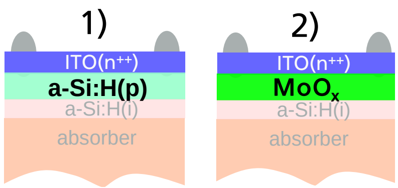

		V_{oc}	J_{sc}	*FF*	*Eta*	Finding
		mV	mA/cm²	%	%	
1	a-Si:H(p)	719	**31.8**	79.6	17.9	-
2	**MoO_x**	712	**33.3**	80.4	19.0	Replacement a-Si:H(p)

Bivour et al., "Molybdenum and tungsten oxide: High work function wide band gap contact materials for hole selective contacts of silicon solar cells", SOLMAT 142, 2015
Bivour et al., "High work function metal oxides for the hole contact of silicon solar Cells, 43rd PVSC, 2016

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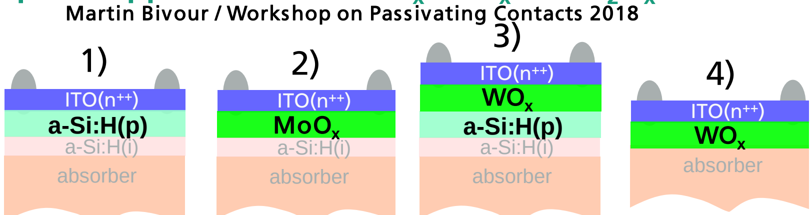

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2	**MoO_x**	712	**33.3**	80.4	19.0	Replacement a-Si:H(p)
3	a-Si:H(p) / **WO_x**	719	29.9	**81.1**	17.5	ITO / a-Si:H(p) improvement
4	**WO_x**	661	33.6	80.1	17.8	Decent passivation

Bivour et al., "Molybdenum and tungsten oxide: High work function wide band gap contact materials for hole selective contacts of silicon solar cells", SOLMAT 142, 2015
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TiO_x / Al Rear Contac

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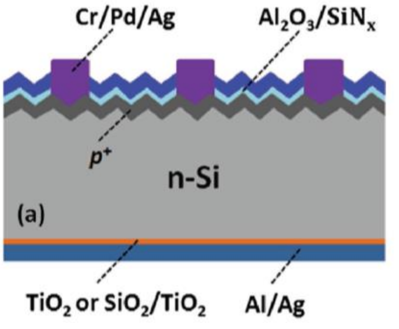
- Al rear contact improved by adding TiO_x or SiO_x/TiO_x below Al



Rear contact type	V_{oc} [mV]	J_{sc} [mA cm^{-2}]	FF [%]	η [%]
Al	585	38.5	78.2	17.6
TiO_2/Al	638	39.2	79.1	19.8
SiO_2/TiO_2/Al	676	39.6	80.7	21.6

Yang et al., "High-Performance TiO2-Based Electron-Selective Contacts for Crystalline Silicon Solar Cells", Adv Mater 28(28), 2016

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Transition Metal Oxides for c-Si Solar Cells

ITO/ MoO_x Front and TiO_x / LiF / Al Rear Contac

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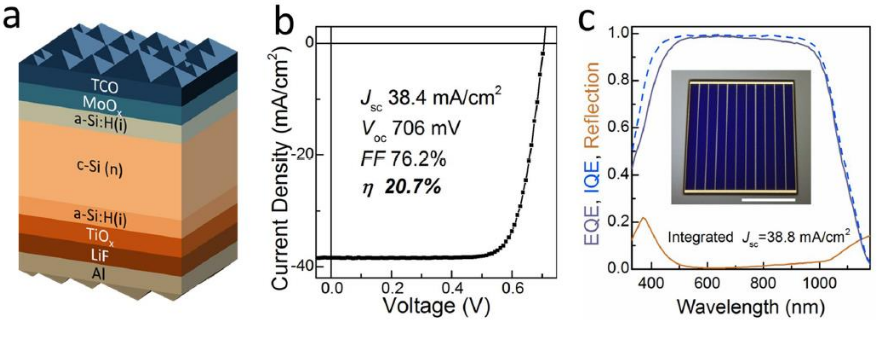
- Champion cell with metal oxides on both contacts
  - Challenges *FF*, rear reflector



Bullock et al., "Stable Dopant-Free Asymmetric Heterocontact SiliconSolar Cells with Efficiencies Above 20%", Accepted: ACS Energy Letters, Jan 2018

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Transition Metal Oxides for c-Si Solar Cells
Hot Topic in the Field of Silicon PV
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Plenty of papers published by other institutes

- UPC
  - G. Gerling
- HZB
  - M. Mews, …
- TU/e
  - B. Macco, J. Melskens
- UNSW
  - C. Lee
- …

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Renaissance of MIS and SIS-Like Contacts
Heterojunction + Induced c-Si Junction
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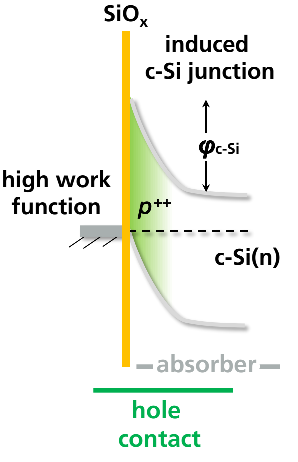
- Explored as alternative to homojunctions in 70´s[1]



[1]Shewchun et al., IEEE Trans. Electron Devices 27(4), 1980

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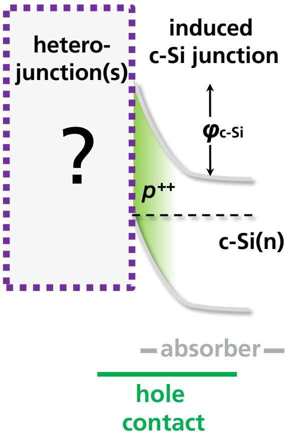
- Explored as alternative to homojunctions in 70´s[1]
- Renaissance by taking advantage of:

1) Improved passivating layers
2) High / low WF thin films[2] adapted from:
   - Organic electronics[3]
   - Thin film solar cells
   - Cathodes/anodes water splitting[4] (H_2/O_2)
   - (Schottky) barrier-height engineering / Fermi level depinning in MOS[5,6], e.g. source / drain contacts and Schottky-gates



[1]Shewchun et al., IEEE Trans. Electron Devices 27(4), 1980
[2]Bullock et al.,IEEE XXXX, 2016
[3]Chen et al., J. Mater. Chem. 20 (13) , 2010
[4]Walter et al., *Chem. Rev.* , 110 (11), 2010
[5]Larson et al., IEEE Trans. Electron Devices 53 (5) , *2006*

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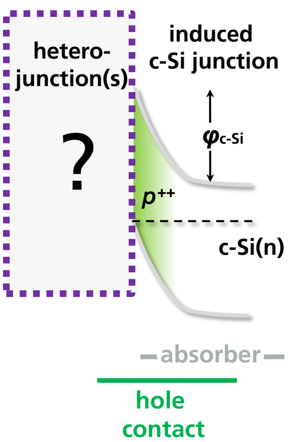
→ Adapted optimization strategies considering "non-classical" losses can be helpful for engineering such heterojunction



