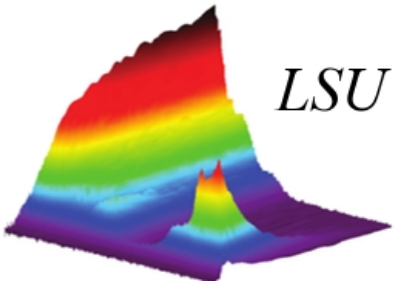
Advances and challenges in making an artificial leaf

Jacinto Sá



ÉCOLE POLYTECHNIQUE
FÉDÉRALE DE LAUSANNE

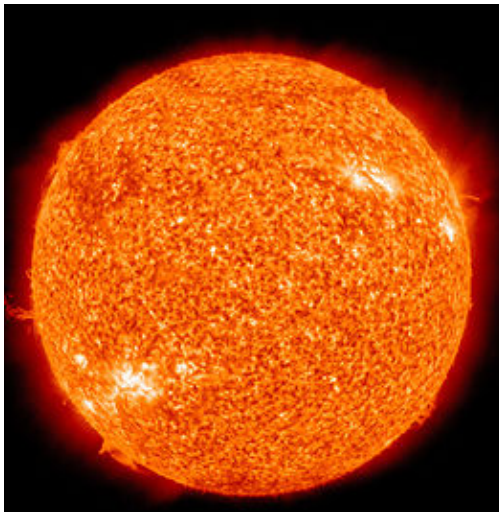
PAUL SCHERRER INSTITUT
PSI

Artificial photosynthesis

'The problem of future begins to interest us [...] Is fossil solar energy the only one that may be used in modern life and civilization? [...] ***for nature is not in a hurry and mankind is***'

Ciamician, Science 36 (1912) 385

Practical artificial leaf

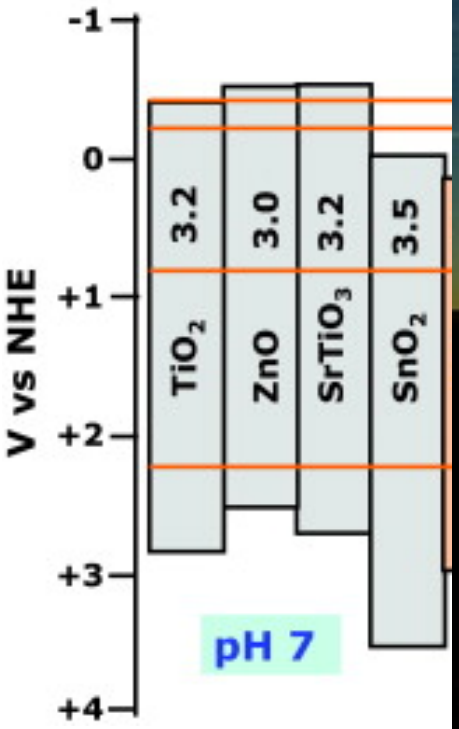


O_2

H_2

- ✧Visible light absorber
- ✧Reduction catalyst (Hydrogen production)
- ✧Oxidation catalyst (Oxygen-evolving catalyst)

