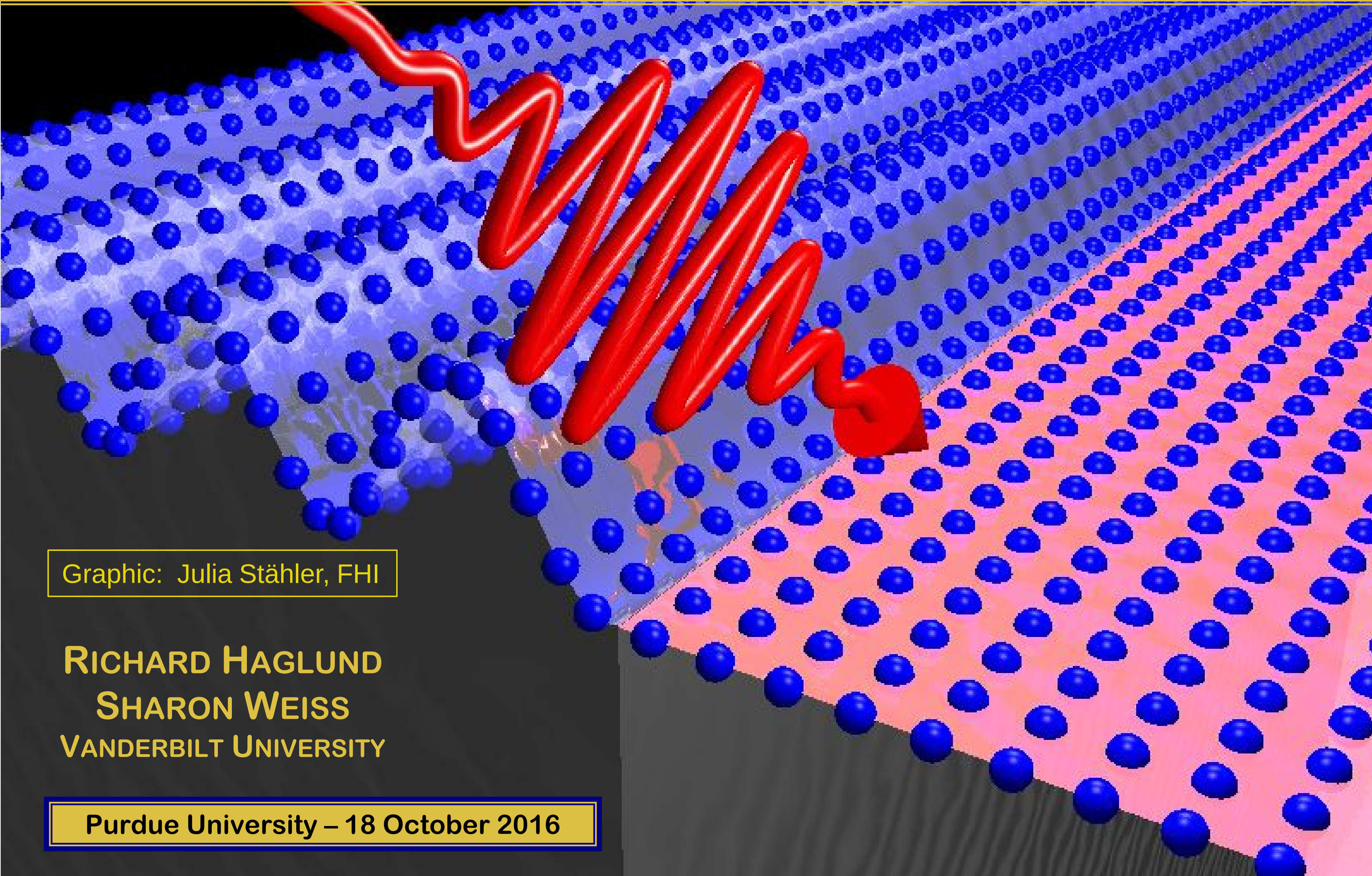
A PHASE-CHANGING OXIDE FOR PS SILICON PHOTONICS



Graphic: Julia Stähler, FHI

RICHARD HAGLUND
SHARON WEISS
VANDERBILT UNIVERSITY

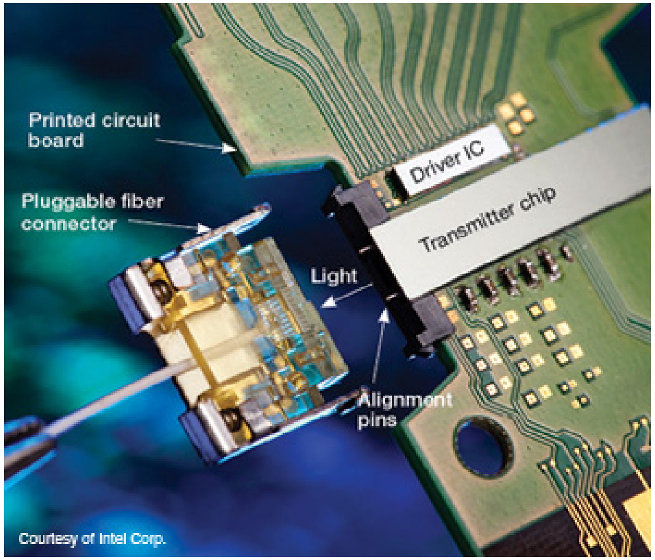
Purdue University – 18 October 2016

The story you are about to hear …

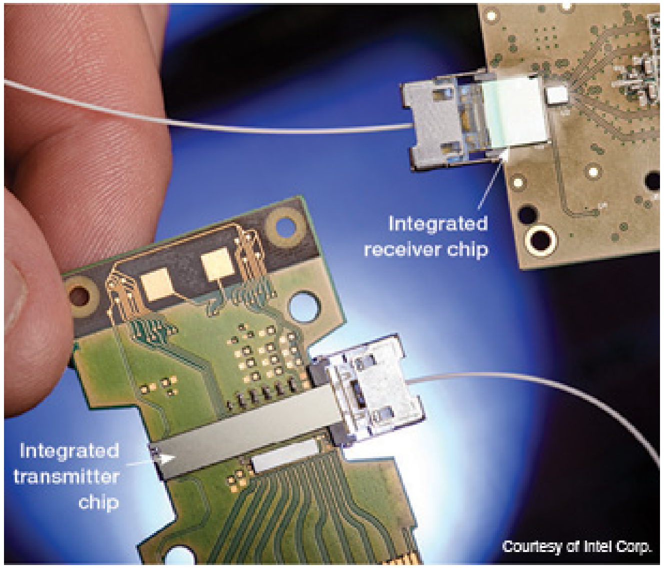
- **Motivation**: all-optical modulators for silicon photonics
- **Prologue**: The hybrid vanadium dioxide-silicon ring resonator
- **Ultrafast dynamics**: The role of epitaxy in PLD-grown films
- **Peroration**: Known unknowns and unknown unknowns

CMOS devices: short of area, speed and band-width.

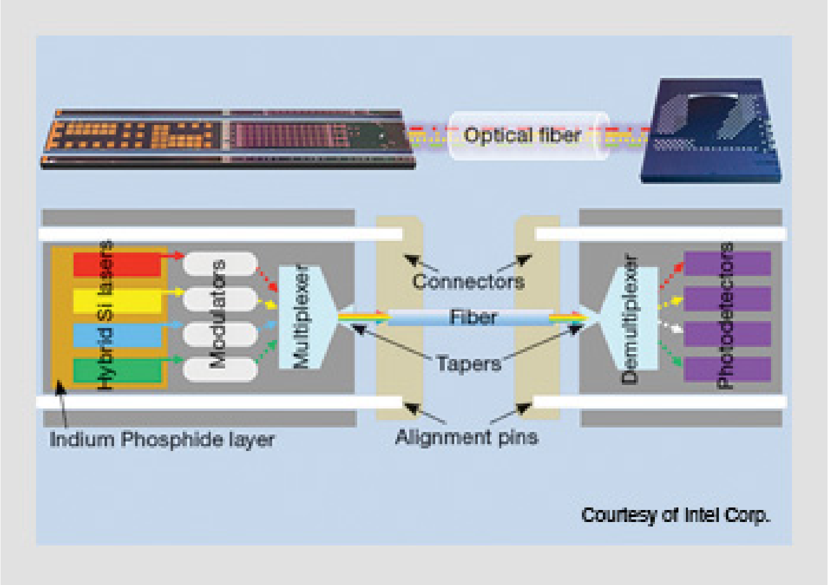
- Eighty per cent of CPU chip area now used for interconnects
- Data communication now more challenging than computation
- Current state-of-the-art of order GHz for modulators, data lines.



Courtesy of Intel Corp.



Courtesy of Intel Corp.



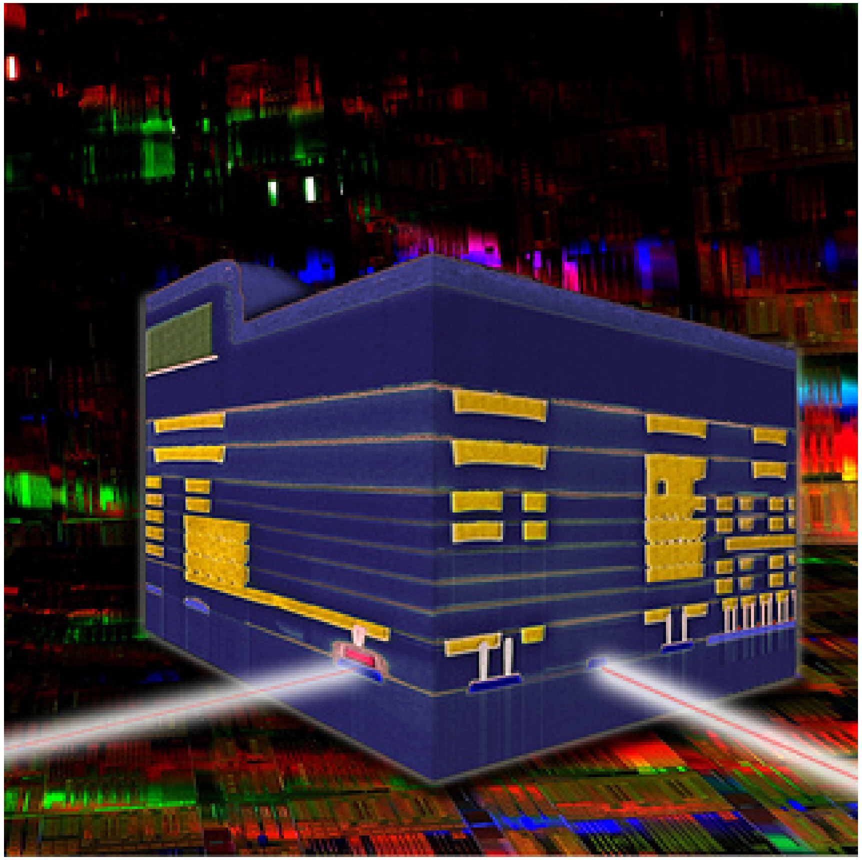
Courtesy of Intel Corp.

Is this the era of silicon nanophotonics?