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Passivating (AND Carrier-Selective) Contacts
Three Cases
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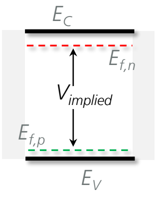
A) passivating



- Gen. / Rec. define implied voltage[1] (iV)

21 [1]Textbook S. M. Sze, Semiconductor devices, Physics and Technology, 2002

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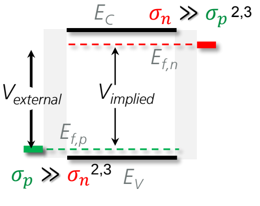
A) passivating + selective



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- Negligible gradient $E_{f,maj}$ in contact region[1,2]

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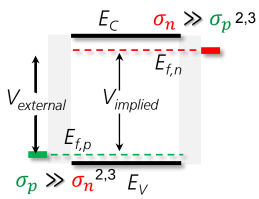
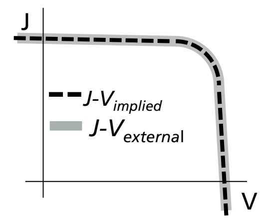


- Gen. / Rec. define implied voltage[1] (*iV*)
- Negligible gradient $E_{f,maj}$ in contact region[1,2]
- → **External voltage matches implied voltage**
- → **"Classical" diode losses**



$$J = J_{gen} - \boldsymbol{J_0}\left\{\exp\left(\frac{q\,V_{implied} + J\boldsymbol{R_S}}{k_B T}\right) - 1\right\} - \frac{qV_{implied} + J\boldsymbol{R_S}}{R_{SH}}$$

23

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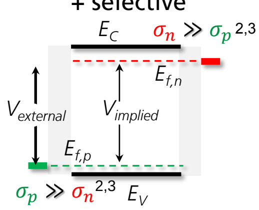
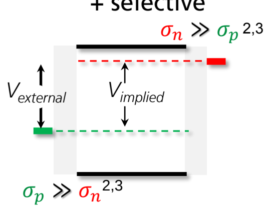


B) poor-passivation
+ selective



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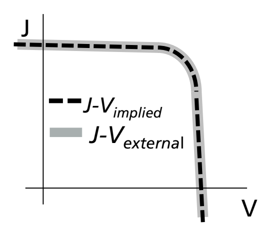

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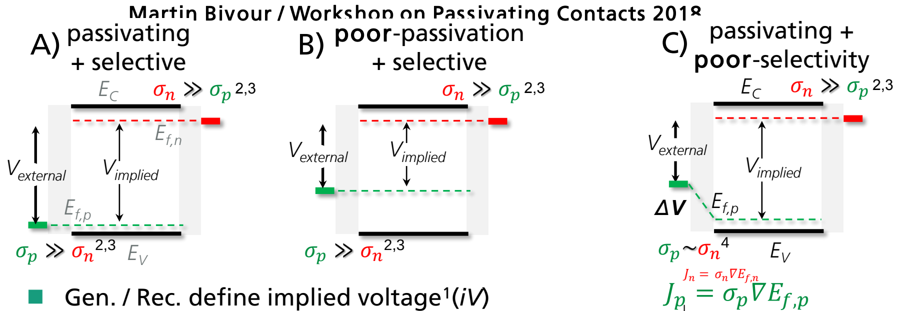
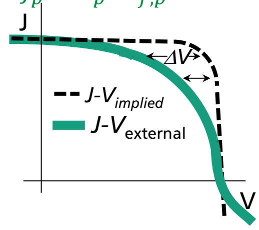


- Gen. / Rec. define implied voltage[1] (iV)
- Significant gradient $E_{f,maj}$ in contact region[1,2]
- → External voltage <u>below</u> implied voltage[4]
- → S-shaped JV, unusual T dependence, …



$$J = J_{gen} - J_0 \cancel{\left\{\exp\left(\frac{qV_{implied} + JR_S}{k_B T}\right) - 1\right\}} - \cancel{\frac{qV_{implied} + JR_S}{R_{SH}}}$$

25

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[3]U. Würfel et al., IEEE-JPV 99, 2014
[4]PhD thesis Bivour, University of Freiburg 2015

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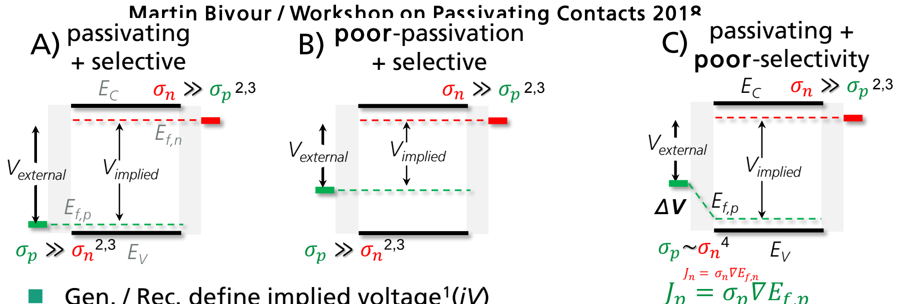

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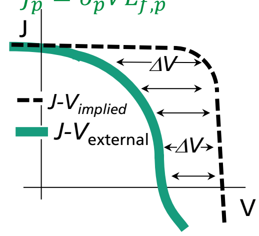

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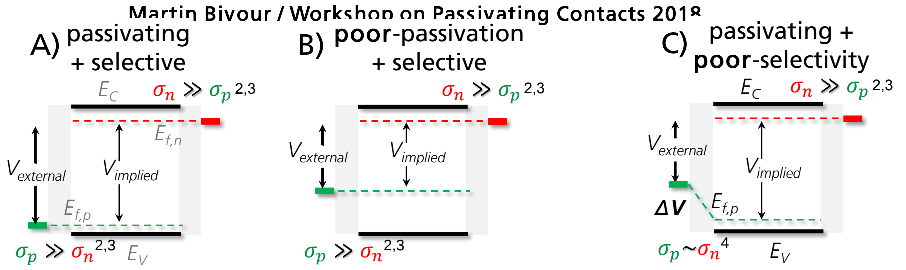

- Non-selective contacts unusual in homojunction Si devices
  - High doping efficiency of p^+ and n^+ regions ($\sigma_p = q\mu_p \boldsymbol{p} \gg \sigma_n = q\mu_n \boldsymbol{n}$)
- More common in thin film, OPV[1] and perovskite solar cells
  - Poor / no doping
  - Selectivity from work function matching / band alignment

→ Also observed in "novel" silicon heterojunctions

[1]Spiesl et al., "On the Impact of Contact Selectivity and Charge Transport on the Open-Circuit Voltage of Organic Solar Cells", *Adv. Energy Mater. 2017, 7, 1601750.*

Passivating (AND Carrier-Selective) Contacts
Simulation Metal Semiconductor Contact