Semico ysis

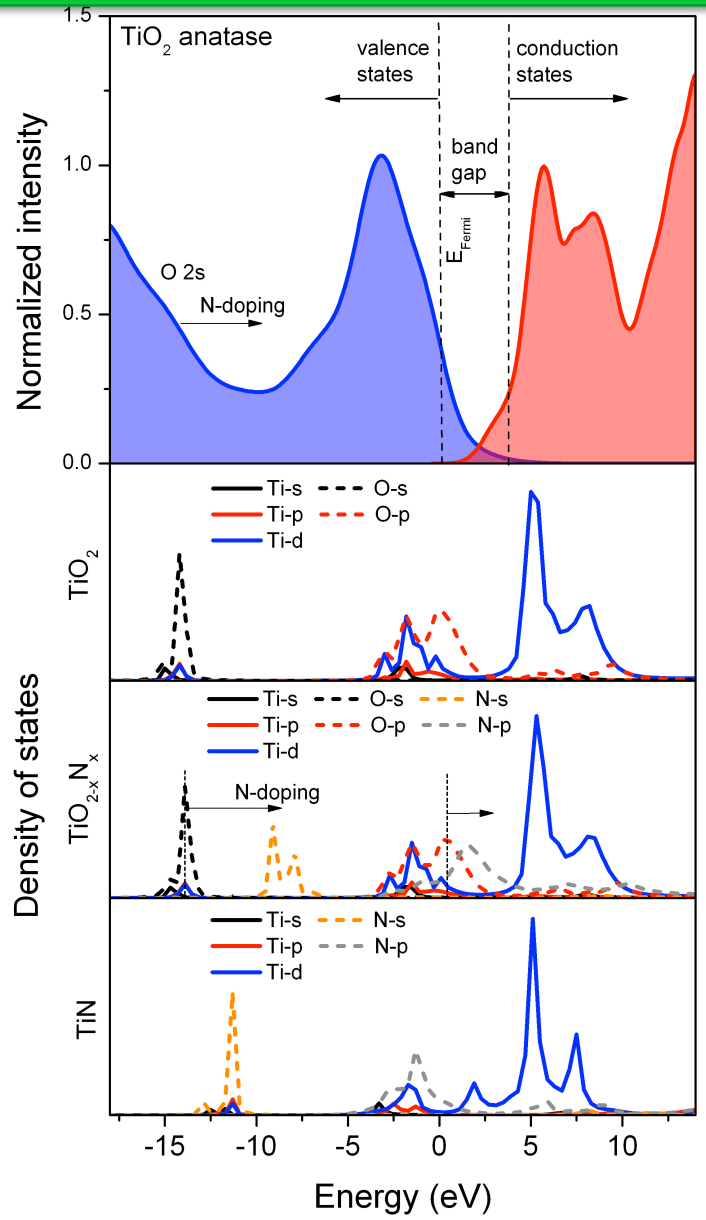


Lianos – J. Hazardou

res

light

Band gap narrowing



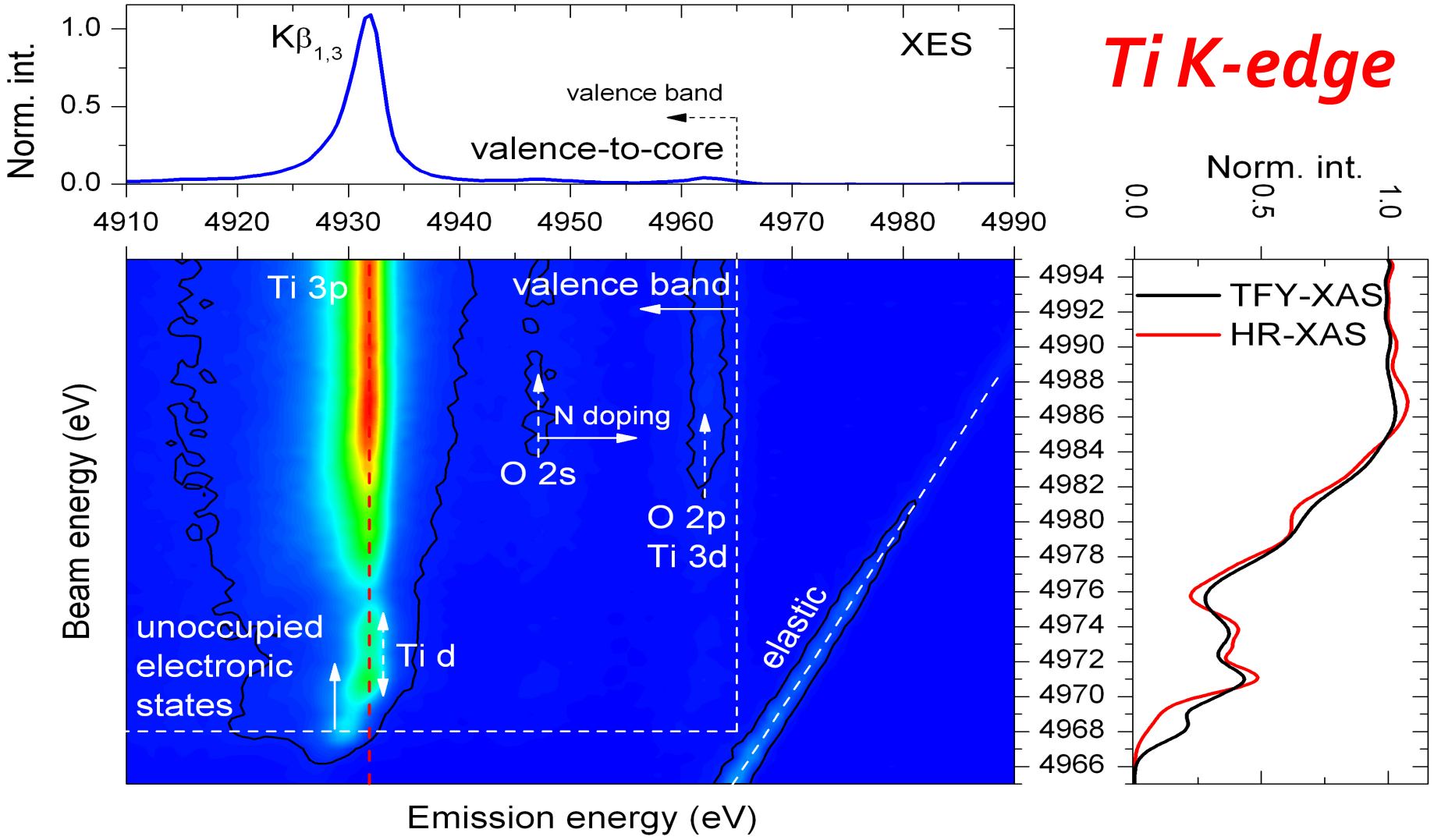
Ti K-edge

Szlachetko & Sá, CrystEngComm *15 (2013) 2583*

Narrowing the band gap

Decreases the number of reactions that can be catalysed

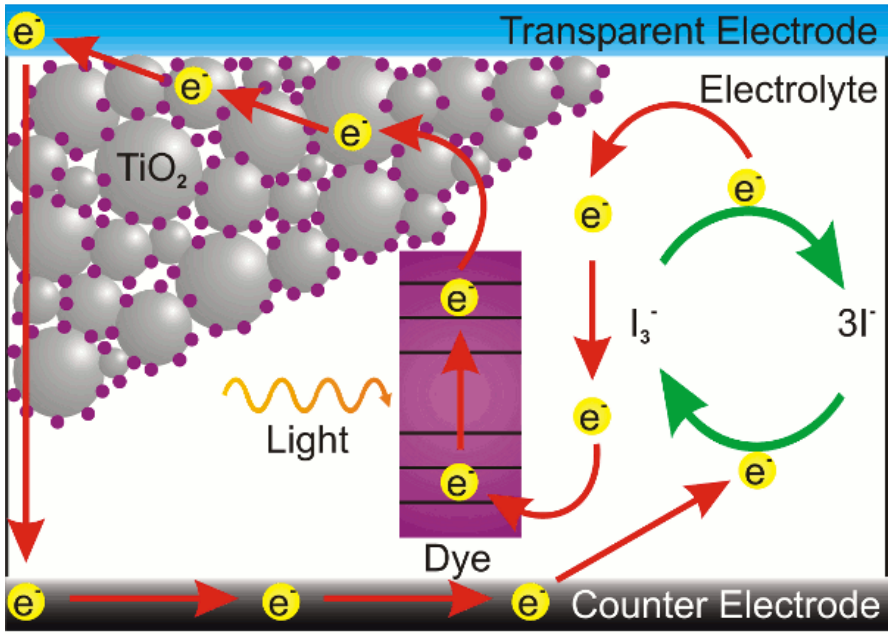
RIXS measurements



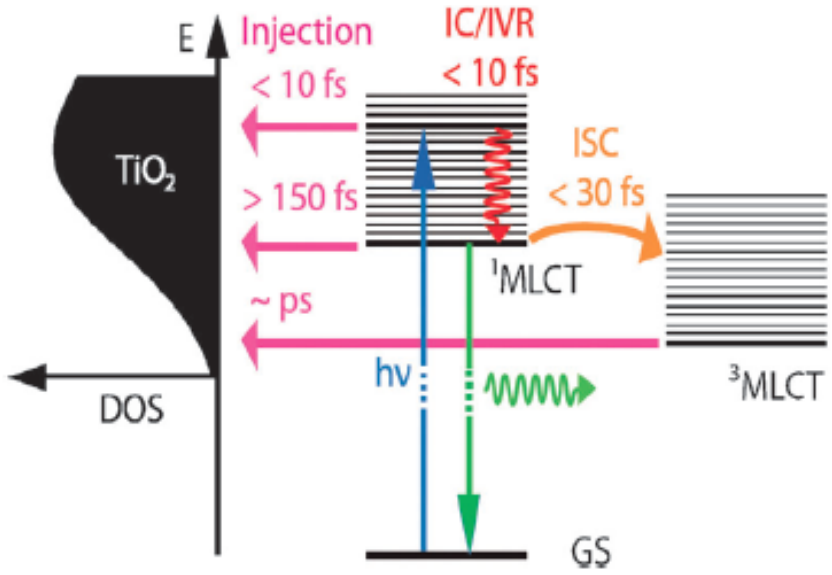
Ti K-edge

Szlachetko & Sá, CrystEngComm *15 (2013) 2583*

Solar light absorbers



http://tikalon.com/blog/blog.php?article=graetzel

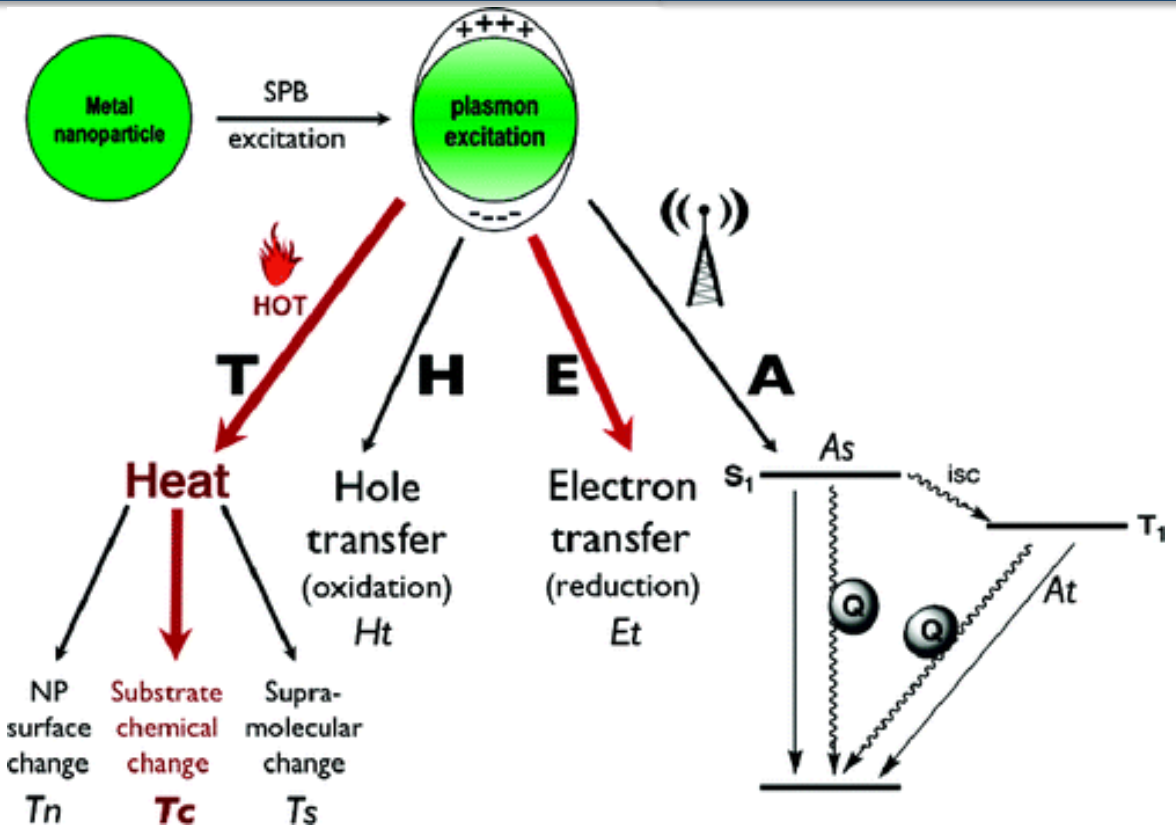


Bräm et al. - PCCP 14 (2012) 7934

- ✧ Requires high coverage (no space for catalysis)
- ✧ High level of expertise to modify and tune absorption
- ✧ Low stability
- ✧ Unreactive hole