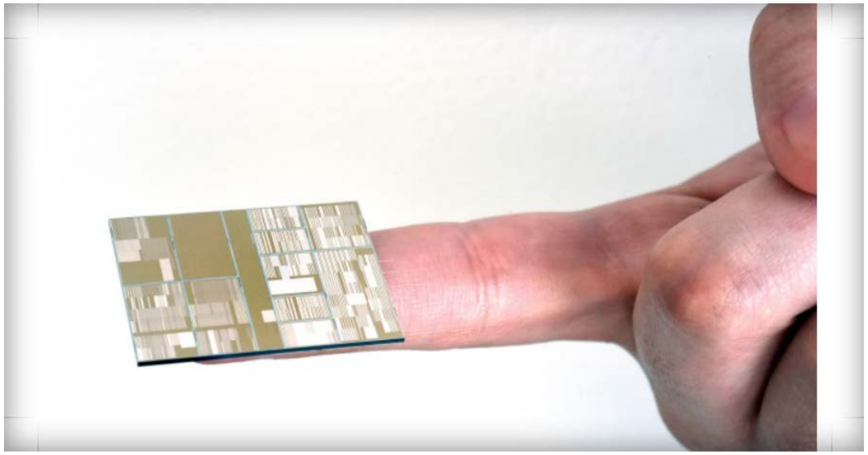
Made in IBM Labs: IBM Lights Up Silicon Chips to Tackle Big Data

- From the lab to the fab: Technology breakthrough demonstrates feasibility of **silicon nanophotonics** for chip manufacturing
- Light pulses can move data at blazing speeds to help solve bandwidth limitations of servers, datacenters and supercomputers
- After more than a decade of research, **silicon nanophotonics** is ready for development of commercial applications

(December 2012 IBM press release)



Inching closer to realization …



- IBM-Samsung-SUNY team announces in 2015
- Uses SiGe technology at the level of 7 nm FINFET on Semiconductor Map
- Challenge remains to find light sources and photo-detectors

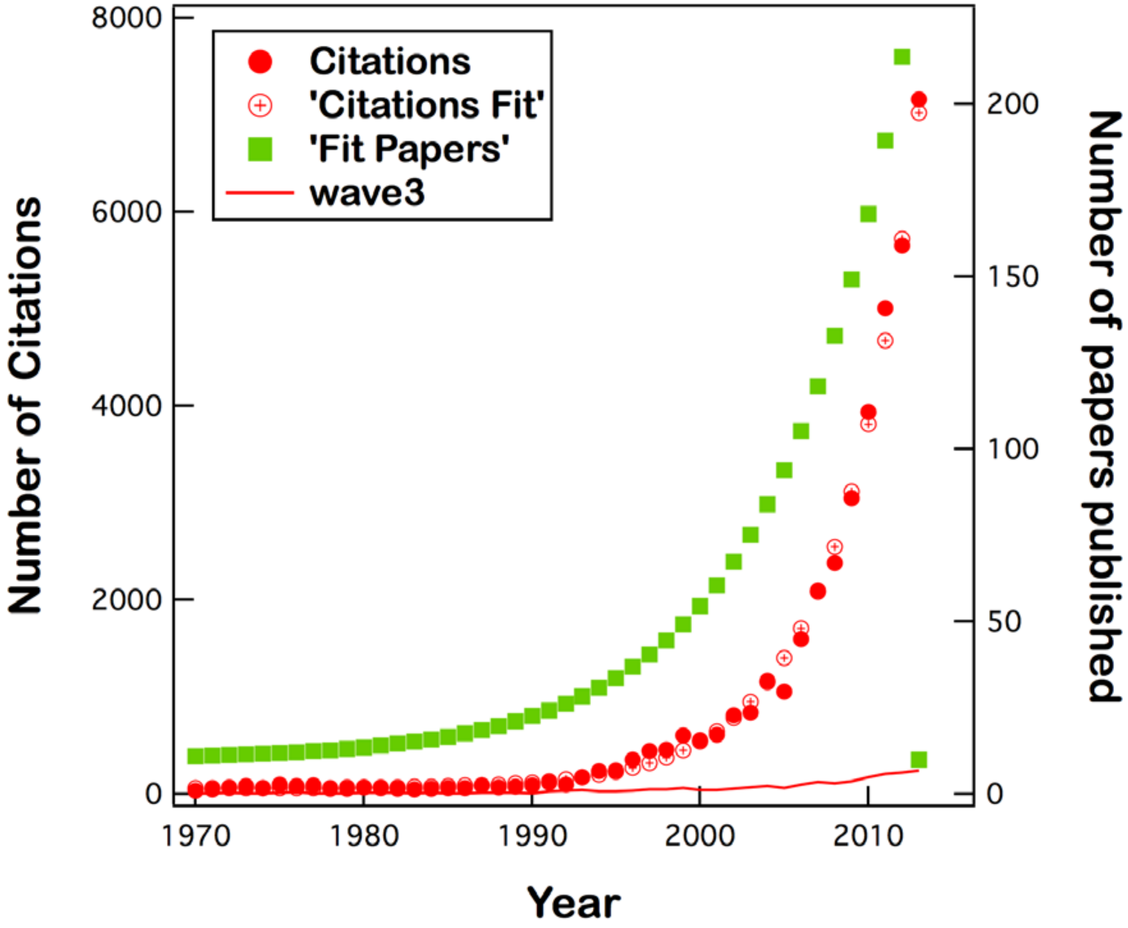
Joel Hruska (http://www.extremetech.com/author/jhruska) on July 9, 2015 at 7:30 am

Why not all-optical switching in all-silicon devices?

- Electro-optic effect is weak in silicon: $\Delta n \sim 0.001$, *ergo*
- Resonant structures must have high Q value, implying
- Relatively large size, long photon confinement, and
- Significant sensitivity to temperature fluctuations.

- ✓ Hybrid structures can separate silicon optics from the poor switching characteristics.
- But materials and processing compatibility are issues for many fast-switching materials (*e.g.*, polymers).
- ✓ Oxides can be compatible with silicon … so look for
- ✓ An oxide that can be grown on silicon devices and has large and ultrafast switching speed.

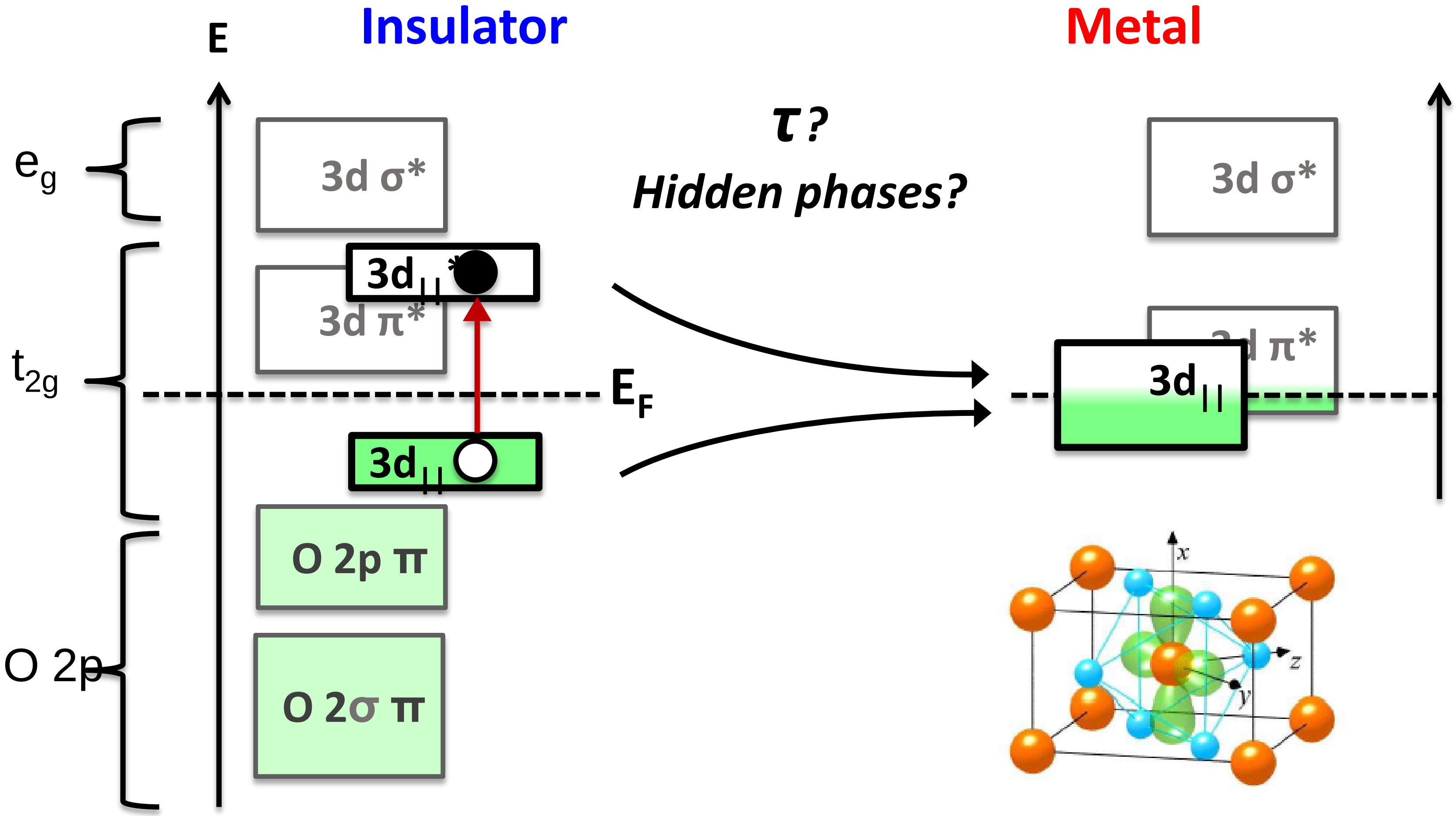
Mad for vanadium dioxide!



VO$_2$: Metal-insulator and structural phase transitions

- **Structural** phase transition changes long-range order from monoclinic to rutile at about 67˚C in bulk single crystals
- Metal-insulator transition is an **electronic** phase transition as well – gives factor 10^4 contrast in electrical resistivity
- Metallization proceeds percolatively in thin films, perhaps also in single crystals (subject of active research)

Origin of the insulator-to-metal transition in VO_2



E

Insulator

Metal

e_g

t_{2g}

O 2p

3d σ*

3d$_{||}$*

3d π*

E_F

3d$_{||}$

O 2p π

O 2σ π

τ?

Hidden phases?

3d σ*

3d π*

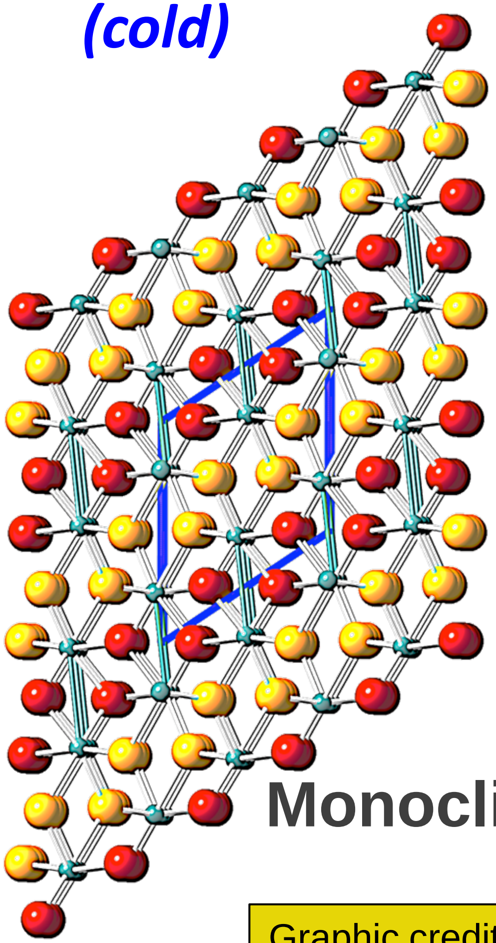
3d$_{||}$

Graphic credit: Marieke Jager, IBL

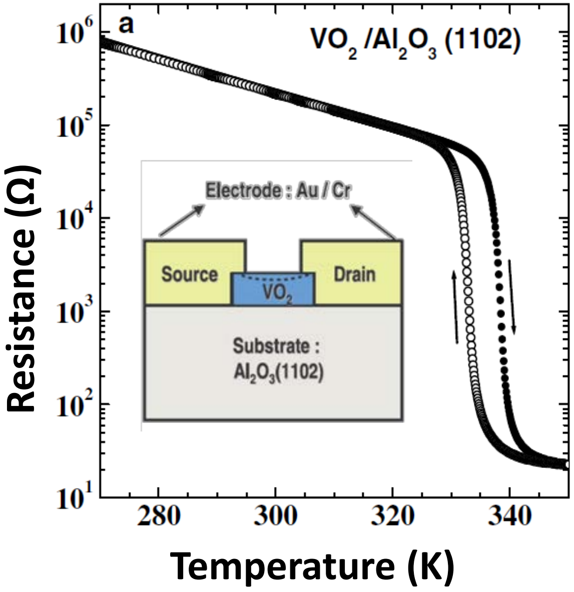
$d_{||}$ orbitals mediate V-V bonding

The monoclinic-to-rutile crystallographic transition

Insulator
(cold)