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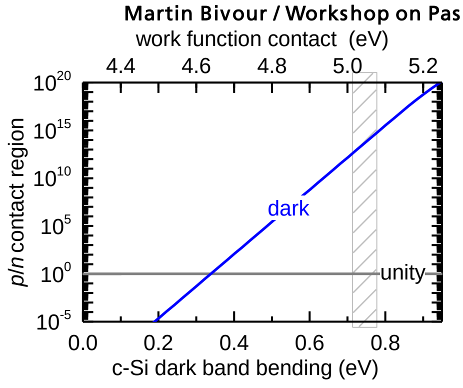
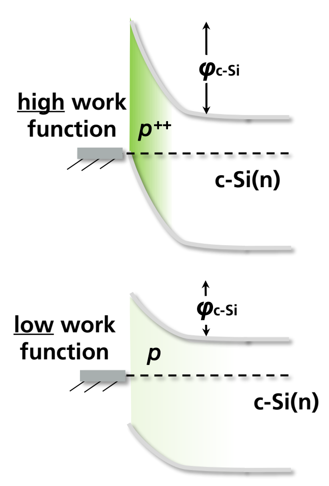
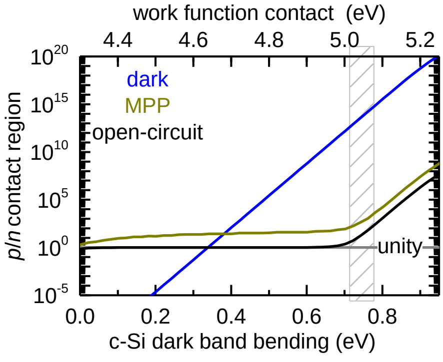
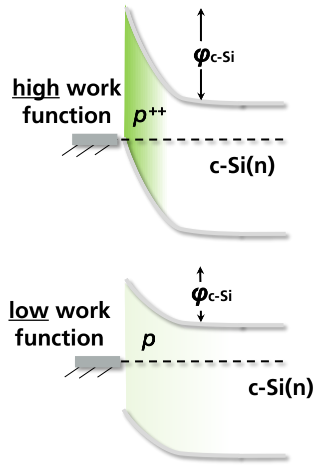




Passivating (AND Carrier-Selective) Contacts
Simulation Metal Semiconductor Contact
Martin Bivour / Workshop on Passivating Contacts 2018





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Passivating (AND Carrier-Selective) Contacts
Alternative Optimization Strategy
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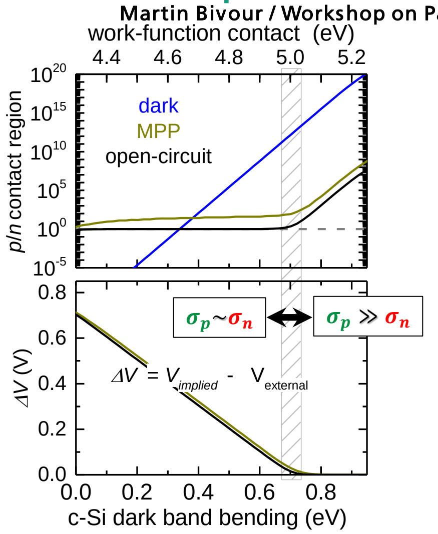

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Passivating (AND Carrier-Selective) Contacts

Alternative Optimization Strategy

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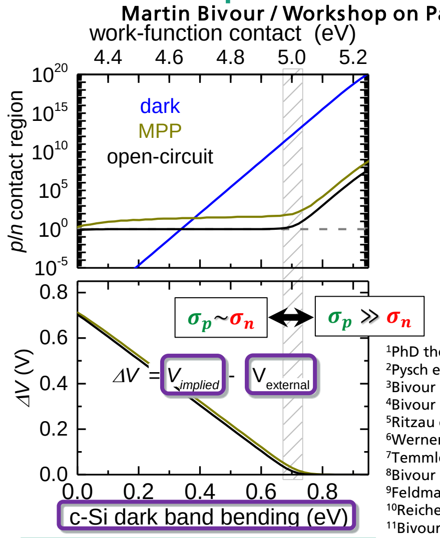

- Considering "non-classical" losses for engineering such heterojunction
  - V_{oc}, iV_{oc}, $\varphi_{c\text{-}Si}$ easily measurable for Si devices
- → Helps to identifying contacts limited by poor selectivity[1-XX]

[1]PhD thesis Bivour, University of Freiburg 2015
[2]Pysch et al., APL 110, 2011
[3]Bivour et al., SOLMAT 106, 2012
[4]Bivour et al., IEEE JPV 4(2), 2014
[5]Ritzau et al., SOLMAT 131, 2014
[6]Wernerus et al., Energy Procedia, 55, 2014
[7]Temmler et al., Proceedings 29th EU PVSEC, 2014
[8]Bivour et al., SOLMAT 142, 2015
[9]Feldmann et al., Energy Procedia, 77, 2015
[10]Reichel et al., APL 118, 2015
[11]Bivour et al., Energy Procedia, 92, 2016
[12]Bivour et al., Energy Procedia, 124, 2017
[13]Neusel et al., Energy Procedia, 124, 2017
[14]Bivour et al., Proceedings 33th EU PVSEC, 2017
[15]Reichel et al., APL 123, 2018

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AGENDA

- Introduction: Passivating (AND carrier-selective) contacts based on high / low work function thin films
- Material screening: Importance of induced c-Si junction
- MoO_x based hole contacts: Simulation, fabrication and selectivity issues

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Material Screening
Metal Contact Layers
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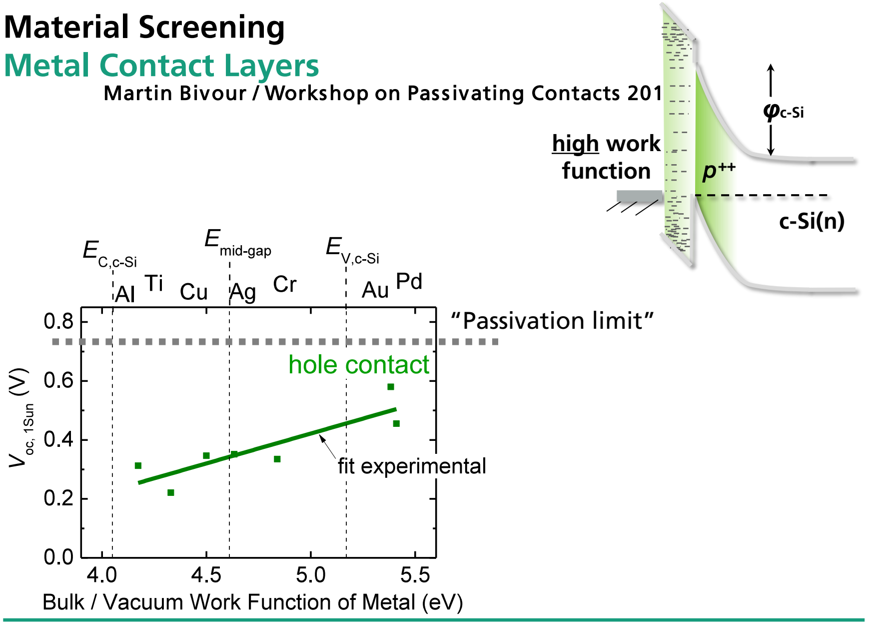