d^{10} metal plasmonic structures

- ✧ High cross-sections
- ✧ Easy to tune and modify
- ✧ Reactive hole
- ✧ High stability



Hallett-Tapley et al. - J. Phys. Chem. C 115 (2011) 10784

Electronic mechanism

NO spectroscopic evidence

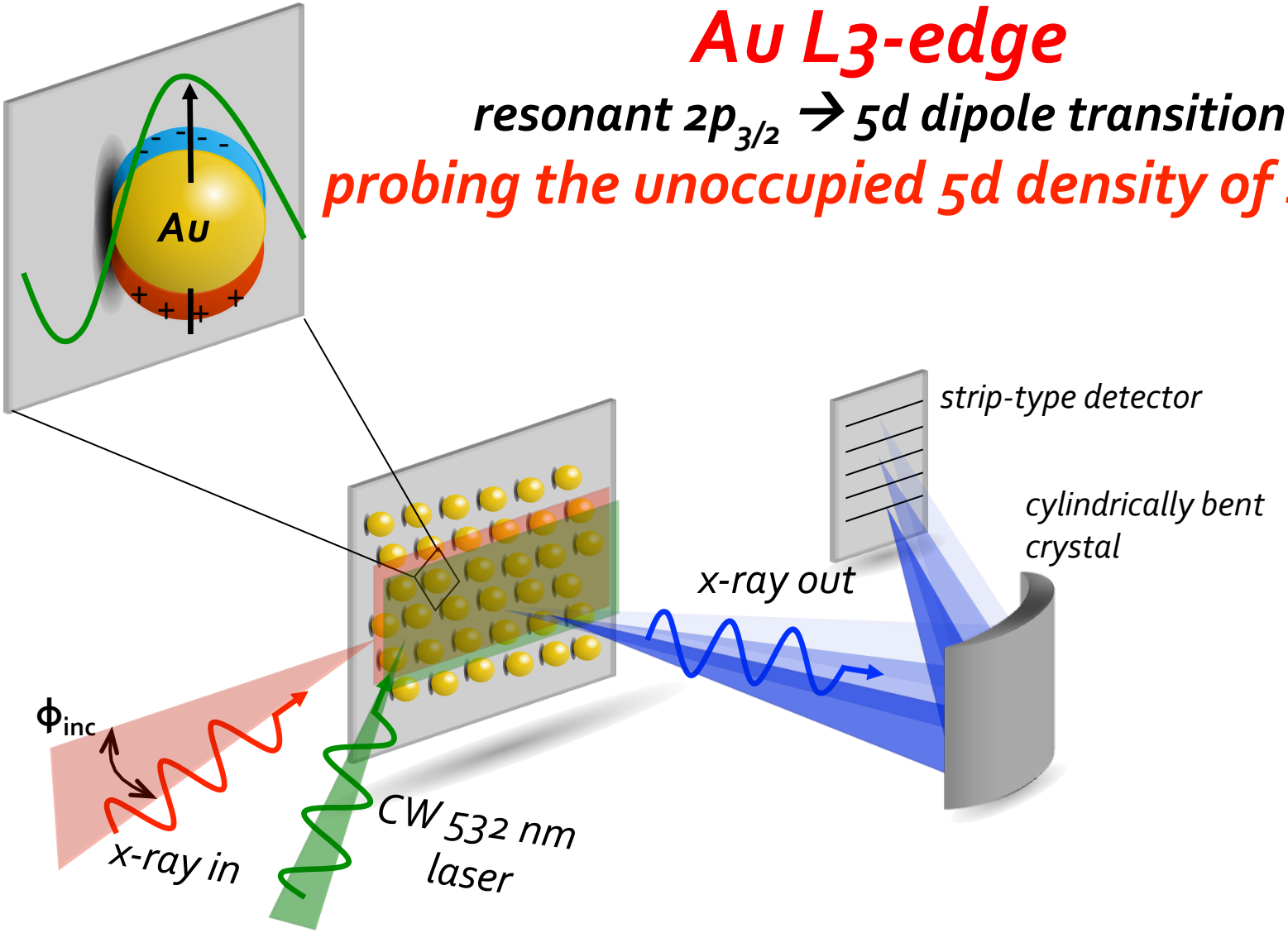
Mubeen et al. - Nature Nanotechnology 8 (2013) 247

Electronic mechanism

Au L3-edge

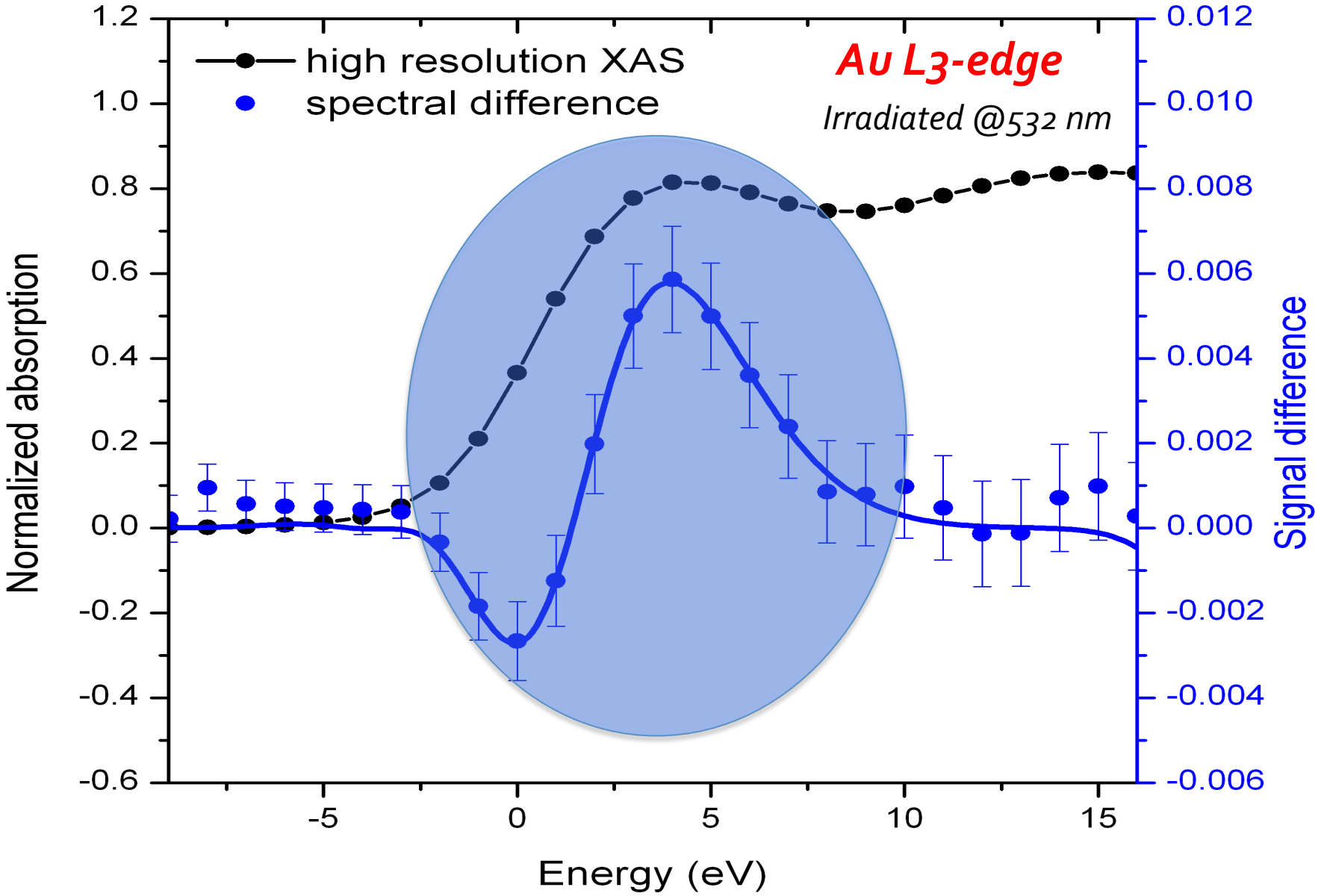
resonant $2p_{3/2} \rightarrow 5d$ dipole transition