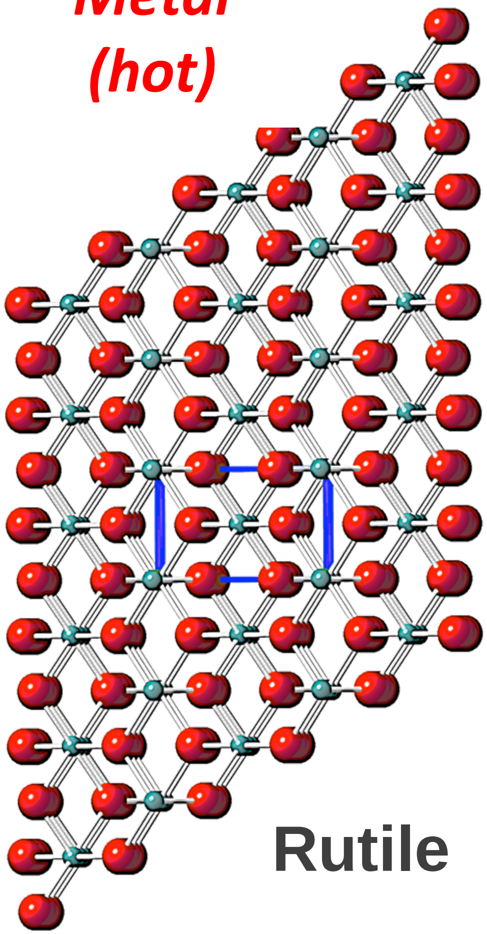
Monoclinic



H. T. Kim et al., New J. Phys. **6** (2004) 52.

Temperature
Pressure
Dopants
Light

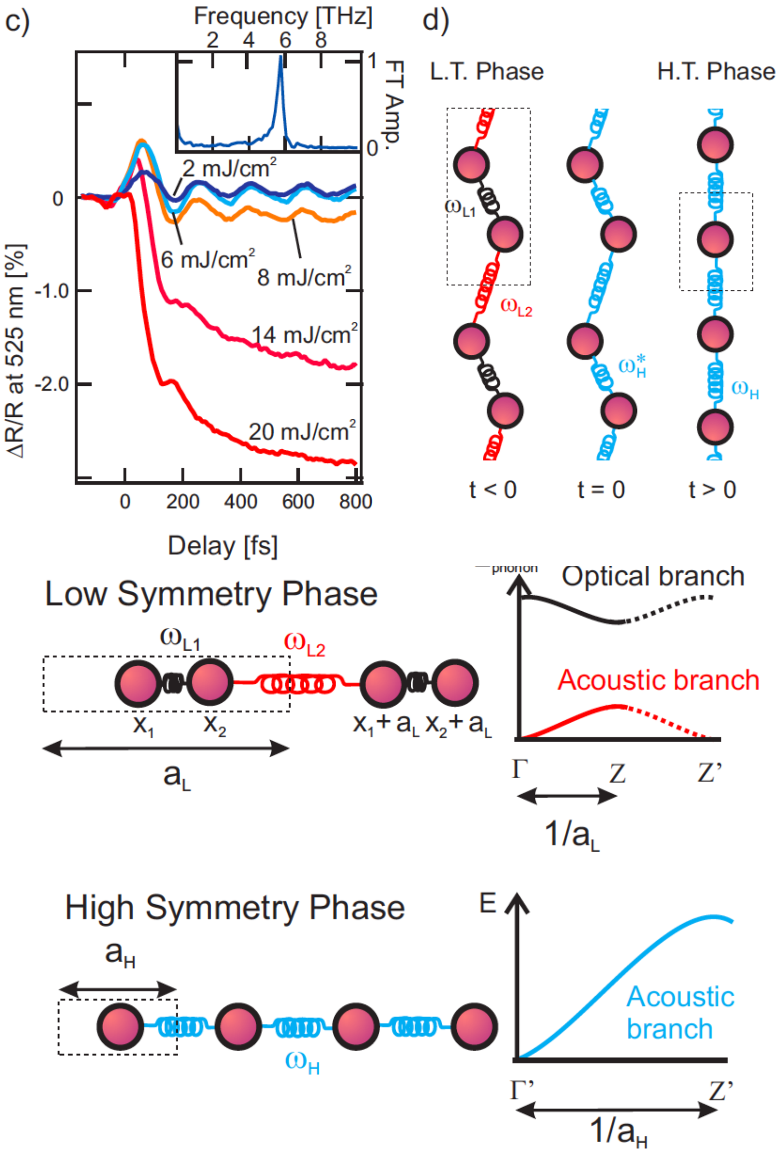
Metal
(hot)

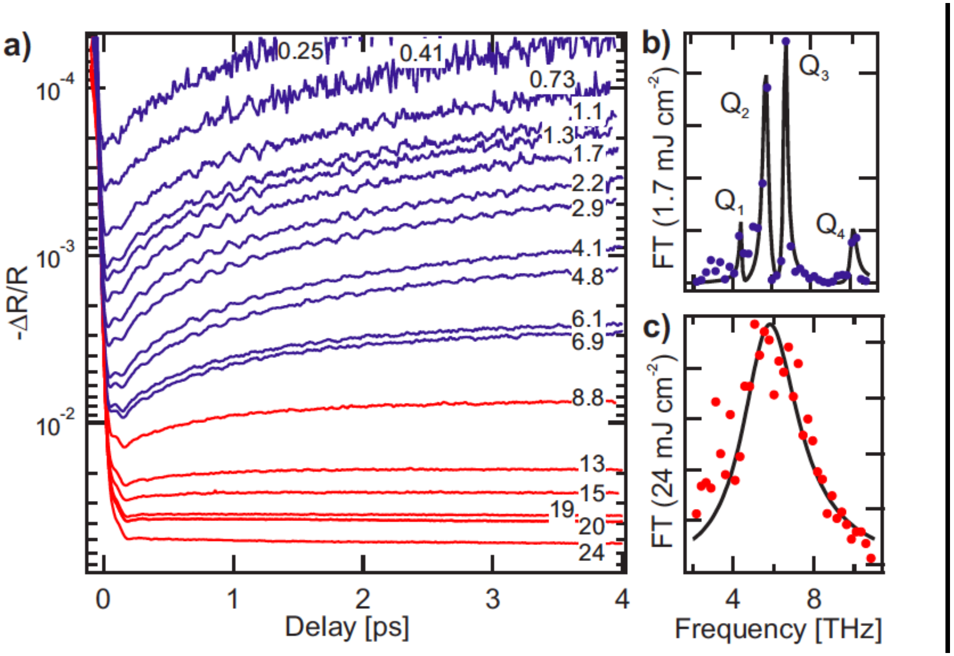


Rutile

Graphic credit: Marieke Jager, IBL

Fluence-dependent ultrafast phase-change dynamics

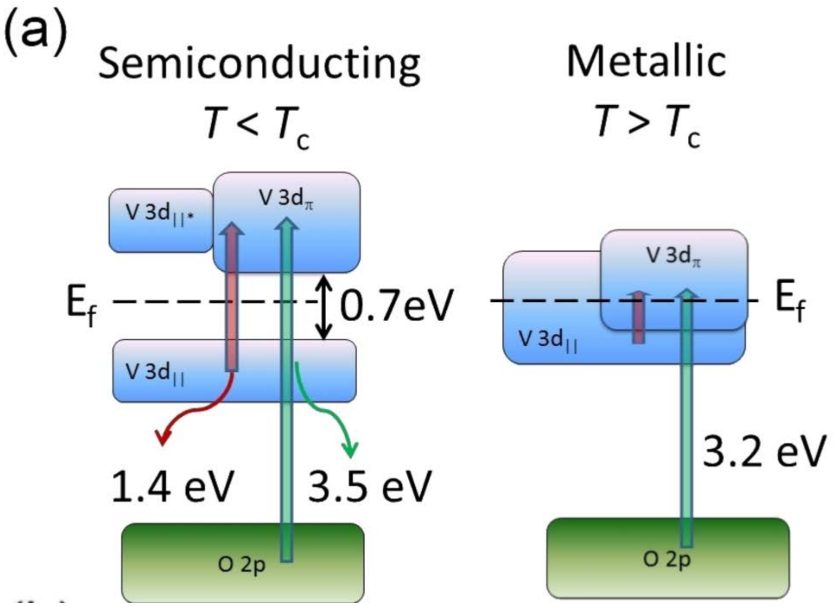


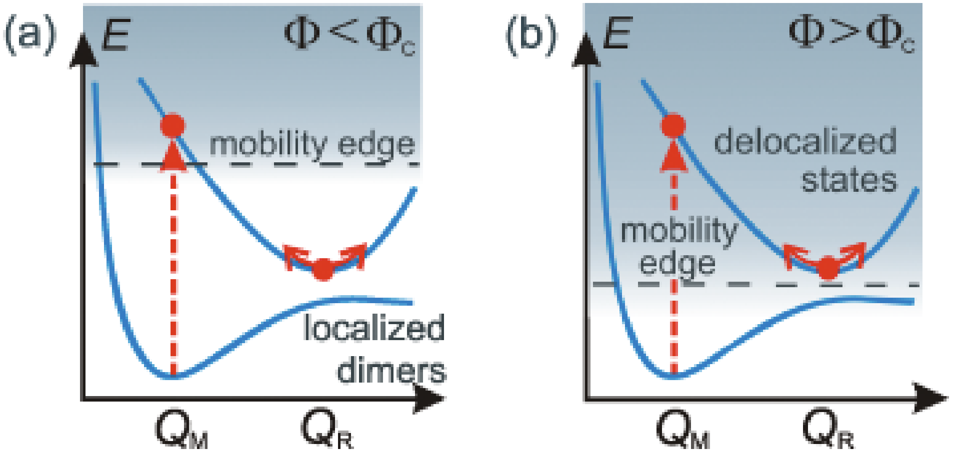


- Below critical fluence observe VO_2 Raman-active phonons
- Above critical fluence single THz mode signals change in lattice symmetry.

Wall et al, Nature Communications (2012)

Thermally *vs* optically driven phase transition





- Thermal (first-order) phase transition determined by **critical temperature**.
- Electronic (**IMT**), structural (**MRT**) phase transitions are not necessarily congruent!

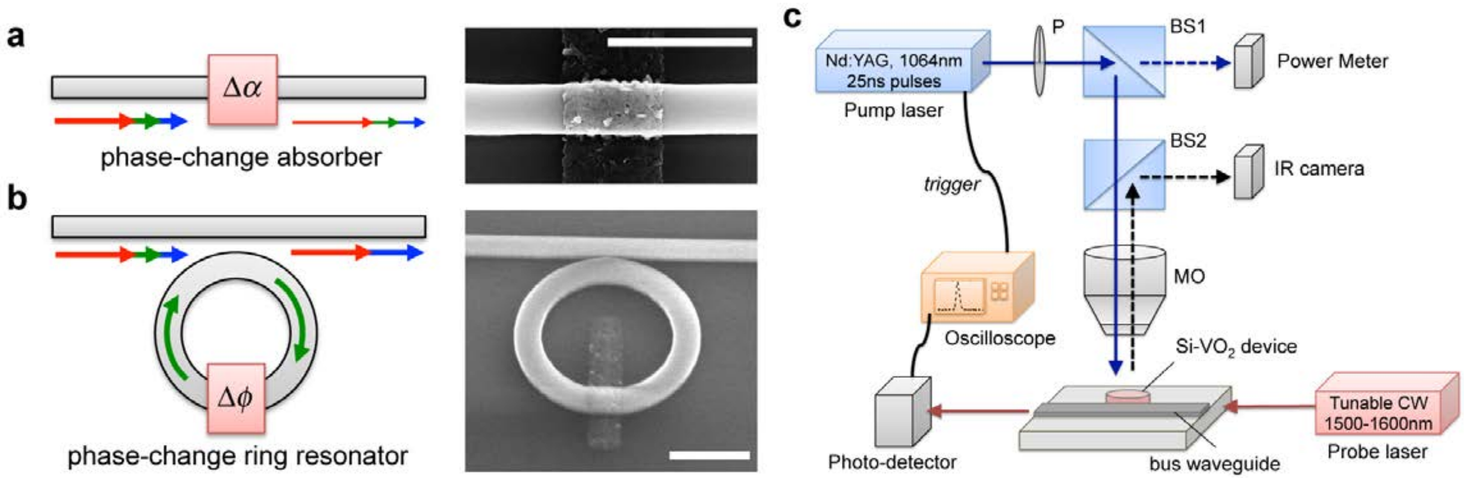
- Electronic (**IMT**, first-order?) phase transition driven by **critical fluence**.
- Structural (**MRT**) transition is driven by **coherent phonon generation**.