33 Data: PhD thesis Bivour, University of Freiburg 2015

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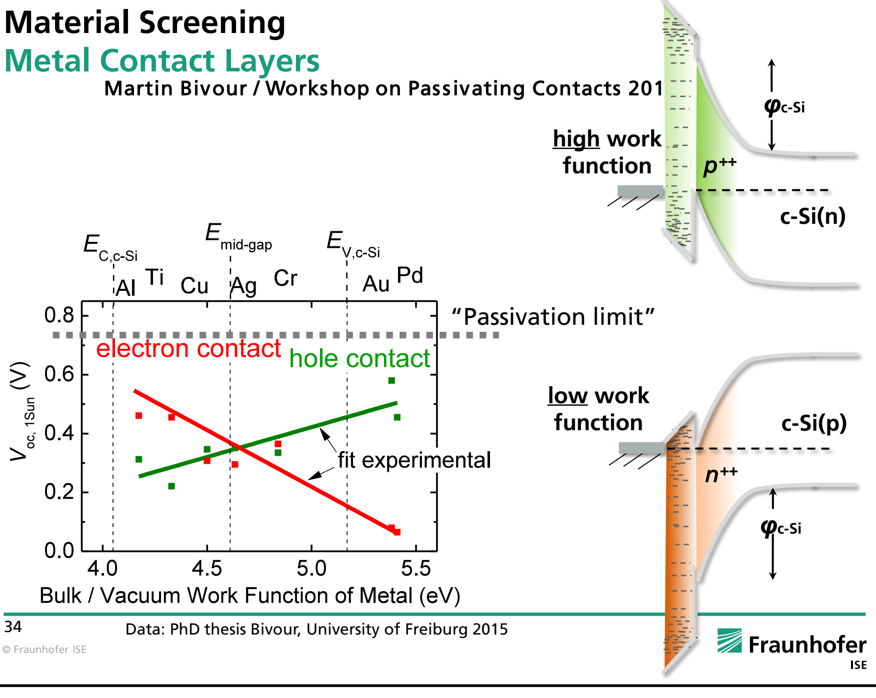
Material Screening
Metal Contact Layers
Martin Bivour / Workshop on Passivating Contacts 201



34 Data: PhD thesis Bivour, University of Freiburg 2015

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Material Screening
Metal Contact Layers

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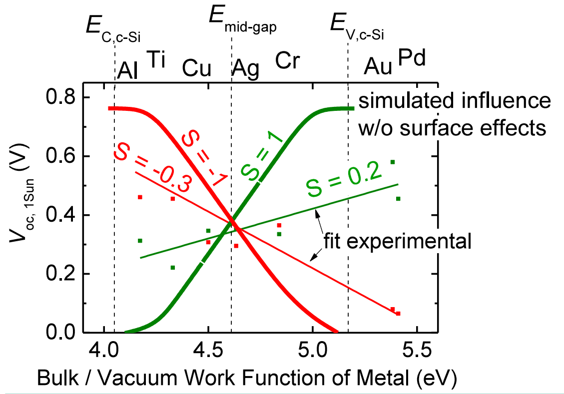
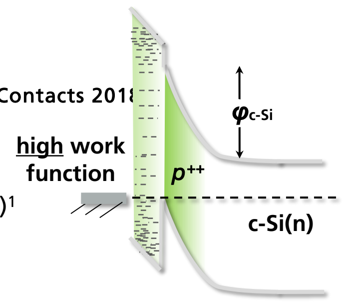
- Strong Fermi-level pinning
  - Experimental pinning factor S is <1, similar to a-Si:H Schottky diodes (S = 0.28)[1]





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Data: PhD thesis Bivour, University of Freiburg 2015
[1]Wronski *et al.*, Solid State Communications 23, 1977

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Material Screening
Metal Contact Layers

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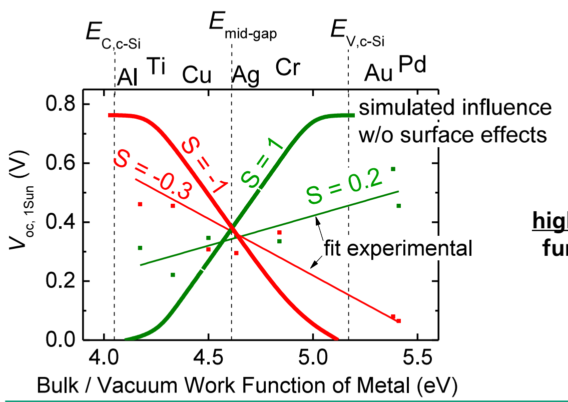

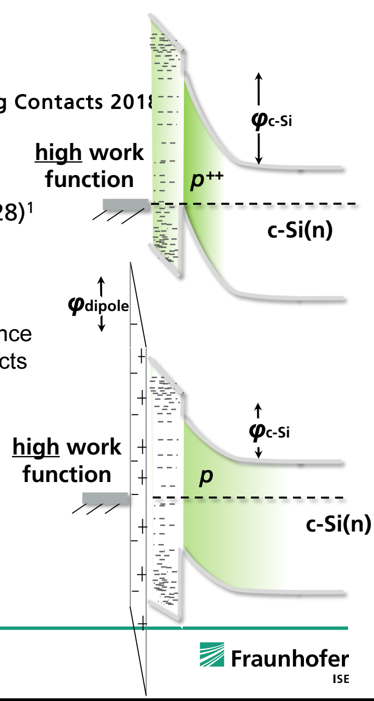

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Data: PhD thesis Bivour, University of Freiburg 2015
[1]Wronski *et al.*, Solid State Communications 23, 1977

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Material Screening
Metal Contact Layers
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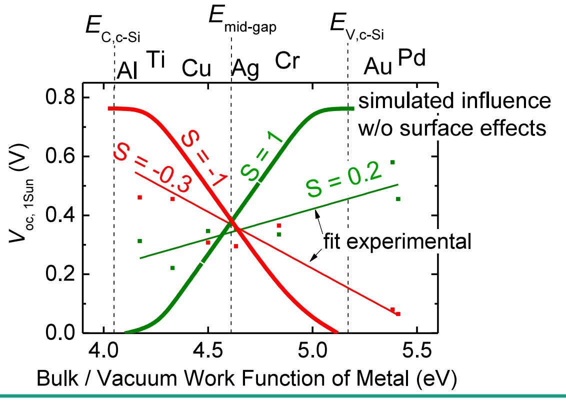

Data: PhD thesis Bivour, University of Freiburg 2015

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Material Screening
Metal Contact Layers
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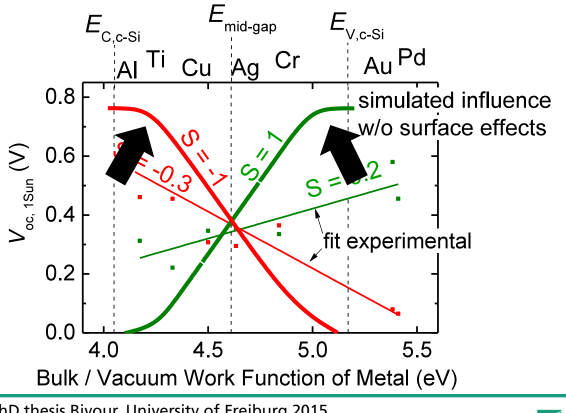
1) Reduction of Fermi-level pinning