probing the unoccupied 5d density of states

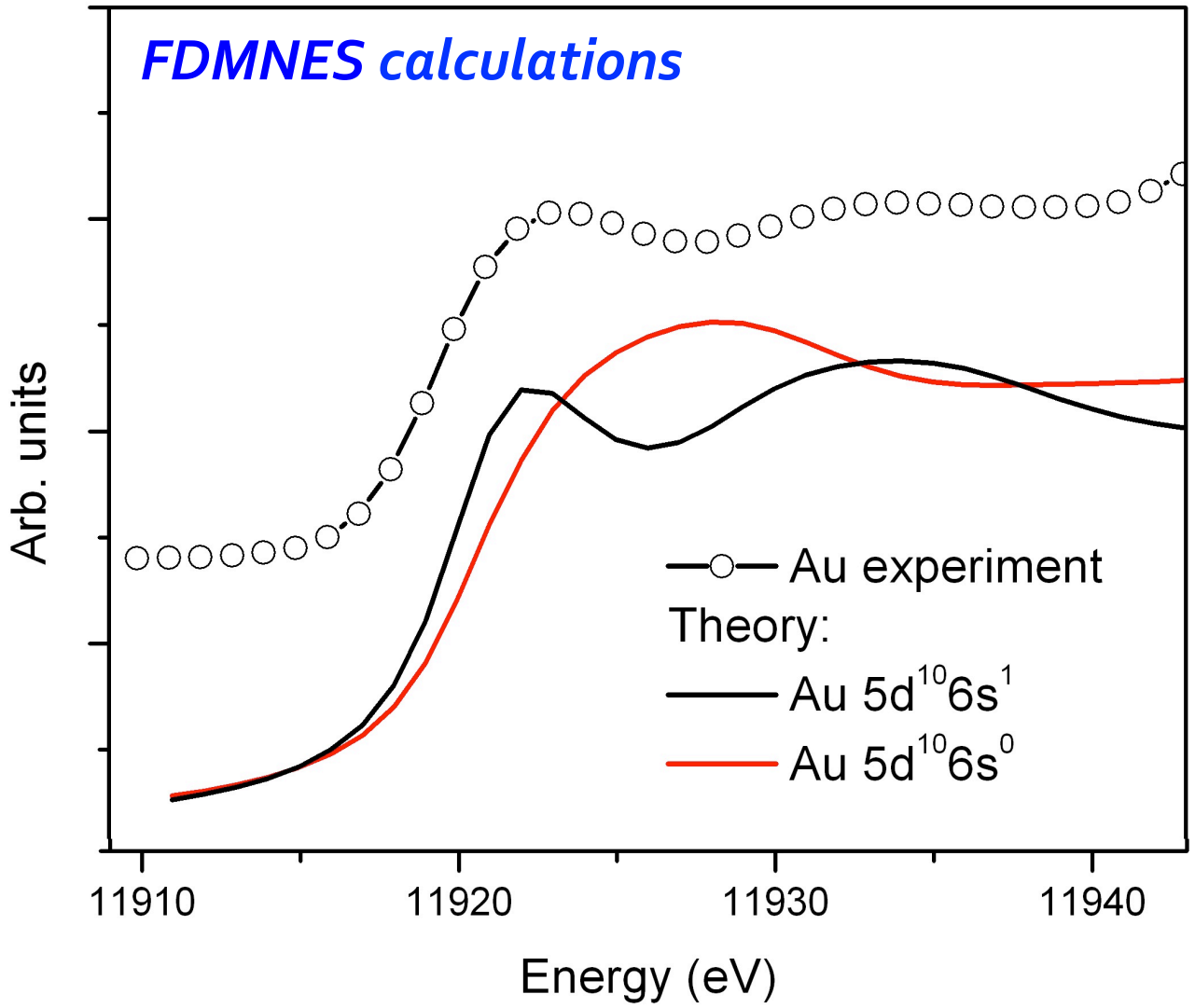


Szlachetko et al. – Chem. Commun. 48 (2012) 10898

Electronic mechanism (XAS)



Electronic mechanism (XAS)



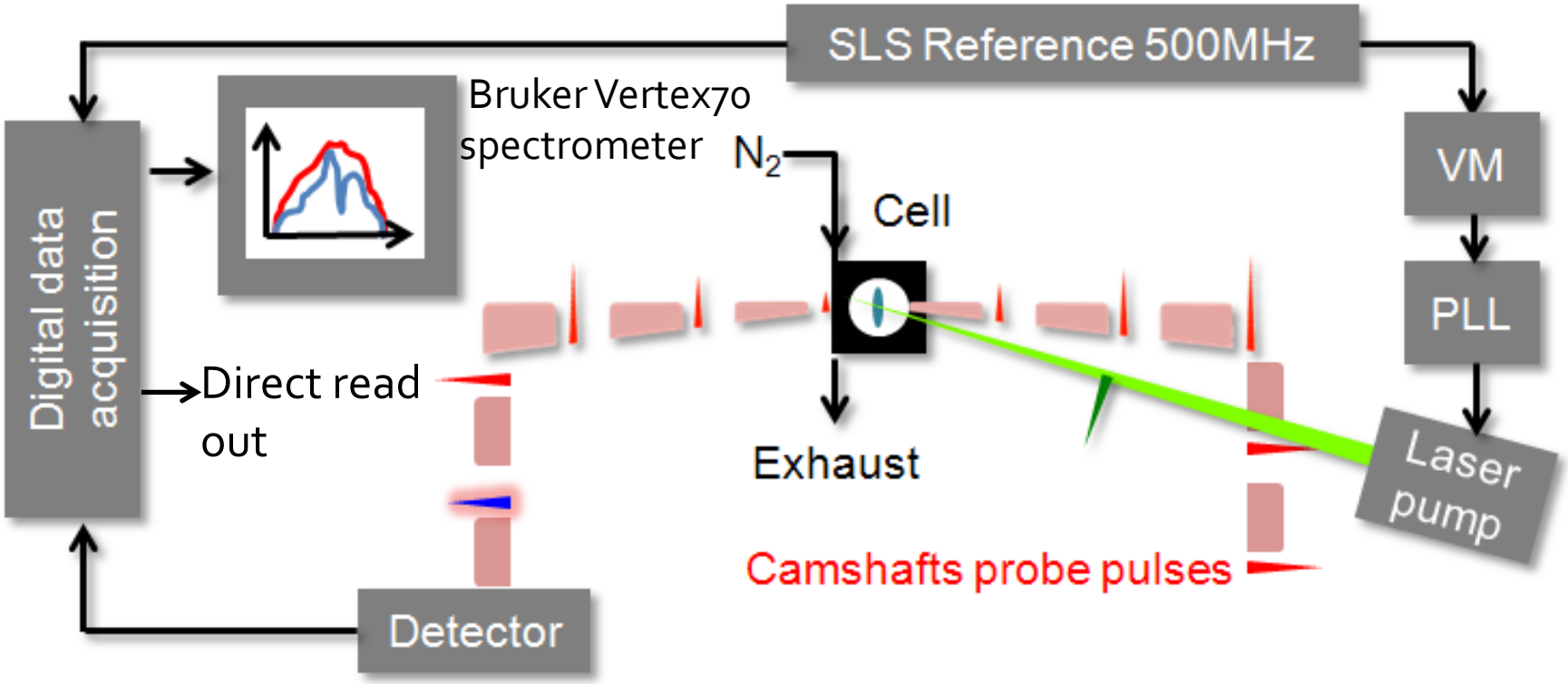
Increase DOS and energy shift

Formation of holes and hot electrons

Sa et al. – (in preparation)