Pashkin, Physical Review B (2011)
van Veenendaal, Physical Review B (2013)

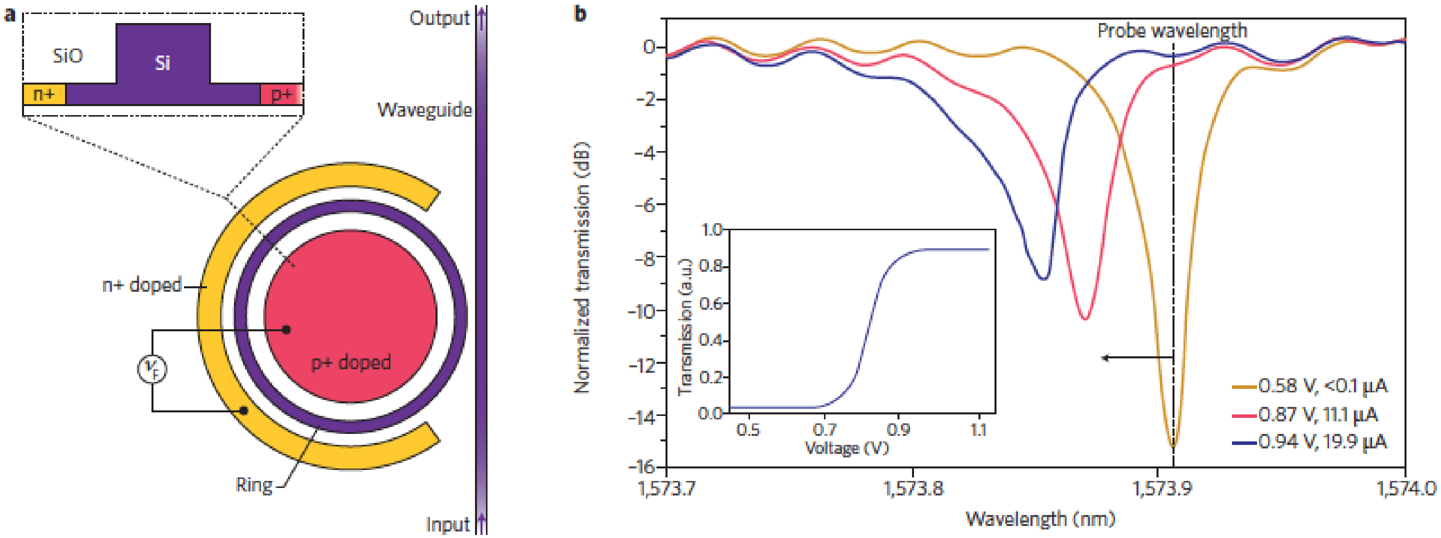
A hybrid ring resonator for ultrafast switching…

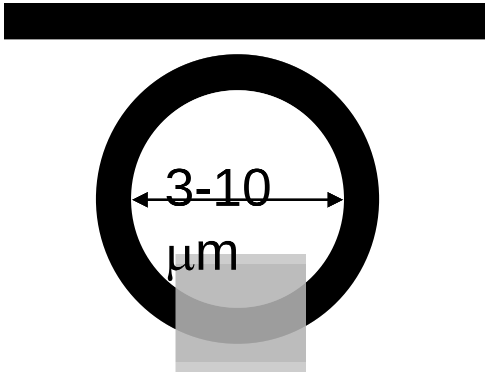
✓ **Motivation**: all-optical modulators for silicon photonics

- **Prologue**: The hybrid vanadium dioxide-silicon ring resonator
  - **Why vanadium dioxide?**
  - **Laser processing for the hybrid VO_2-Si ring resonator**
  - **Performance characteristics of the hybrid ring resonator**
- **Ultrafast dynamics**: The role of epitaxy in PLD-grown films
- **Peroration**: Known unknowns and unknown unknowns



It's the Δn, stupid! What about VO_2 for THz switching?





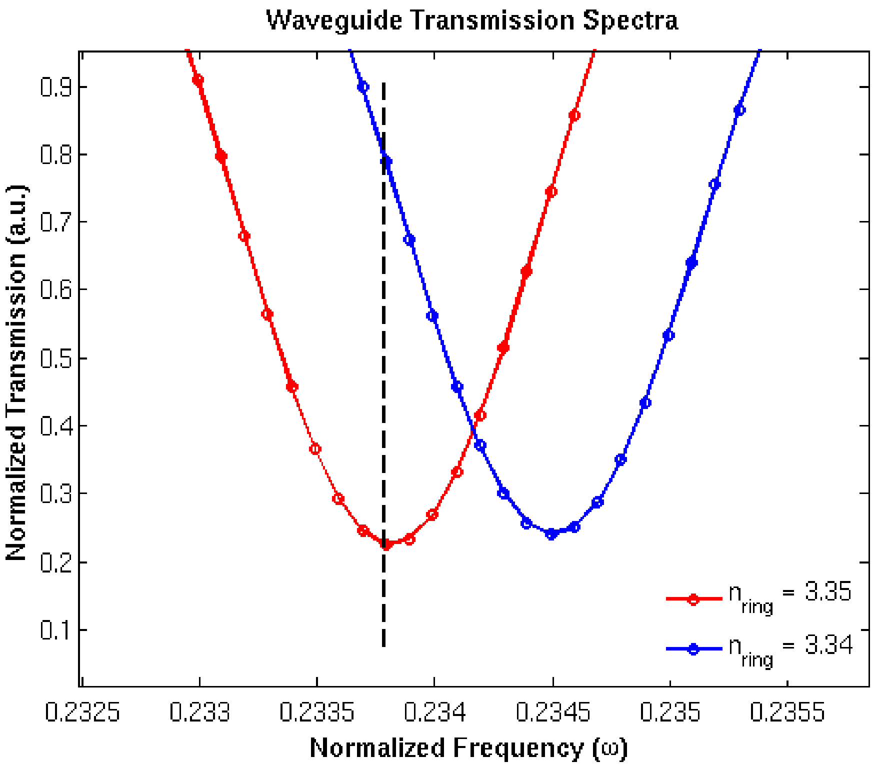
$$2\pi R\cdot\Delta n = m\cdot\Delta\lambda$$

$$\tau_{cavity} = \frac{\lambda^2}{2\pi nc\cdot\Delta\lambda}$$

Ryckman, Nag et al., *Optics Express* **21**, 10753 (2013)

Ring resonator – a canonical waveguide modulator

- FDTD simulations show how nonlinear coupling of ring and waveguide can shift resonant frequency
- Simulated transmission shows: for Δn = 0.01 resonance ω shifts by 0.75% , i.e., Δλ~3 nm near λ = 1550nm
- Simulated electric field distribution at resonance frequency and detuned from it by Δn = 0.01

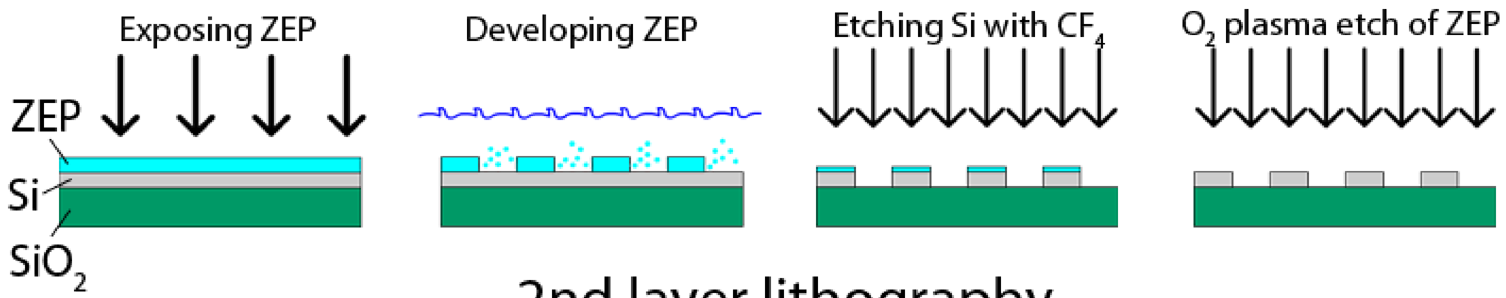


Ring resonator as logic circuit: three-port router

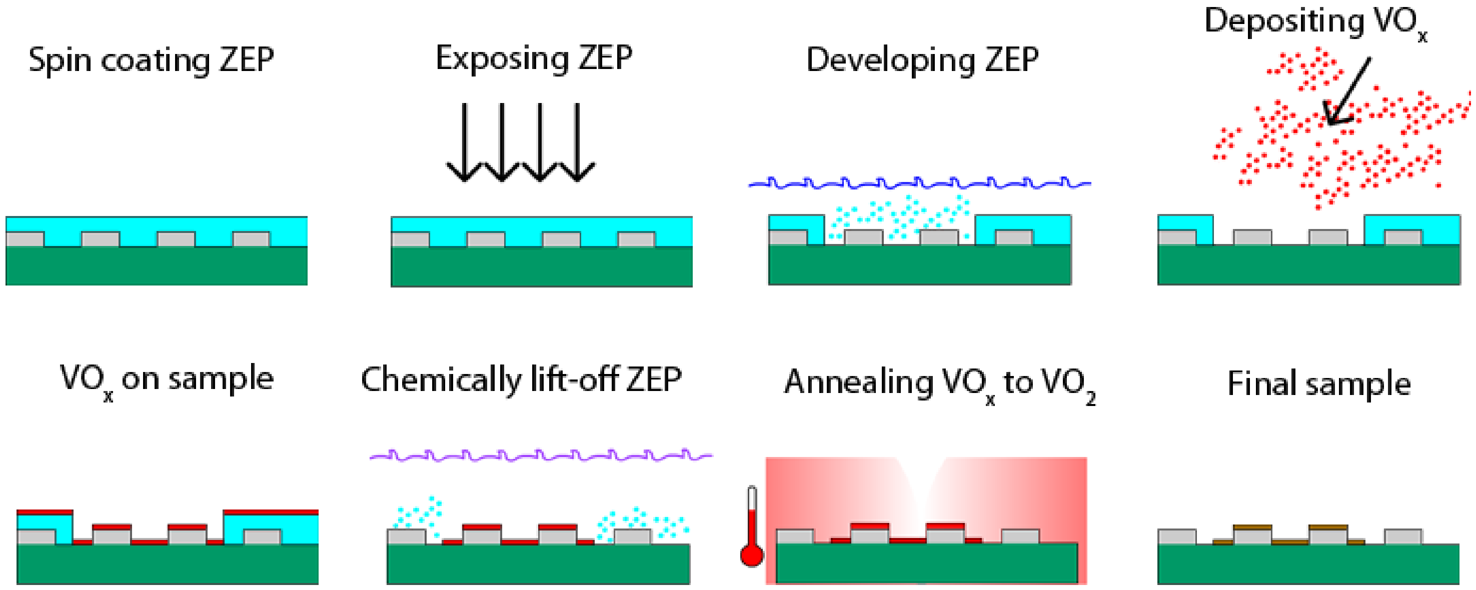
- If ring is resonant, pulses are **dropped** from wave-guide and stored in (**added to**) the resonator.
- If ring is **not** resonant, pulses continue in the wave-guide.
- Various combinations of ring resonators can perform logic operations on light pulses

Some tricky fabrication issues … but it works.