Data: PhD thesis Bivour, University of Freiburg 2015

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Material Screening
Metal Contact Layers
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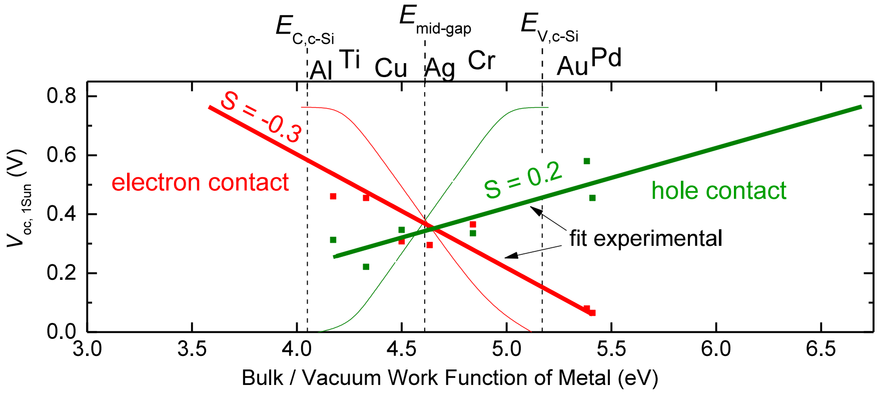
1) Reduction of Fermi-level pinning
2) Materials with much higher / lower work function



39 Data: PhD thesis Bivour, University of Freiburg 2015

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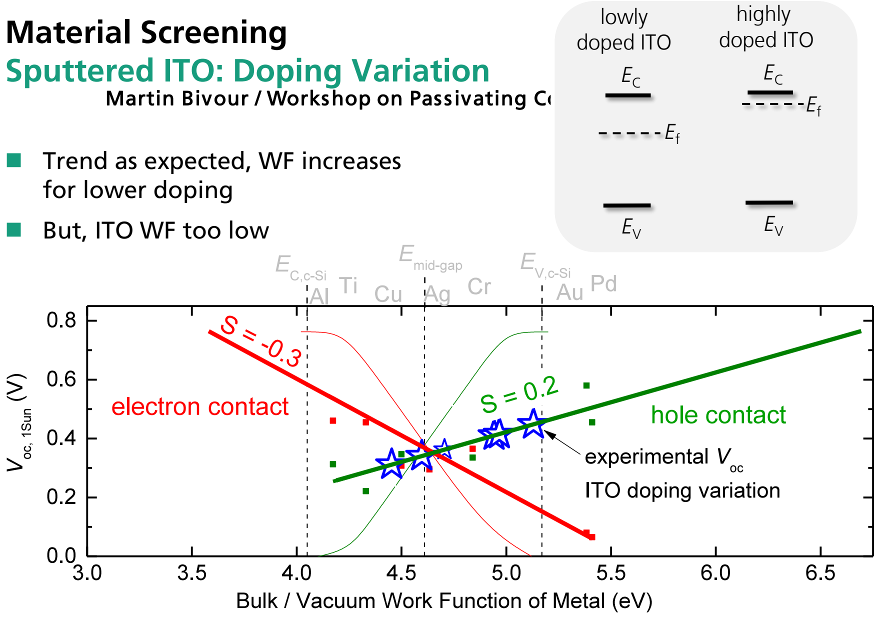
Material Screening
Sputtered ITO: Doping Variation
Martin Bivour / Workshop on Passivating C

- Trend as expected, WF increases for lower doping
- But, ITO WF too low



40 Data: PhD thesis Bivour, University of Freiburg 2015

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Material Screening

ALD TiO_x: Influence of Electrode Material

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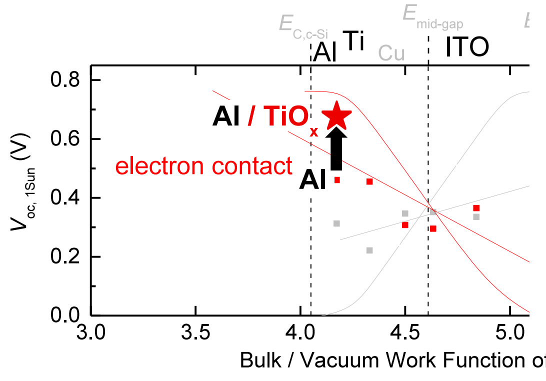
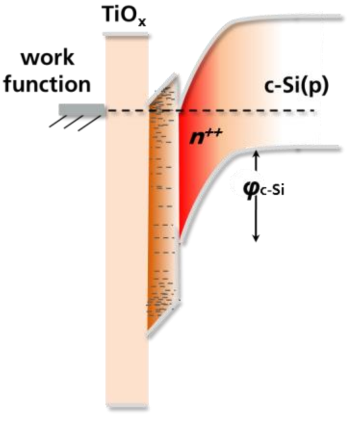
- Low WF Al contact is improved by adding TiO_x[1,2]





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Data: Matsui *et al.*, Energy Procedia 124, 2017
[1]Agrawal et al., APL, 104, 2014
[2]Yang et al., Adv Mater 28(28), 2016

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Material Screening

ALD TiO_x: Influence of Electrode Material

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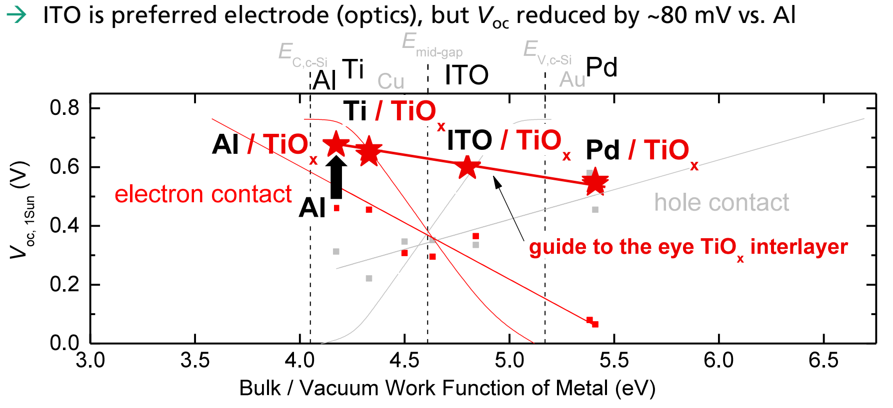
- Low WF Al contact is improved by adding TiO_x[1,2]
- But strong influence of electrode WF

→ ITO is preferred electrode (optics), but V_{oc} reduced by ~80 mV vs. Al



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Data: Matsui *et al.*, Energy Procedia 124, 2017
[1]Agrawal et al., APL, 104, 2014
[2]Yang et al., Adv Mater 28(28), 2016

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Material Screening
High Work Function Transition Metal Oxides (TMOs)
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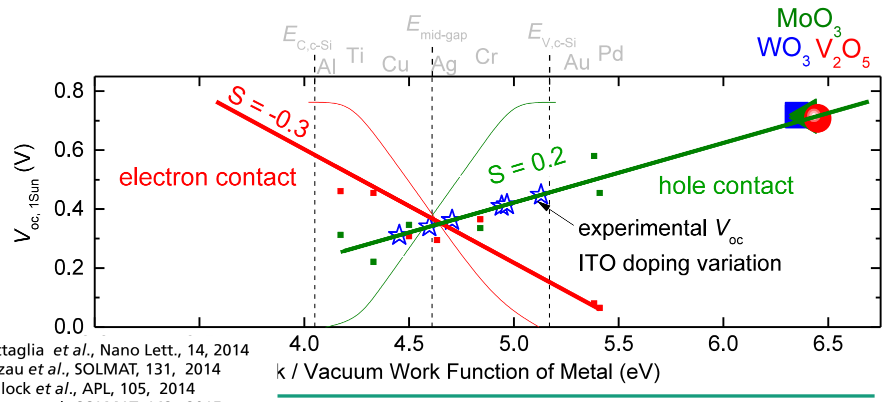
- Promising results for MoO_3, WO_3 and V_2O_5.