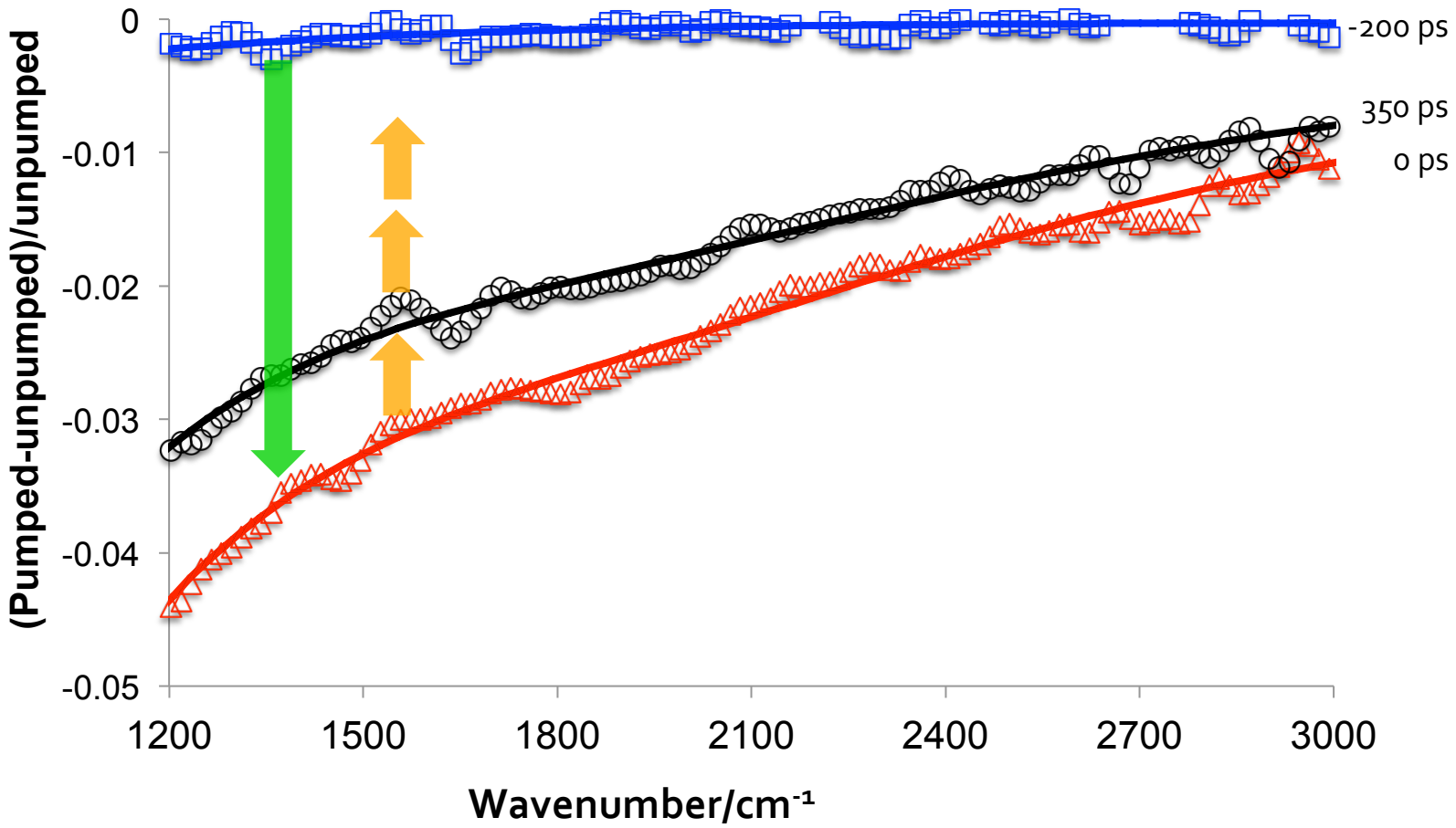
Electron transfer

Hot electrons have enough energy to overcome Schottky barrier?