1st layer lithography

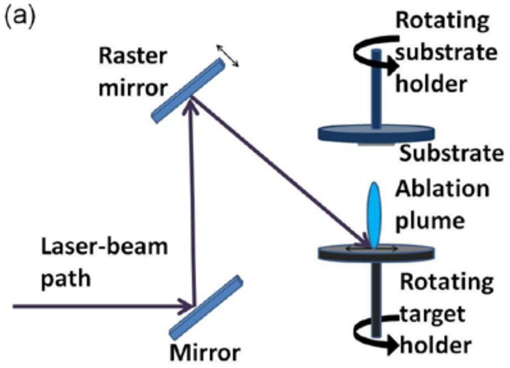


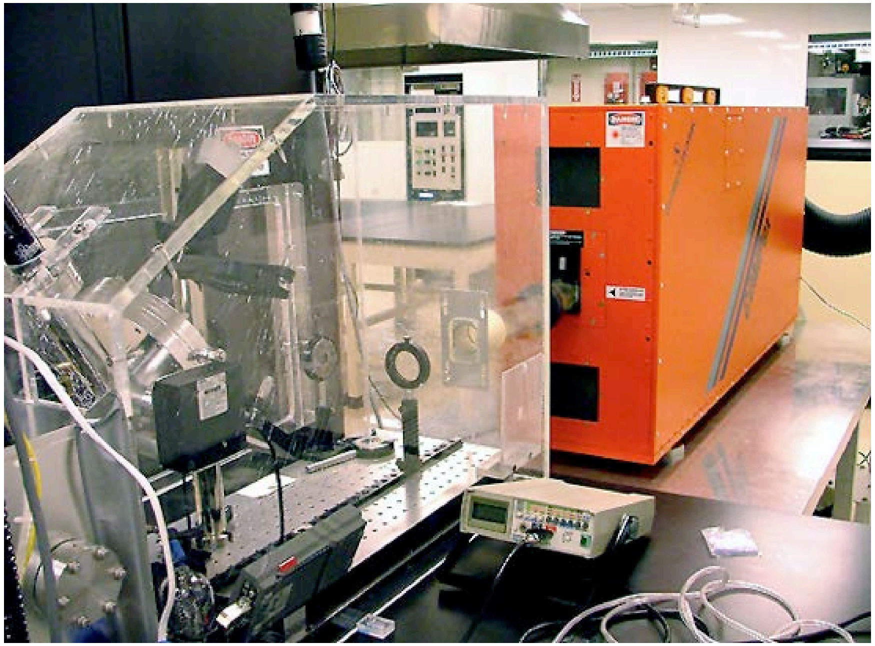
2nd layer lithography



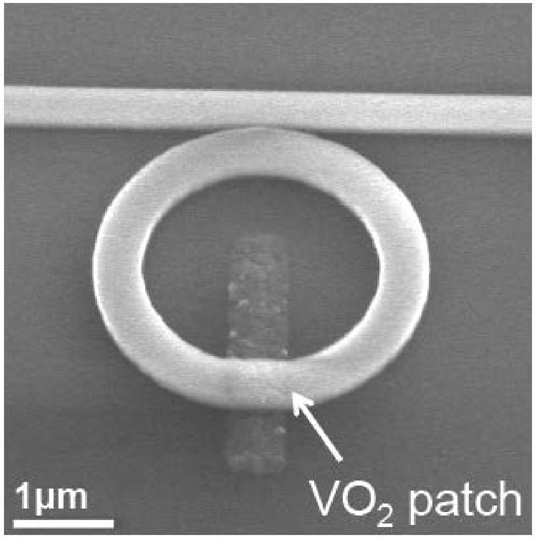
Pulsed laser deposition of VO_2

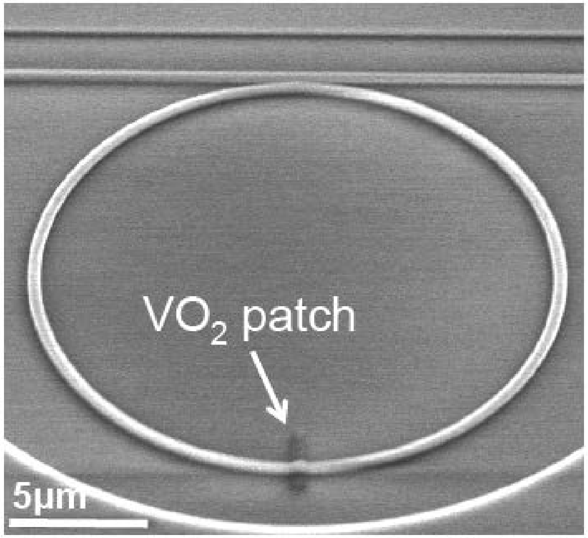
- Vanadium metal target in 10 mTorr O_2
- KrF pulsed laser ablation (248 nm), prf 10-25 Hz, fluence 1.2 J/cm^2
- Pulsed laser deposition yields amorphous $VO_{1.7}$
- Anneal 2-30 min in 250 mTorr O_2 to get polycrystalline VO_2
- Thickness by shot count and/or profilometry.

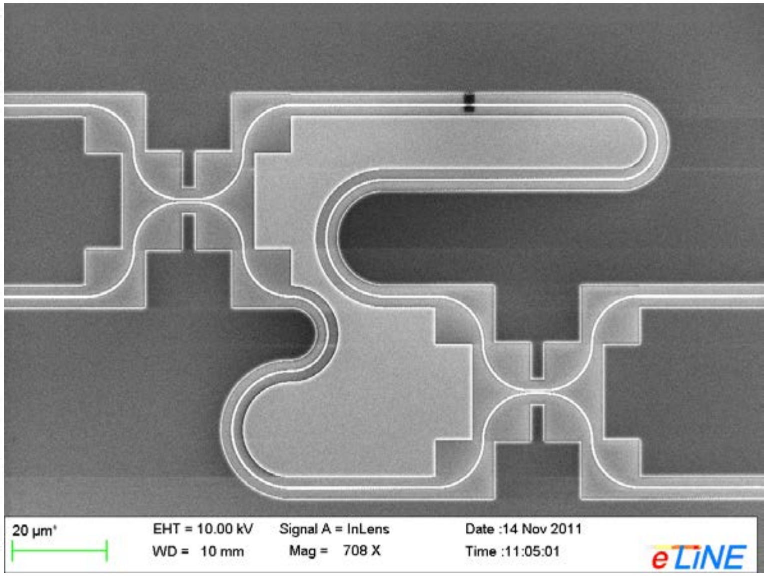




Multiple rings and interferometers fabricated

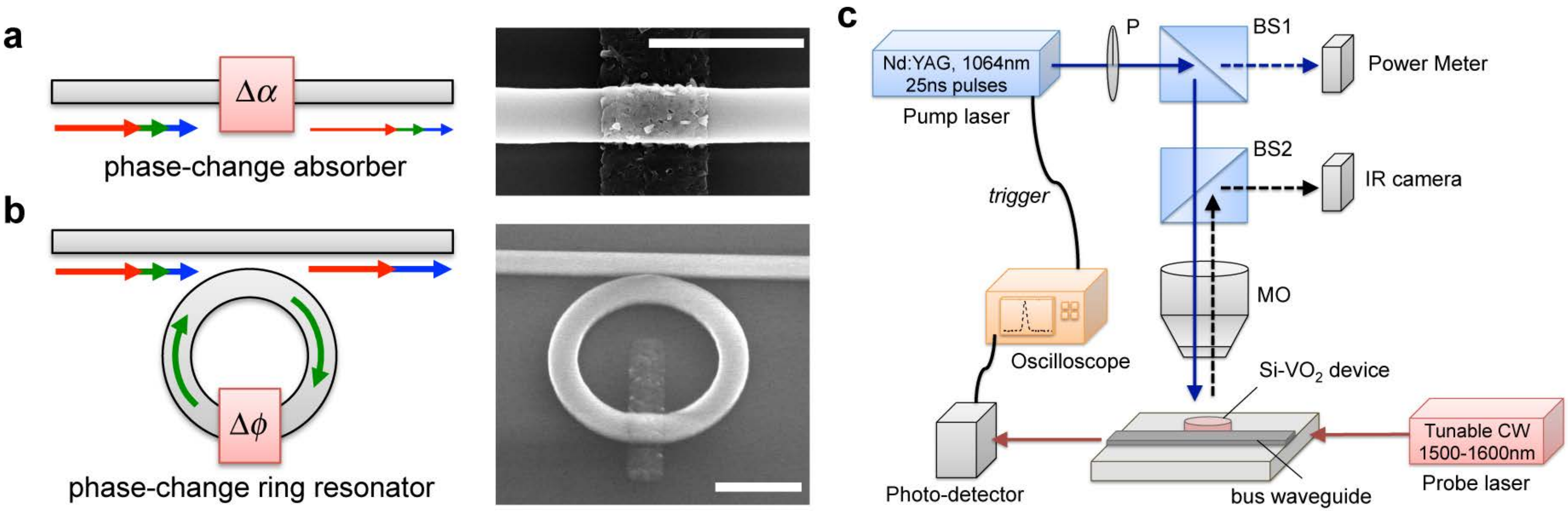






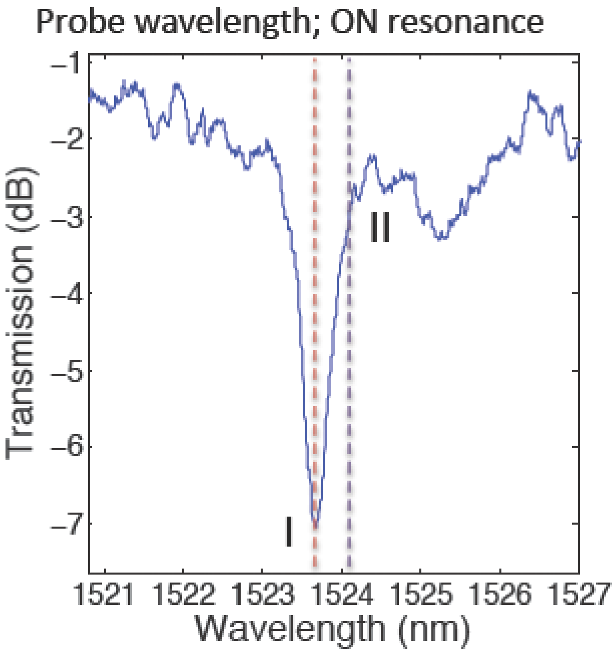
- VO_2 deposited by pulsed laser deposition in first experiments
- More recently have used electron-beam evaporation
- V_2O_4 powder rather than metallic vanadium target (cheaper)
- Initial fabrication at Vanderbilt using Raith e-Line EBL system
- Subsequent “industrial strength” fabrication at Oak Ridge National Laboratory Center for Nanophase Materials Sciences

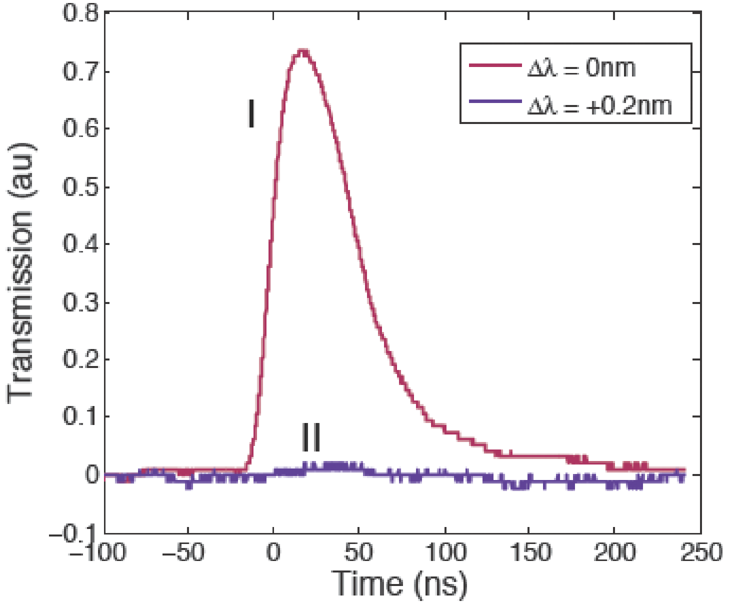
Hybrid Si-VO$_2$ modulator structures



- Objective: show single-pulse switching, establish threshold
- As before, tunable cw probe beam in resonance region
- (a) wave-guide absorber, (b) hybrid ring resonator
- (c) Schematic of experimental layout

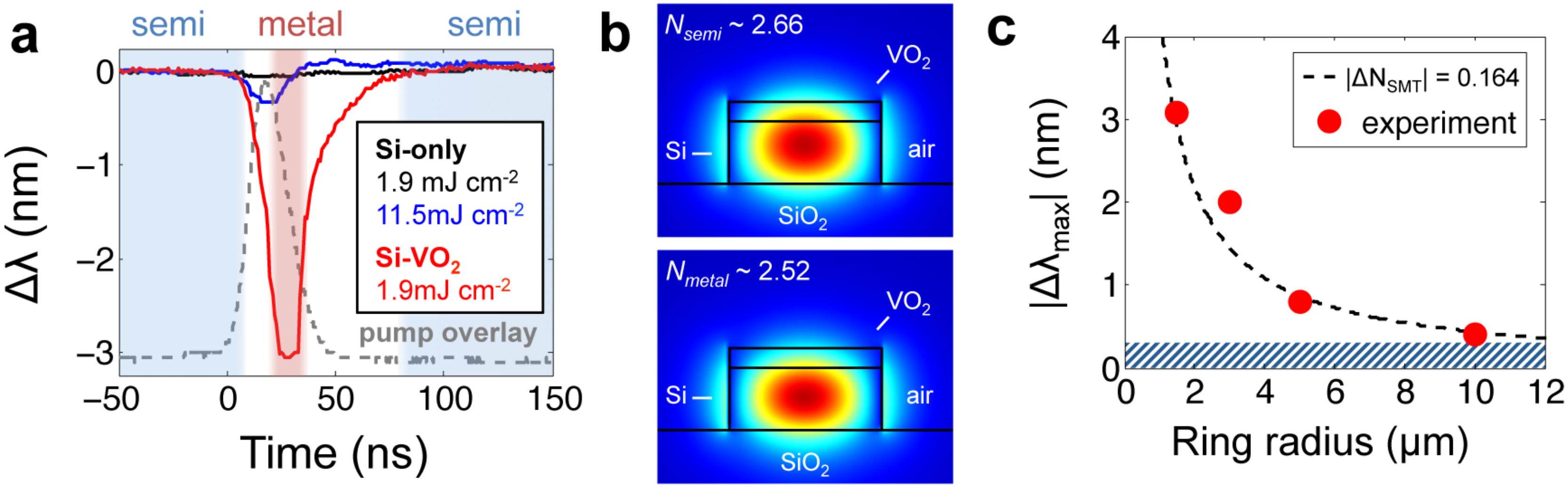
On- *vs* off-resonance transmission vs time





- Test on large-diameter (10 µm) ring resonator
- Tune the probe wavelength on (I) and off (II) resonance
- Relaxation time longer than pulse duration …
- Consistent with relaxation of metallic (rutile) phase of VO_2