Battaglia *et al.*, Nano Lett., 14, 2014
Ritzau *et al.*, SOLMAT, 131, 2014
Bullock *et al.*, APL, 105, 2014
Bivour *et al.*, SOLMAT, 142, 2015
Gerling *et al.*, SOLMAT, 145, 2016
Mews *et al.*, SOLMAT, 158, 2016

Data: PhD thesis Bivour, University of Freiburg 2015

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Material Screening
Induced c-Si Junction / Effective Work Function
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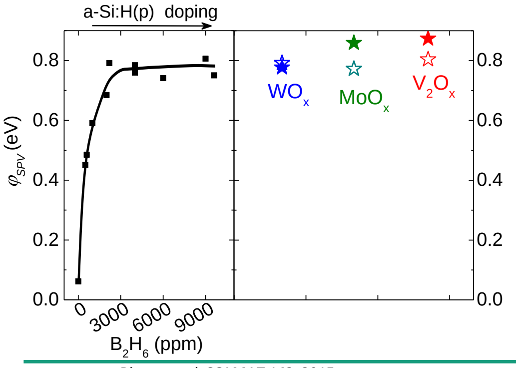
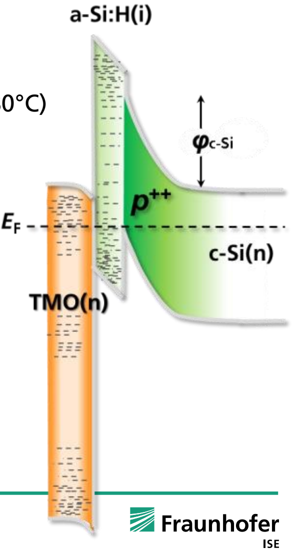
Surface photo-voltage measurements:

- $\varphi_{c\text{-}Si}$ similar for TMOs and highly doped a-Si:H(p)
- $\varphi_{c\text{-}Si}$ reduced by annealing for MoO_x and V_2O_x (180°C)





Bivour et al, SOLMAT 142, 2015

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- MoO_x based hole contacts: Simulation, fabrication and selectivity issues

Metal Oxide Based Si Heterojunction

Work Function → Sufficient High c-Si Hole Density

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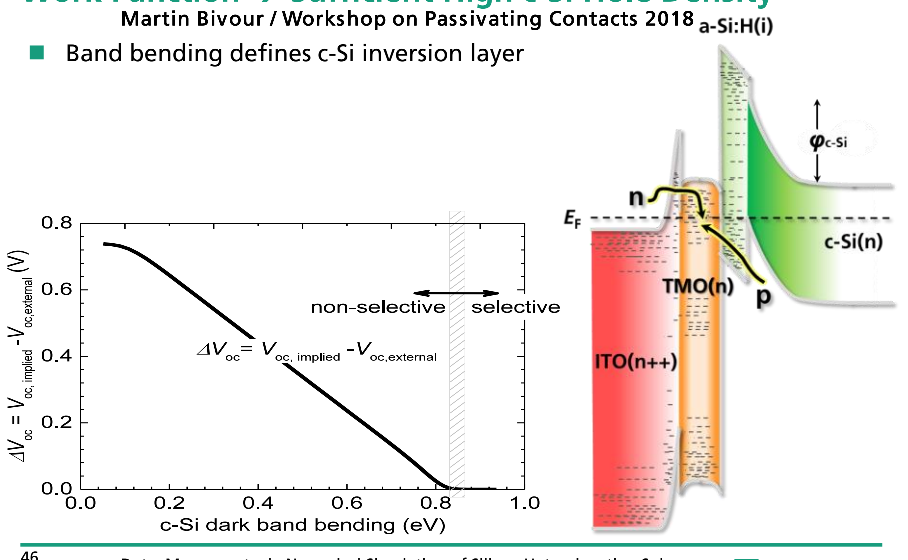
- Band bending defines c-Si inversion layer



Data: Messmer et. al., Numerical Simulation of Silicon Heterojunction Solar Cells Featuring Metal Oxides as Carrier-Selective Contacts, Accepted IEEE-JPV, 2017

Metal Oxide Based Si Heterojunction

Extraction of Holes from the c-Si Inversion Layer

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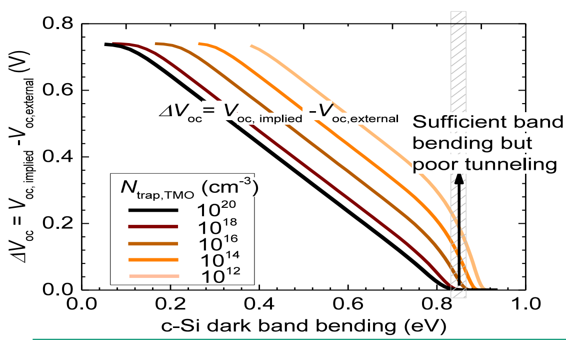
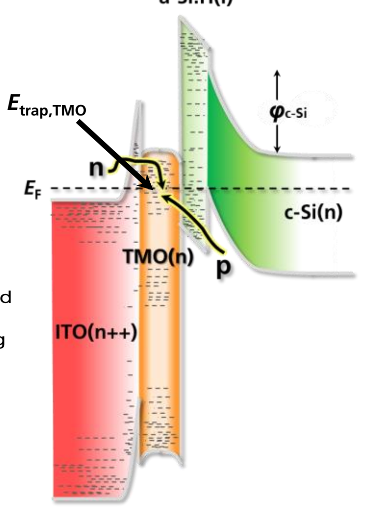
- Band bending defines c-Si inversion layer
- Inefficient tunneling for
  - Too low metal oxide trap density





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Data: Messmer et. al., Numerical Simulation of Silicon Heterojunction Solar Cells Featuring Metal Oxides as Carrier-Selective Contacts, Accepted IEEE-JPV, 2017

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Metal Oxide Based Si Heterojunction