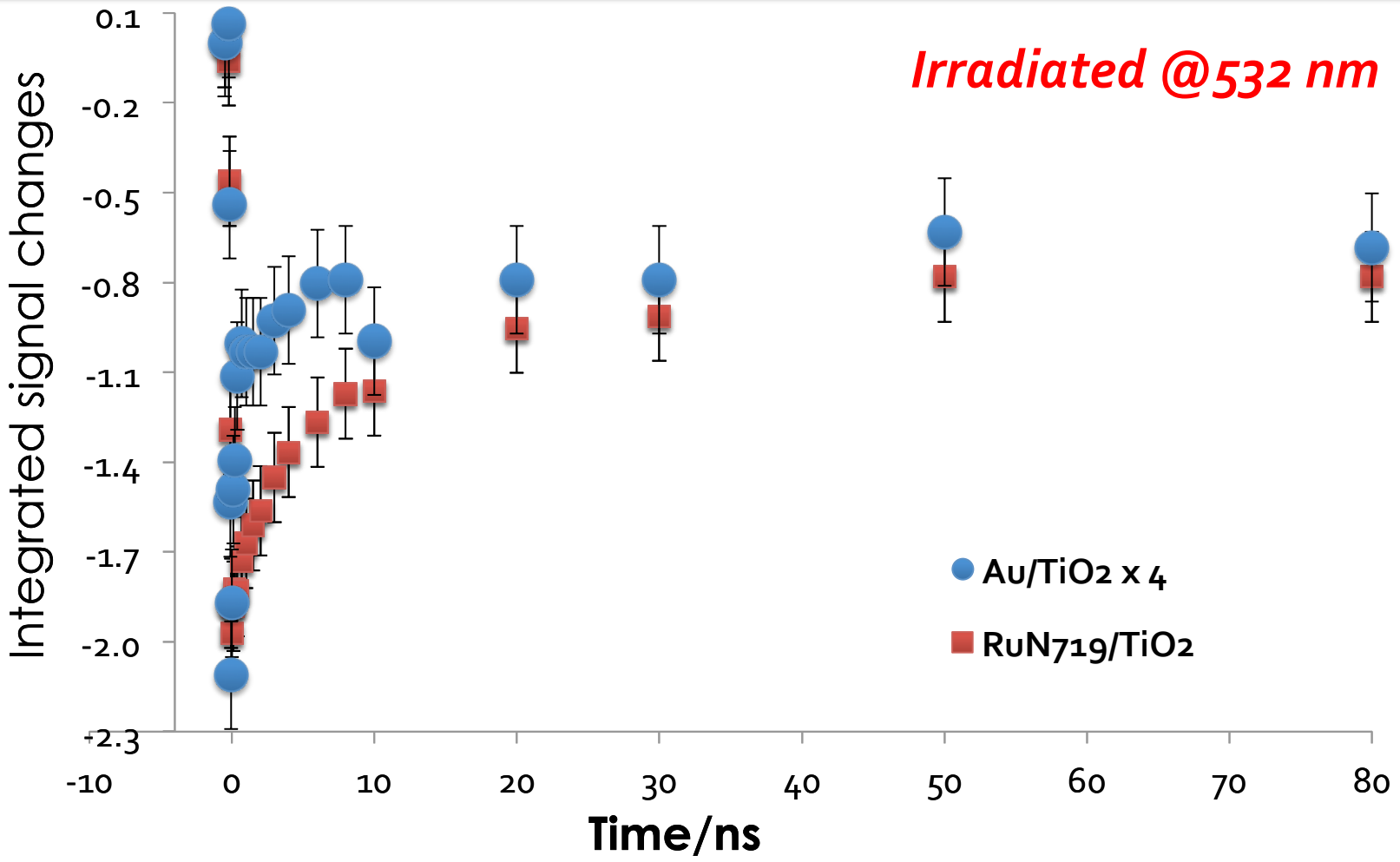
Sá et al. - Analyst 138 (2013) 1966

Electron transfer



Sá et al. - *in preparation*

Electron transfer



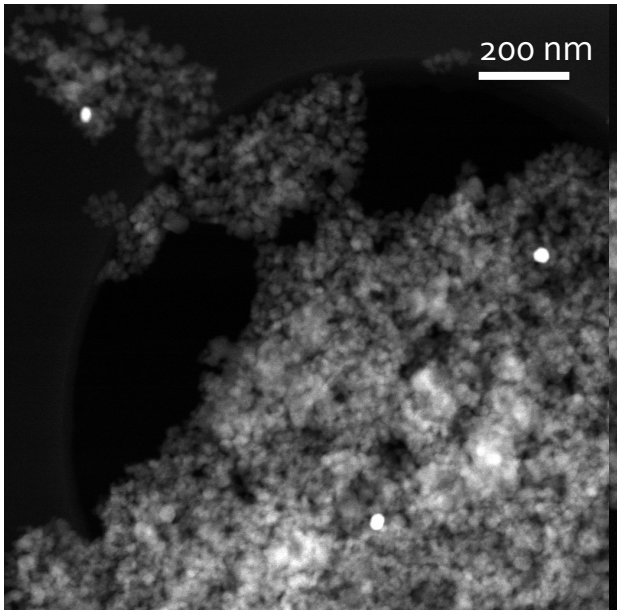
τ_{dye}fast = 1.9 ± 0.42 ns

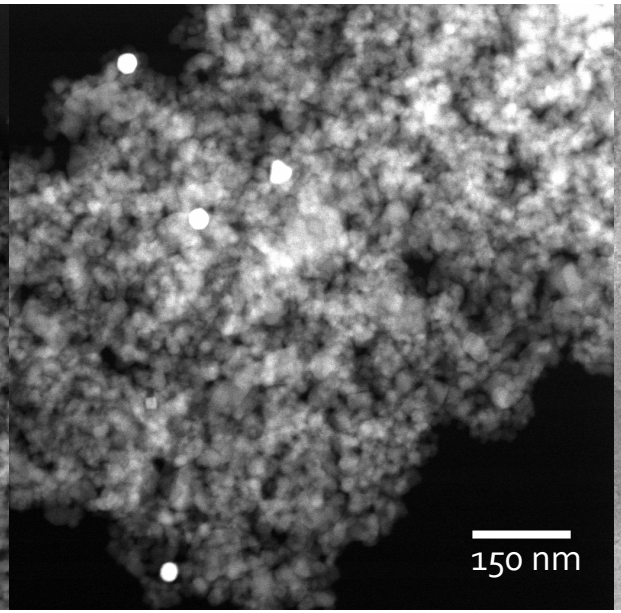
τ_{dye}slow =16.0 ± 3.1 ns

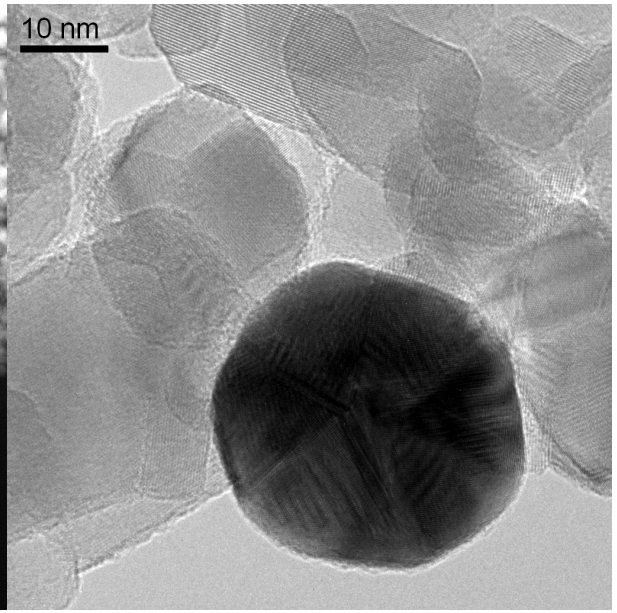
τ_{Au}fast = 0.21 ± 0.03 ns

τ_{Au}slow = 18.1 ± 10.3 ns

Summary







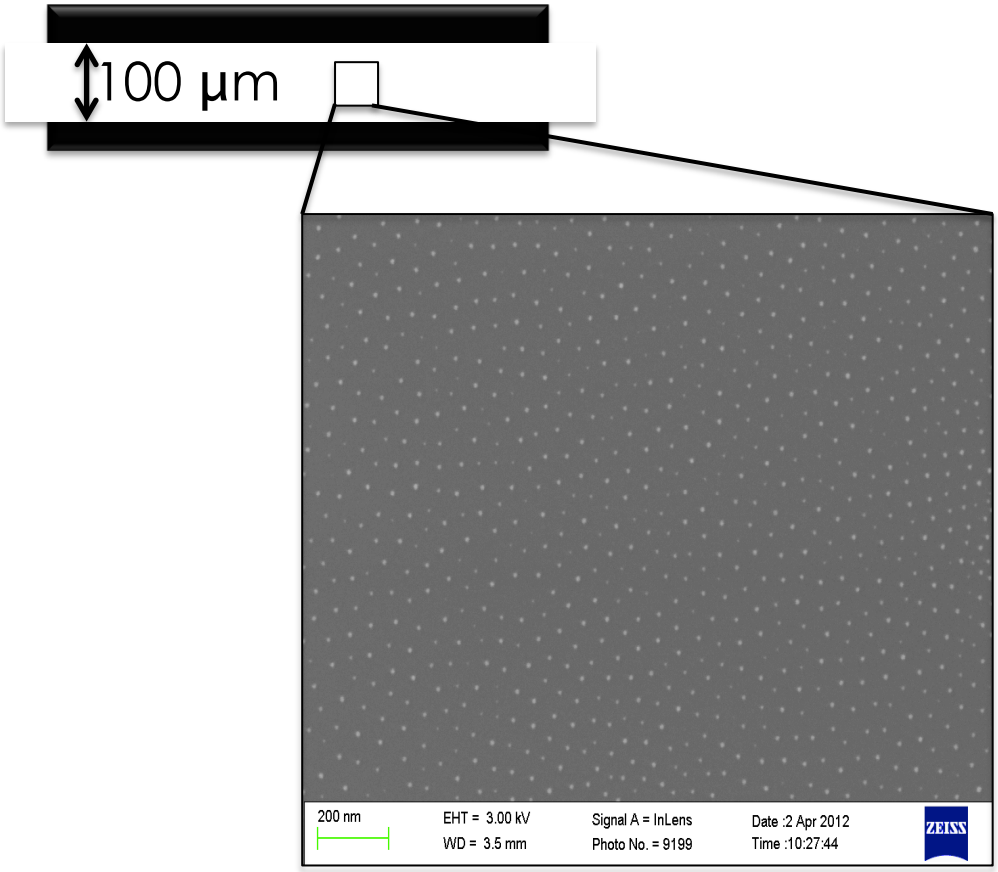
- ✧ Plasmons can work as visible light absorbers
- ✧ Low surface coverage

Thermal??