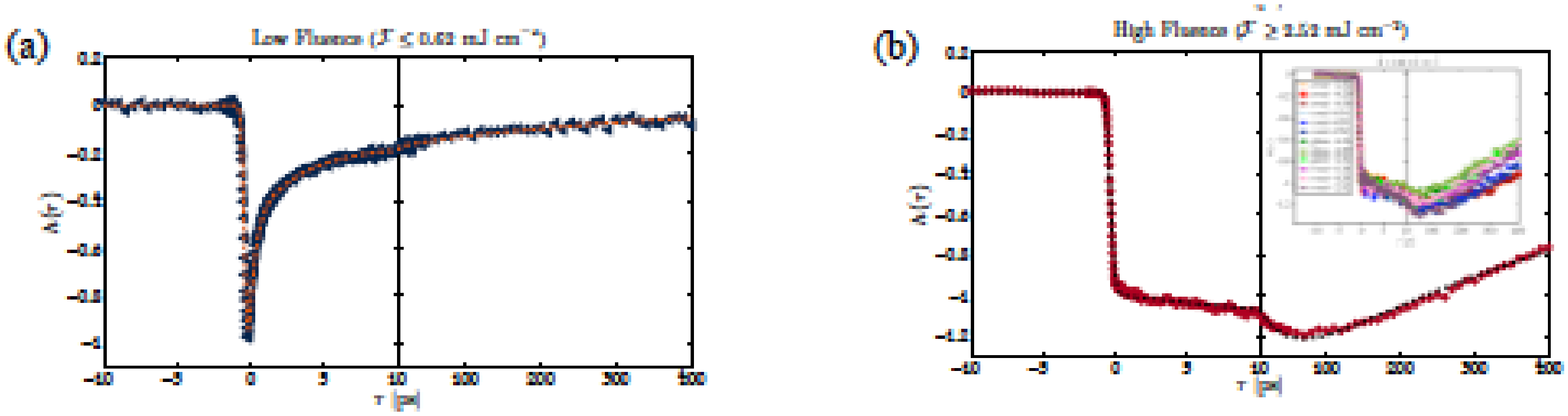
Ring resonator performance summary



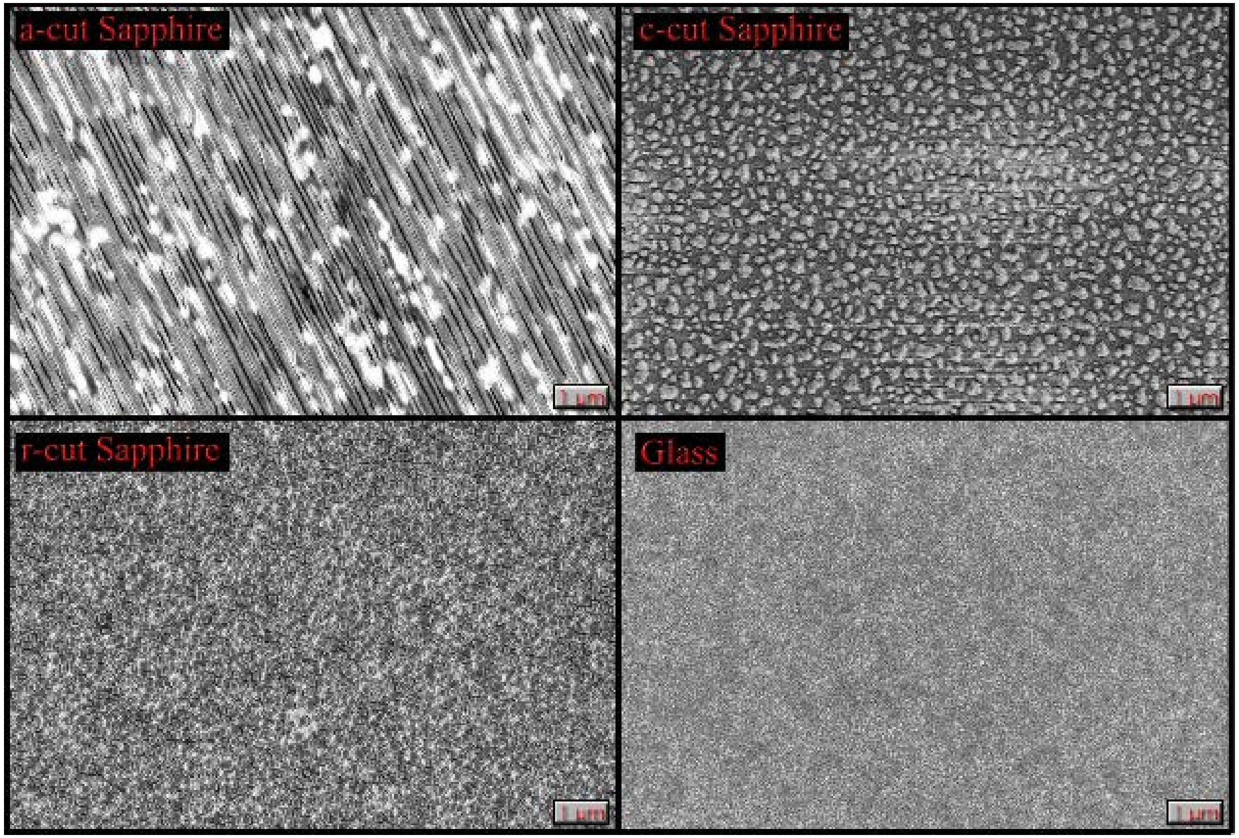
- (a) Wavelength shift *vs* time for Si-only and Si-VO_2 hybrid
- (b) RDTD mode simulation for the hybrid waveguide, with a 70 nm thick VO_2 patch
- (c) Magnitude of the wavelength blue shift for the photo-induced SMT in hybrid ring resonators of varying ring radii with fixed 500 nm VO_2 patch length

What about the slow recovery time of rutile VO_2?

- ✓ **Motivation**: all-optical modulators for silicon photonics
- ✓ **Prologue**: The hybrid vanadium dioxide-silicon ring resonator
- ● **Ultrafast dynamics**: The role of epitaxy in PLD-grown films
  - o **A puzzle: apparently inhomogeneous dynamics in VO_2 crystals**
  - o **Effects of epitaxy in dynamics of PLD-grown thin VO_2 films**
  - o **A puzzle solved? Universal scaling of dynamics in thin films**
- ● **Peroration**: Known unknowns and unknown unknowns



Ultrafast dynamics for distinctly different thin films



SEM images of the four samples studied in this work. All samples are the same thickness (80 nm) and were grown using the same protocol

Samples grown on the **a-cut** and **c-cut** sapphire have a lateral correlation length of
$\Lambda_1 \sim 500$ nm

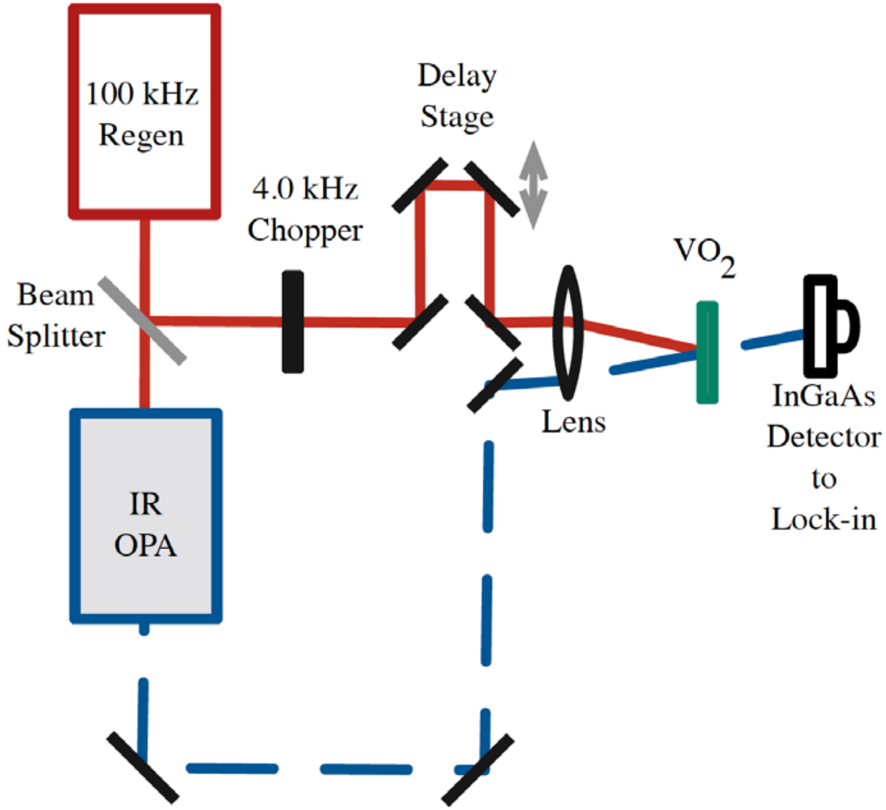
Samples grown on the **r-cut** sapphire have a lateral correlation length of
$\Lambda_2 \sim \Lambda_1/10 \approx 50$ nm

Samples on glass have only short range order.

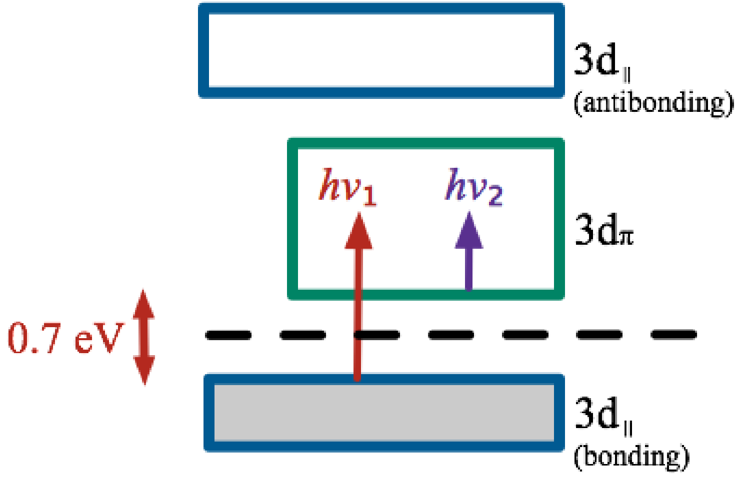
Brady *et al*., *J Phys Condensed Matter* (2016)

A simple experiment to compare four thin VO_2 films

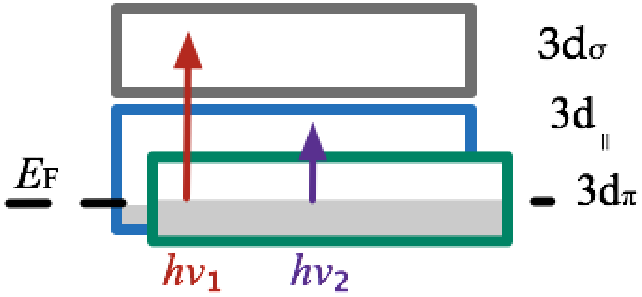
Photoexcite at 800 nm $\hbar\omega \sim 2E_{gap}$
Probe at 3100 nm $\hbar\omega \sim E_{gap}/2$