Extraction of Holes from the c-Si Inversion Layer

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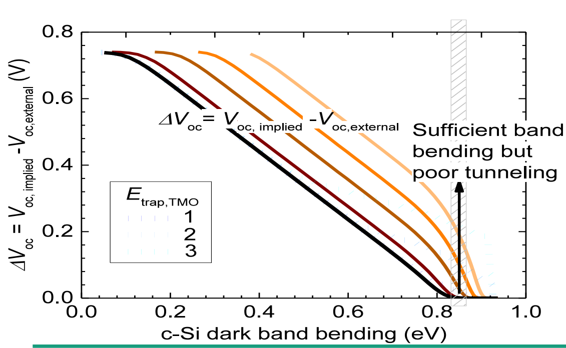
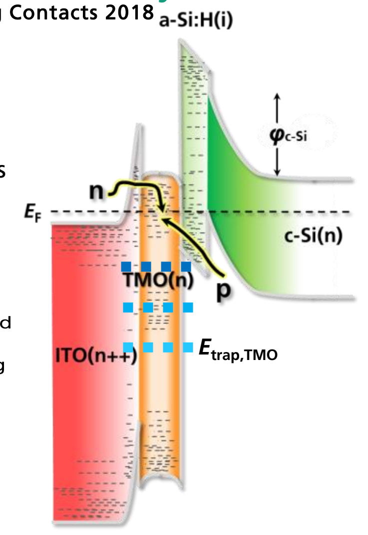
- Band bending defines c-Si inversion layer
- Inefficient tunneling for
  - Too low metal oxide trap density
  - Unsuited energetic distribution of traps





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Data: Messmer et. al., Numerical Simulation of Silicon Heterojunction Solar Cells Featuring Metal Oxides as Carrier-Selective Contacts, Accepted IEEE-JPV, 2017

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Metal Oxide Based Si Heterojunction

Extraction of Holes from the c-Si Inversion Layer

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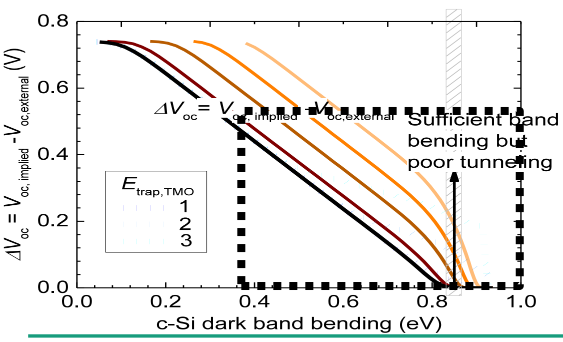

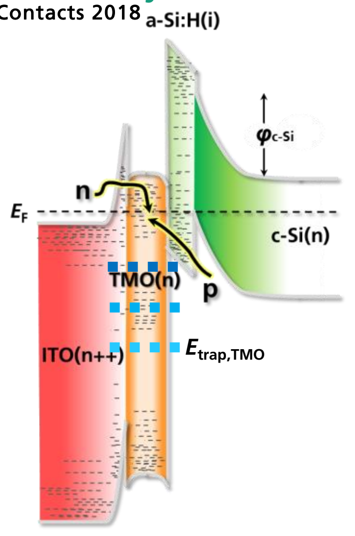

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Data: Messmer et. al., Numerical Simulation of Silicon Heterojunction Solar Cells Featuring Metal Oxides as Carrier-Selective Contacts, Accepted IEEE-JPV, 2017

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Engineering the Metal Oxide Properties

Evaporation, Sputtering and ALD of MoO_x

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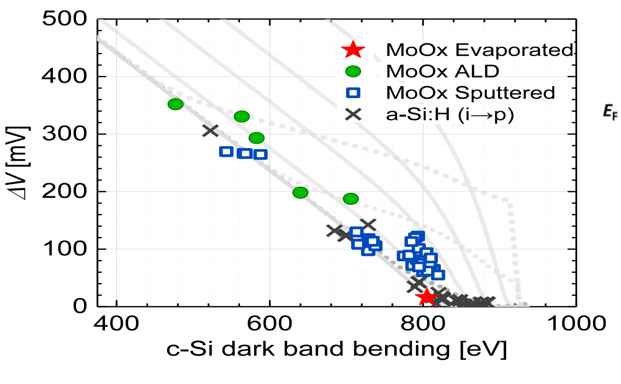

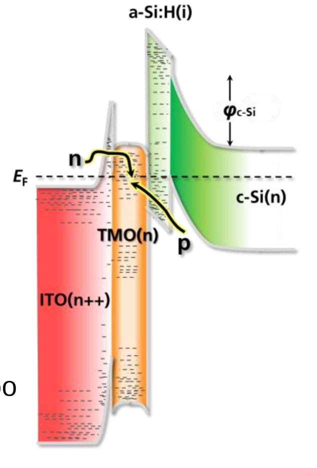

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Data:
Bivour et al., Energy Procedia, 92, 2016
Bivour et al., Energy Procedia, 124, 2017
Bivour et al., Proceedings 33th EU PVSEC, 2017

ALD films deposited at TU/e: Bart Macco

TU/e

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Engineering the Metal Oxide Properties
Evaporation, Sputtering and ALD of MoO_x
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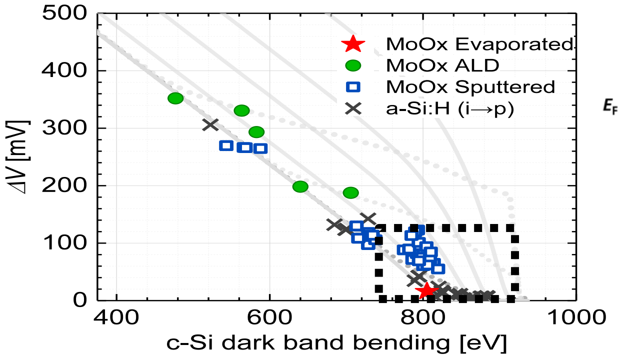

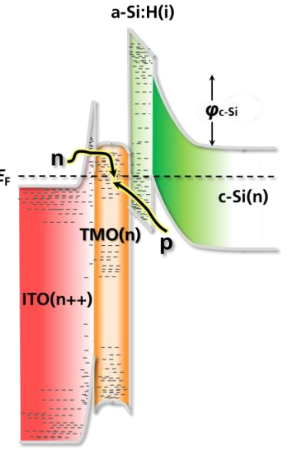

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Data:
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Bivour et al., Energy Procedia, 124, 2017
Bivour et al., Proceedings 33[th] EU PVSEC, 2017

ALD films deposited at TU/e: Bart Macco

TU/e

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Evaporated MoO_x
Loss of Selectivity with Annealing
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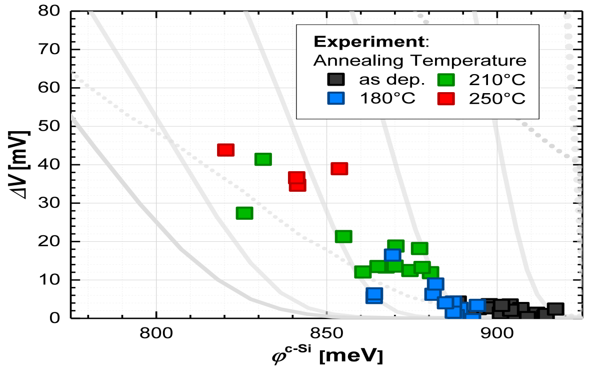