Ethanol steam reforming

$H_2O + CH_3CH_2OH$

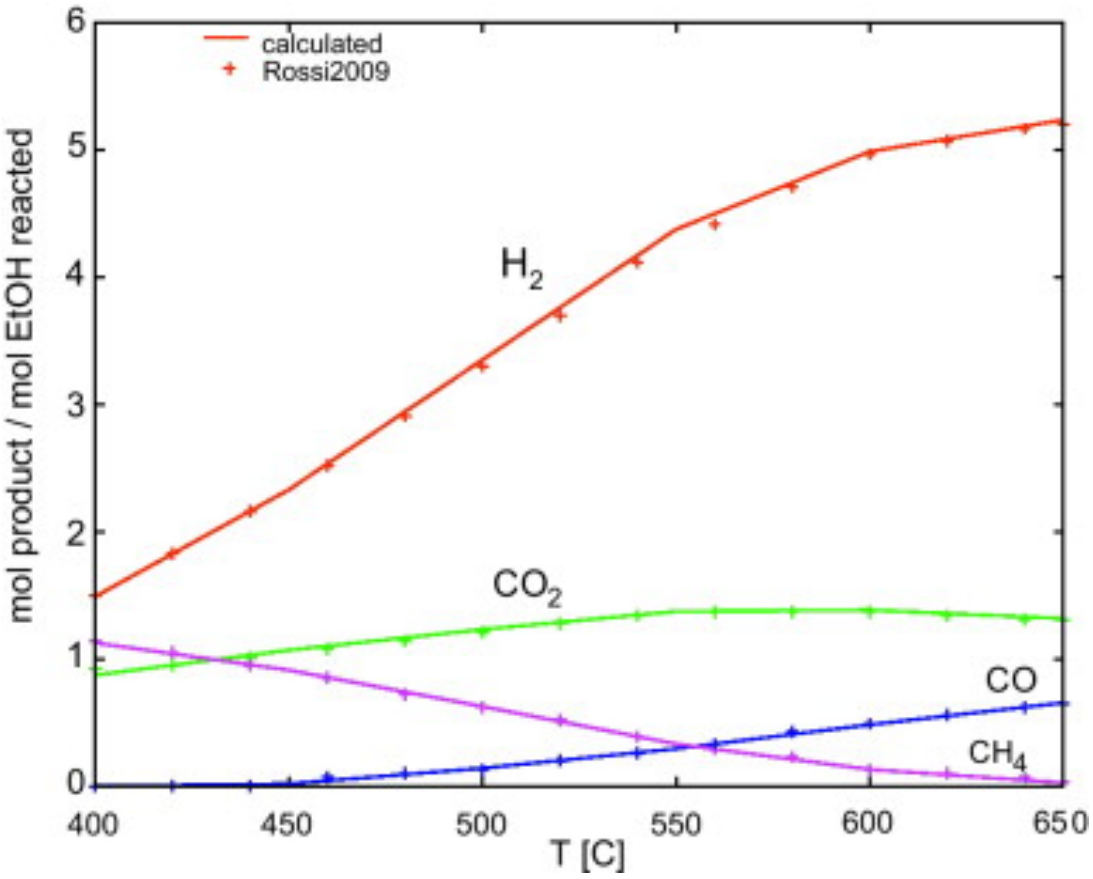
Irradiated @532 nm



Steam reforming ethanol

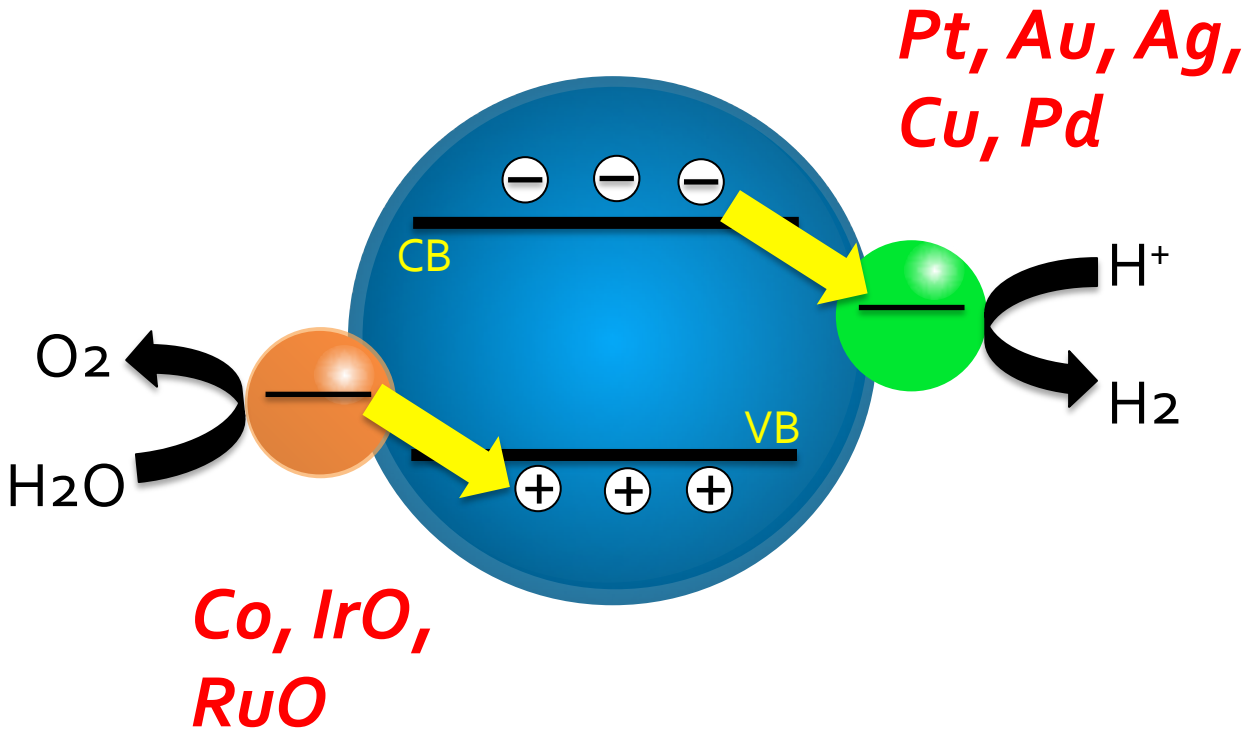
MS analysis:

CO, CO2, H2, CH4

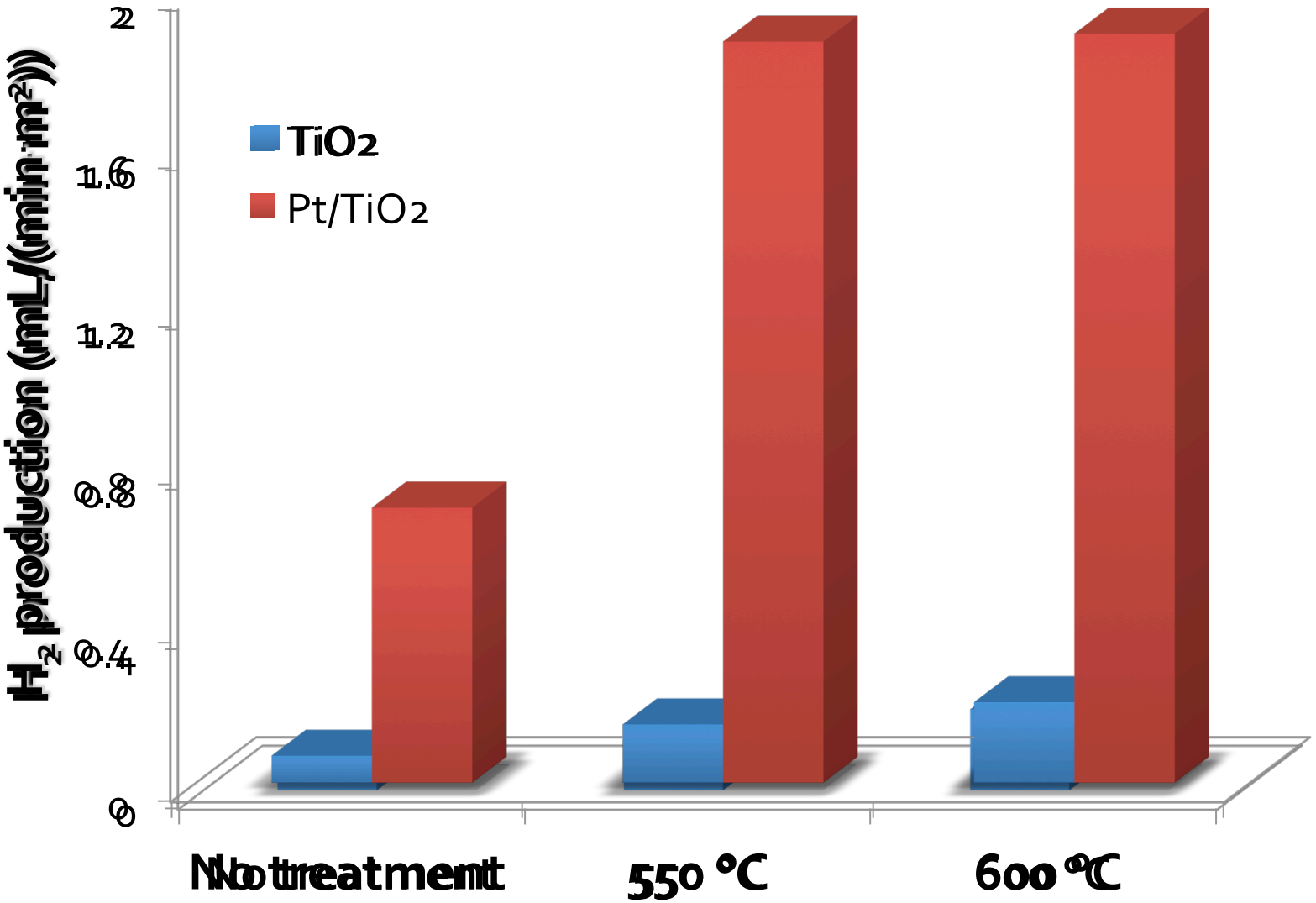


Alvarado & Garcia – Chem. Eng. J. 165 (2010) 649

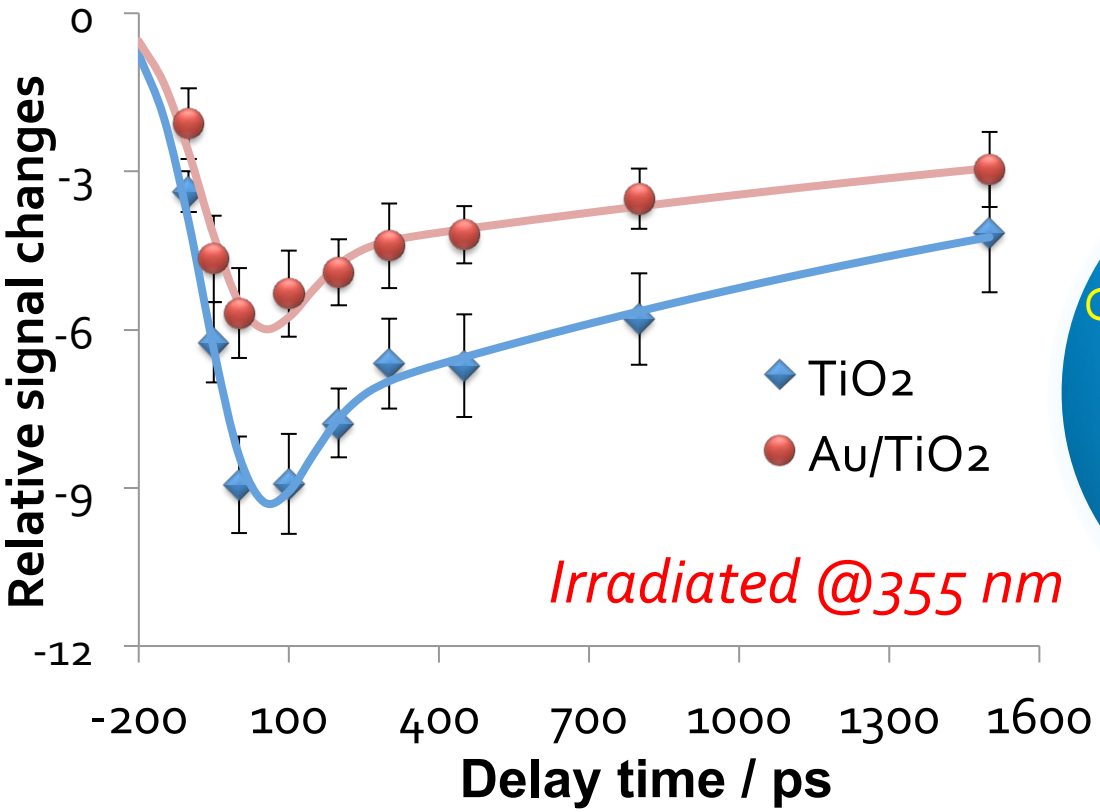
Co-catalysts

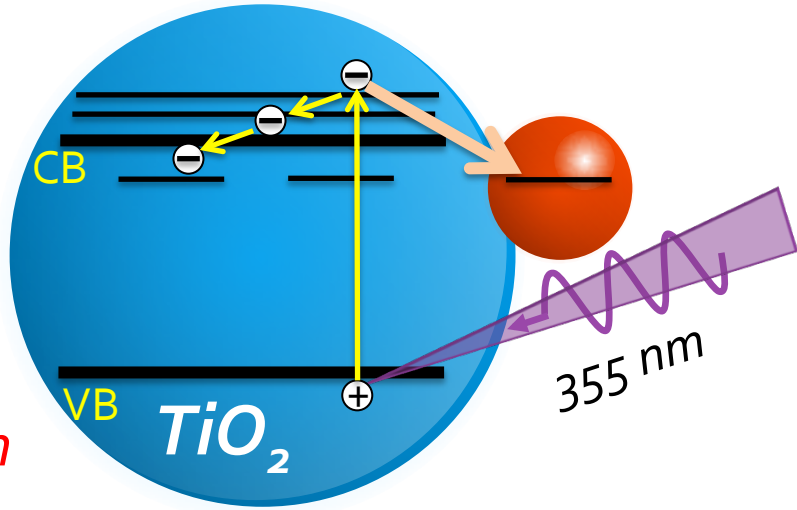


Co-catalyst for H_2 production



Co-catalyst effect





✧ 30-40% less electrons detected

✧ Occurs with Au, Ag, Pt,…

Sá et al. - Analyst 138 (2013) 1966

Oxygen evolution

Wireless Solar Water Splitting Using Silicon-Based Semiconductors and Earth-Abundant Catalysts

Reece et al. Science 334 (2011) 645

Future Plans

- ✧ Development of broad band plasmonic structures
- ✧ Reduce metal content
- ✧ Synthesize composite materials for photocatalysis and thermally driven processes
- ✧ Fuel production from alternative sources

'I want to see the world evolve into a better place and with the help of science, I am trying to achieve that aim'

Prof. Robert S. Lang

Acknowledgements

FONDS NATIONAL SUISSE
SCHWEIZERISCHER NATIONALFONDS
FONDO NAZIONALE SVIZZERO
SWISS NATIONAL SCIENCE FOUNDATION

Engineering and Physical Sciences
Research Council

Thank you for your kind attention