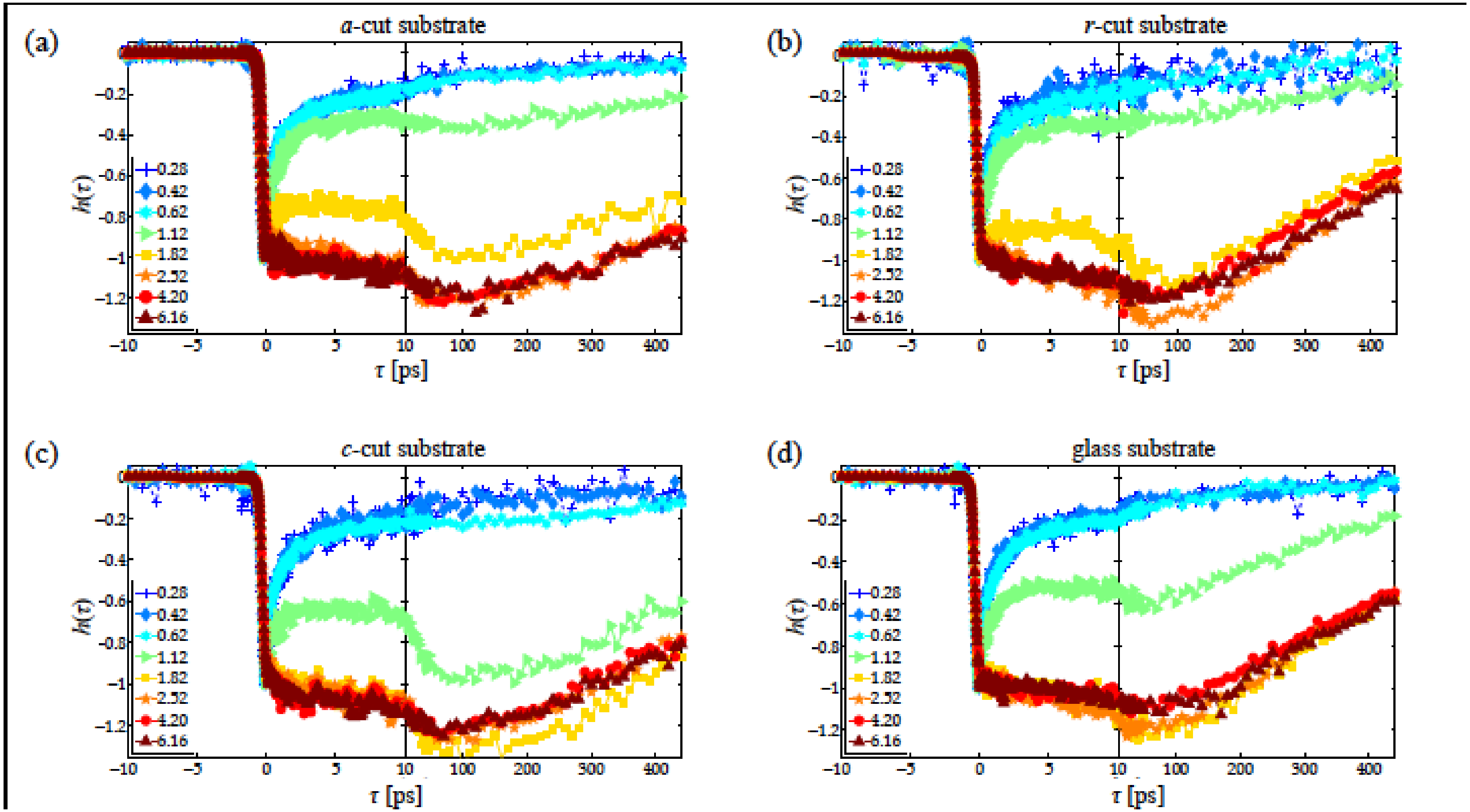
In M1 phase, probing free-carrier absorption in $3d_\pi$ orbitals



In R phase, probing free-carrier absorption near E_F



Ultrafast dynamics in quartet of VO_2 samples



- Note similarity between below threshold (blue) and above threshold (gold, brown, amber) decay histories **across all sample types**

Nucleation time for rutile phase: 40.5±1.2 ps

$$\frac{\Delta T}{T}(t)=\frac{\sigma\sqrt{\pi}}{2}\sum_{j=1}^{3}A_j\left[1-\exp\left(-\frac{t}{\tau_a}\right)\right]\exp\left(\frac{\sigma^2}{4\tau_j^2}-\frac{t}{\tau_j}\right)\operatorname{erfc}\left(\frac{\sigma}{2\tau_j}-\frac{t}{\sigma}\right)$$

$\tau_1 \Rightarrow (1/e)$ thermalization time

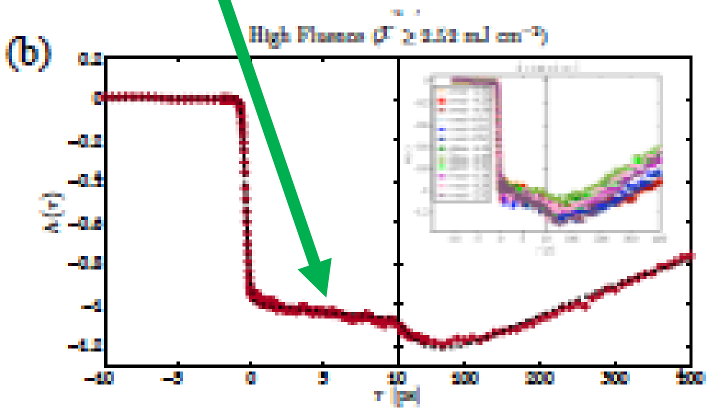
$\tau_2 \Rightarrow (1/e)$ interband transition coupling time

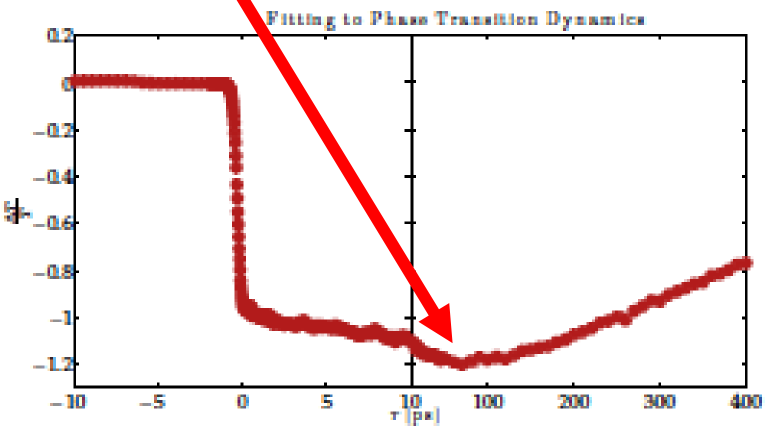
$\tau_3 \Rightarrow (1/e)$ thermal coupling time to substrate

$\tau_a \Rightarrow$ nucleation time for metallic phase $= 40.5 \pm 1.2$ ps

Johnson-Mehl-Avrami-Kolmogorov phase nucleation theory (1940)

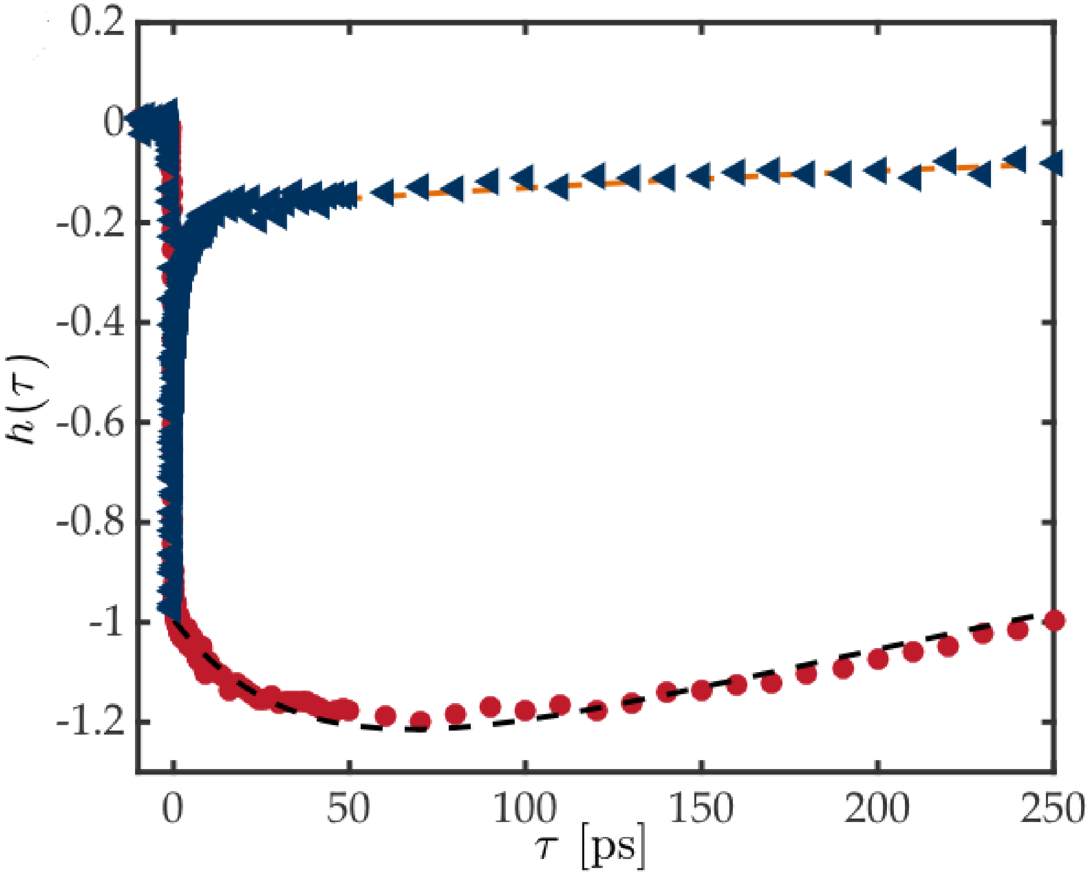
* **Limiting switching speed if rutile transition completed: 25 GHz**





Only two dynamical regimes regardless of epitaxy!

$$\frac{\Delta T}{T}(t) = \frac{\sigma\sqrt{\pi}}{2}\sum_{j=1}^{3} A_j \left[1-\exp\left(-\frac{t}{\tau_a}\right)\right] \exp\left(\frac{\sigma^2}{4\tau_j^2}-\frac{t}{\tau_j}\right)\operatorname{erfc}\left(\frac{\sigma}{2\tau_j}-\frac{t}{\sigma}\right)$$



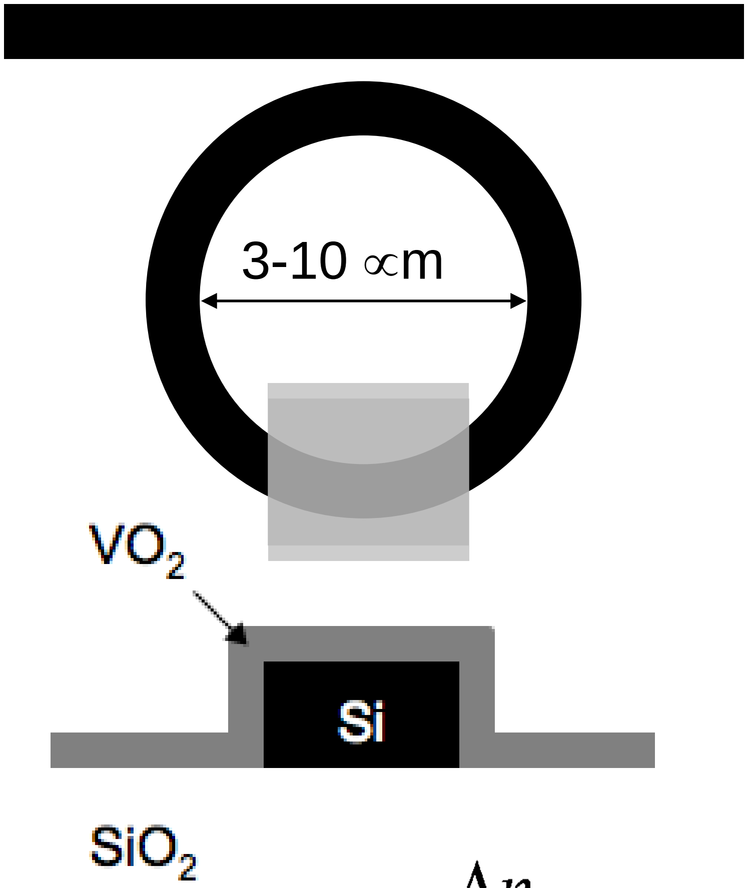
- Fast (semiconductor) dynamics at fluences below ~1.5 mJ/cm^2
- At this fluence, switching response of hybrid VO^2-Si ring resonator stable!
- Slow (JMAK nucleation) dynamics at fluences above 3 mJ/cm^2
- For slow dynamics, Δn reaches maximum 1.3; hybrid ring needs only Δn ~0.15!

So what have we learned?

- ✓ **Motivation**: all-optical modulators for silicon photonics
- ✓ **Prologue**: The hybrid vanadium dioxide-silicon ring resonator
- ✓ **Ultrafast dynamics**: The role of epitaxy in PLD-grown films
- ● **Peroration**: Known unknowns and unknown unknowns
  - o Comparing pulsed laser deposition, sputtering and e-beam evaporation
  - o Challenges for on-chip, THz switching speeds
  - o Next steps: ultrafast switching at the VO_2 band edge

Peroration, n. The explosion of an oratorical rocket. It dazzles, but to an observer having the wrong kind of nose its most conspicuous peculiarity is the smell of the several kinds of powder used in preparing it. (Ambrose Bierce, *The Devil's Dictionary*)

How to process VO_2 for optimal mesoscale response?