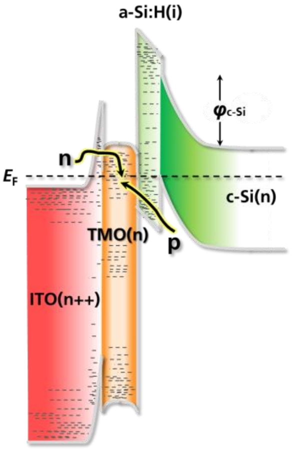

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Data:
Neusel et al., Energy Procedia, 124, 2017
Bivour et al., Proceedings 33[th] EU PVSEC, 2017

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FF Degradation with Annealing
MoO_x Thickness and a-Si:H(i) Buffer
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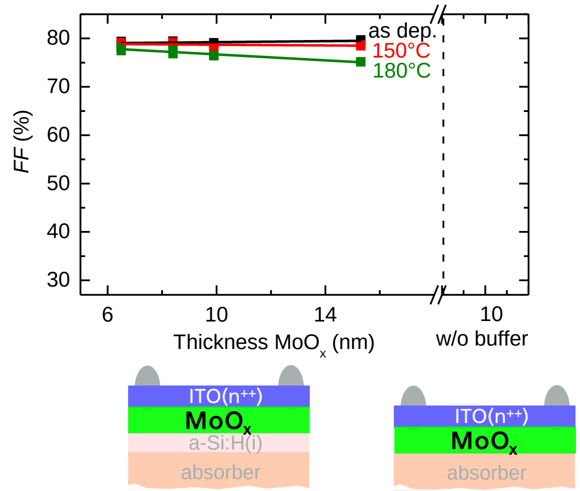
- Degradation less pronounced for:
  - Thinnest MoO_x



Neusel et al., Energy Procedia, 124, 2017
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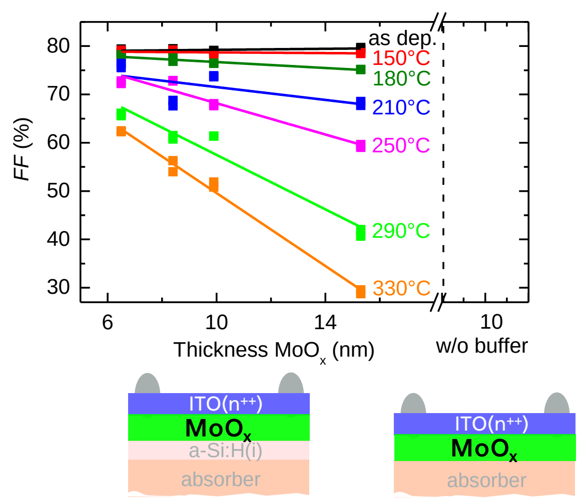

Neusel et al., Energy Procedia, 124, 2017
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FF Degradation with Annealing
MoO_x Thickness and a-Si:H(i) Buffer
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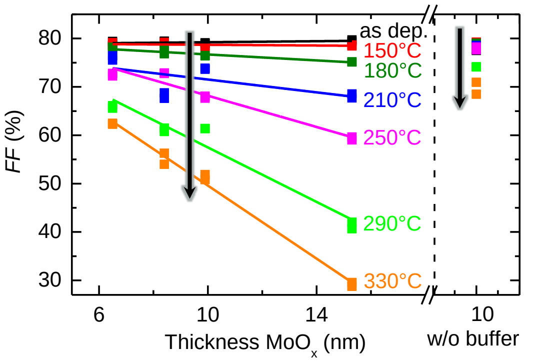
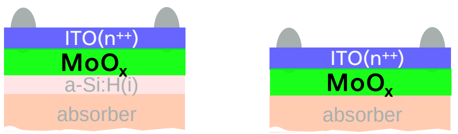
- Degradation less pronounced for:
  - Thinnest MoO_x
  - w/o a-Si:H(i) buffer





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Neusel et al., Energy Procedia, 124, 2017

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Status Quo: Textured Front Side MoO_x vs. Homojunction
ITO / MoO_x / a-Si:H(i)
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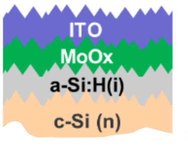
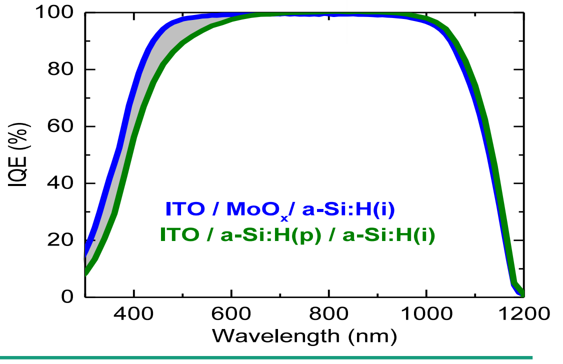
- Promising efficiency
  - Annealing 140°C
  - So far, not compatible with 200°C back-end

J_{sc} (mA/cm²)	V_{oc} (mV)	*FF* (%)	η (%)
40.3	699	80.2	22.6





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Status Quo: Textured Front Side MoO_x vs. Homojunction

ITO / MoO_x / a-Si:H(i)

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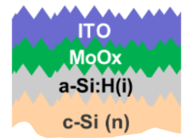
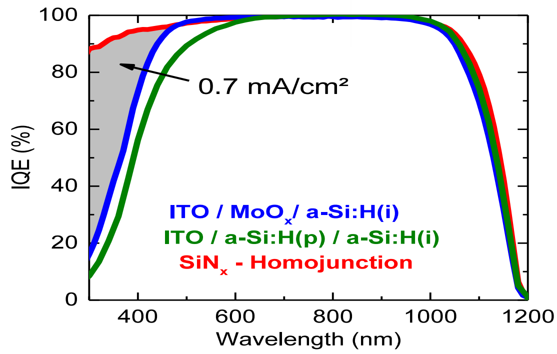


- Promising efficiency
  - Annealing 140°C
  - So far, not compatible with 200°C back-end
- Good blue response
  - Gap to homojunction reduced to only 0.7 mA/cm²
  - Now absorption in ITO significant

J_{sc} (mA/cm²)	V_{oc} (mV)	*FF* (%)	η (%)
40.3	699	80.2	22.6



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Summary

Challenges:

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- Integration of "novel" thin films in silicon device architectures
  - Thermal and chemical stability for subsequent processing and interaction with adjacent films (oxidation/reduction, hydrogen)
  - Long term stability?
- More c-Si solar cell specific understanding for material engineering
  - Intentional doping / alloying to tailor properties, ….

Good news:

- "New" materials currently explored for c-Si solar cells by many groups
  - Basic applicability of metal oxides for selective contacts shown
  - Higher transparency and high *FF*
- Work function engineering for proper induced c-Si junction
- More heterojunction specify properties for efficient transport (band line-up, tunneling, …)

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Especially Bachelor / Master Students
L. Neusel, F. Zähringer, C. Messmer, J. Temmler, R. Sharma, P. Hettich

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aufgrund eines Beschlusses des Deutschen Bundestages

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Thank You Very Much for Your Attention!

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