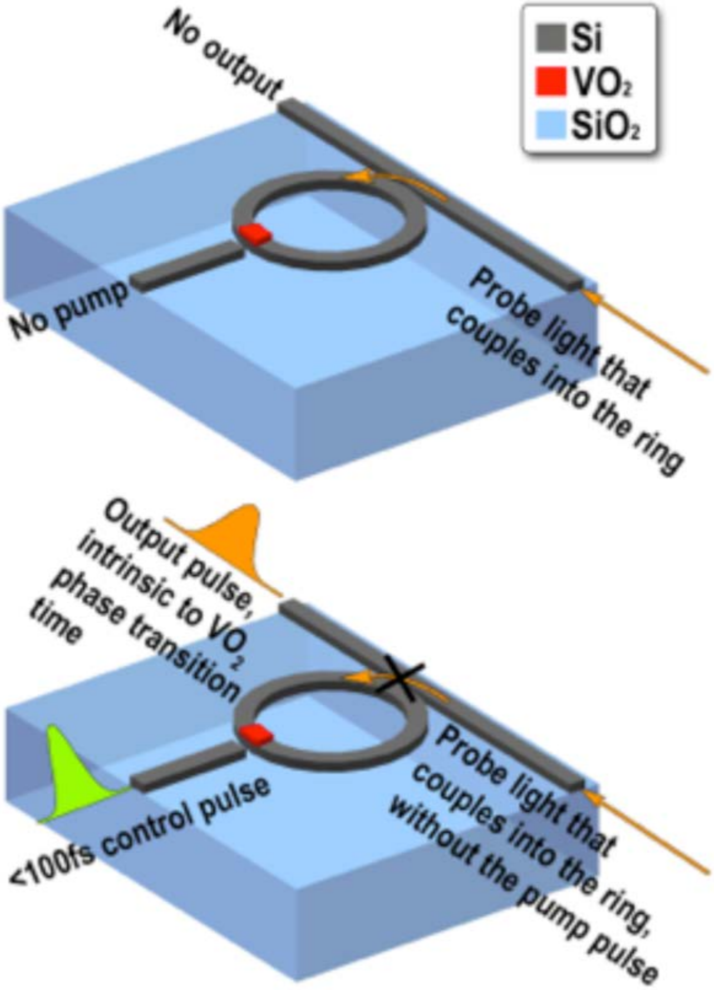


$$\Delta n$$

$$\Delta n(t)$$

- ✓ Δn in VO_2 during IMT ≤0.2 seems to be sufficient for phase change.
- ✓ Soon will know how much Δn is needed for ultrashort pump pulses
- ⊙ **But we don’t know $\Delta n(t)$!**
- ? Best semiconducting phase?
- ? How does doping or strain matter?
- ? How much VO_2 is really needed?
- ? Effects of nanoscale domains?
- ? **Can we reach 100 fJ per bit?**

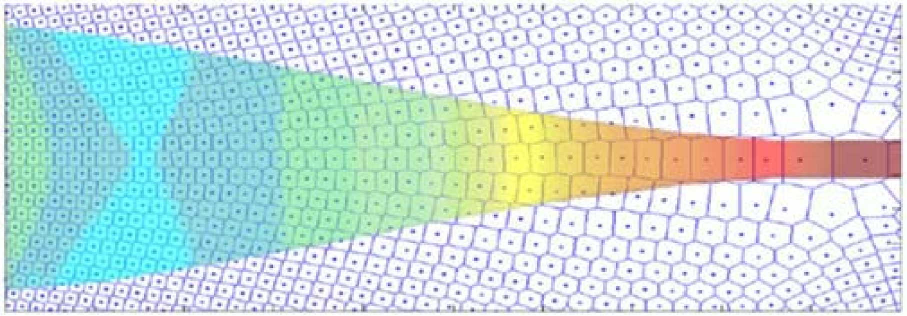
The dream of all-optical switching

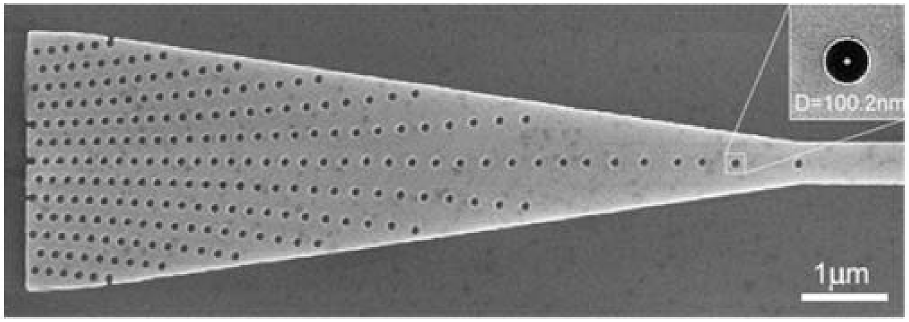


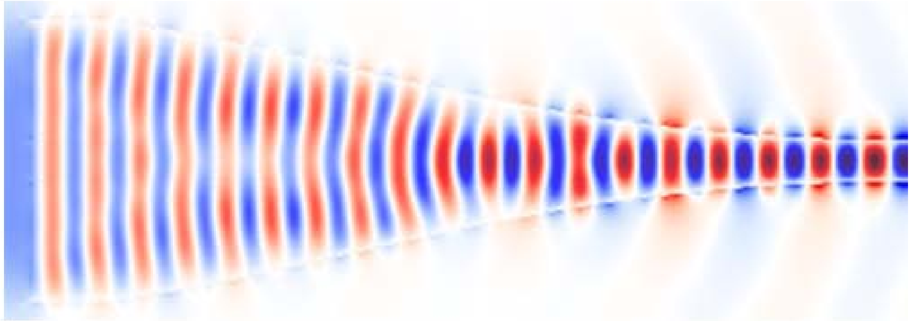
- Proof-of-principle experiment at GHz frequencies required only $\Delta n \sim 0.13$!!
- Comparable response to all-silicon ring resonators
- To go to Tbps switching, need to demonstrate
  - Efficient coupling of pump light into sub-micron VO_2 patch
  - Efficient pumping of VO_2 phase transition at near band-edge wavelength

Optical pumping may require transformation optics

- Have to couple via silicon waveguide with tapered coupling to VO_2 “patch”
- Design based on FDTD simulations of electromagnetic field patterns
- Field patterns (bottom) show low loss, efficient coupling …
- … but still have to consider dispersion in sub-100 fs pump pulses



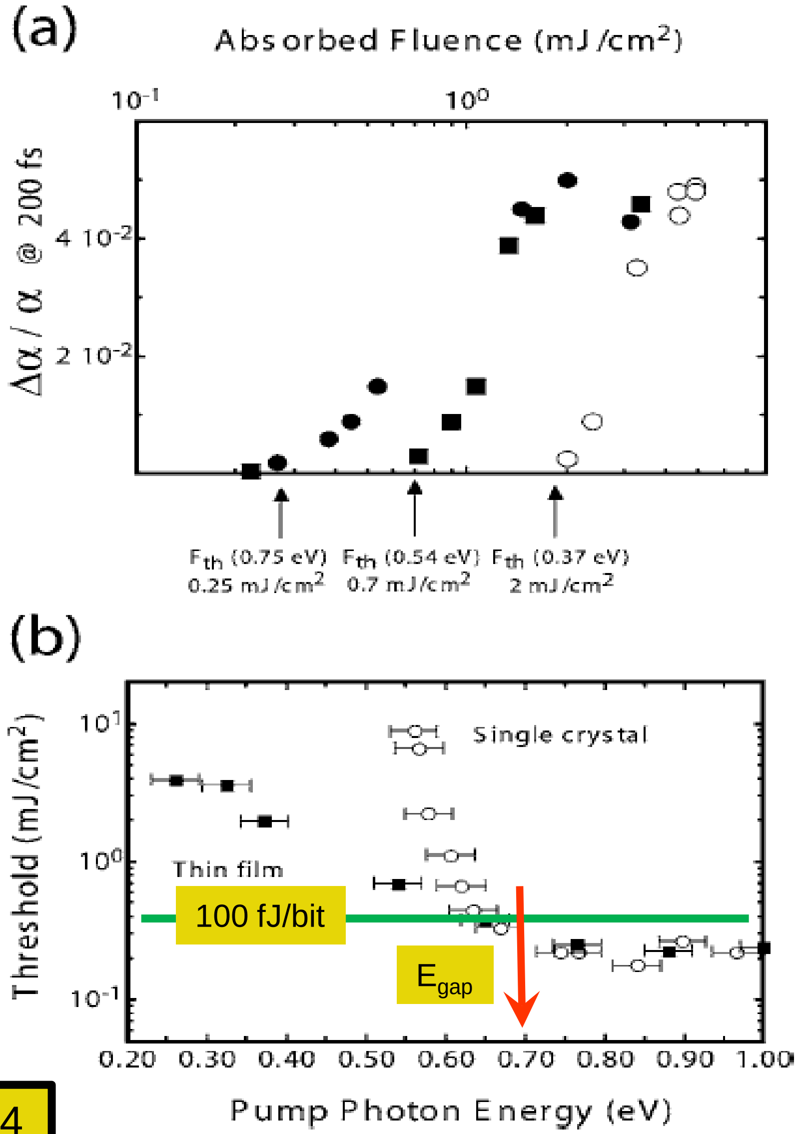




Markov, Valentine, Weiss, *Optics Express* **20**, 14705

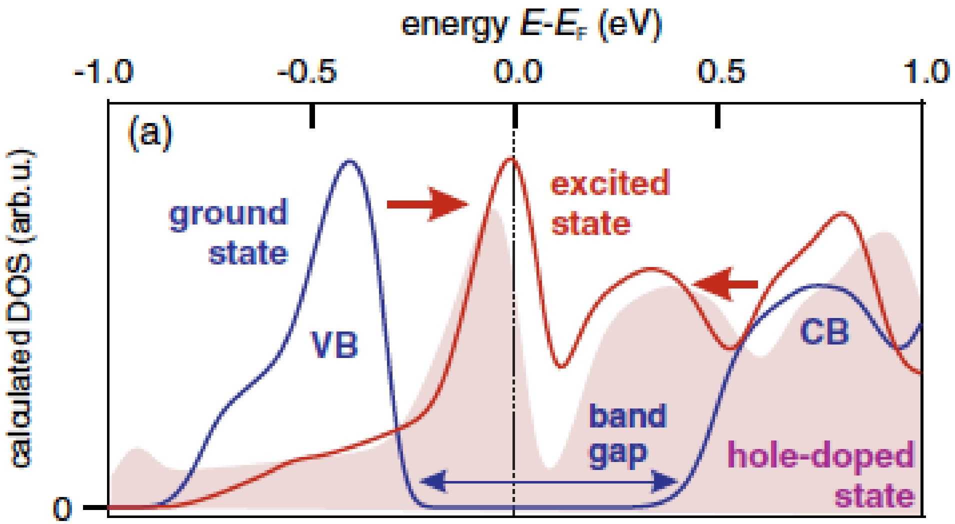
And need to pump the transition at ~ 1500 nm!

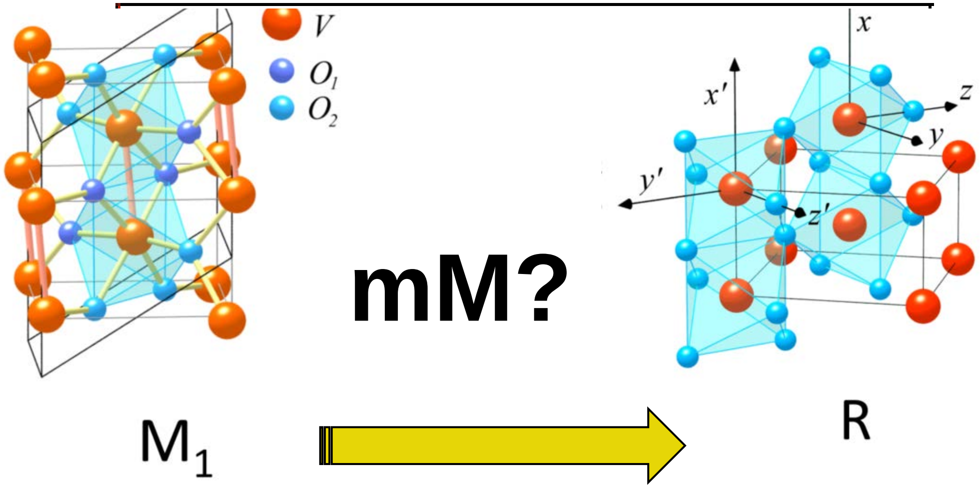
- Virtually all experiments up to now pump at ~ 2·E_{gap}
- Deposits excess energy into phonons, drives phase transition to rutile state
- Differential transition and threshold measurements show optimum pumping wavelengths from 1000 to 1500 nm!
- Pump-probe experiments underway at Vanderbilt …



Rini, Cavalleri *et al.*, *Appl Phys Lett* **92**, 181904

And then there is the mysterious mM phase …



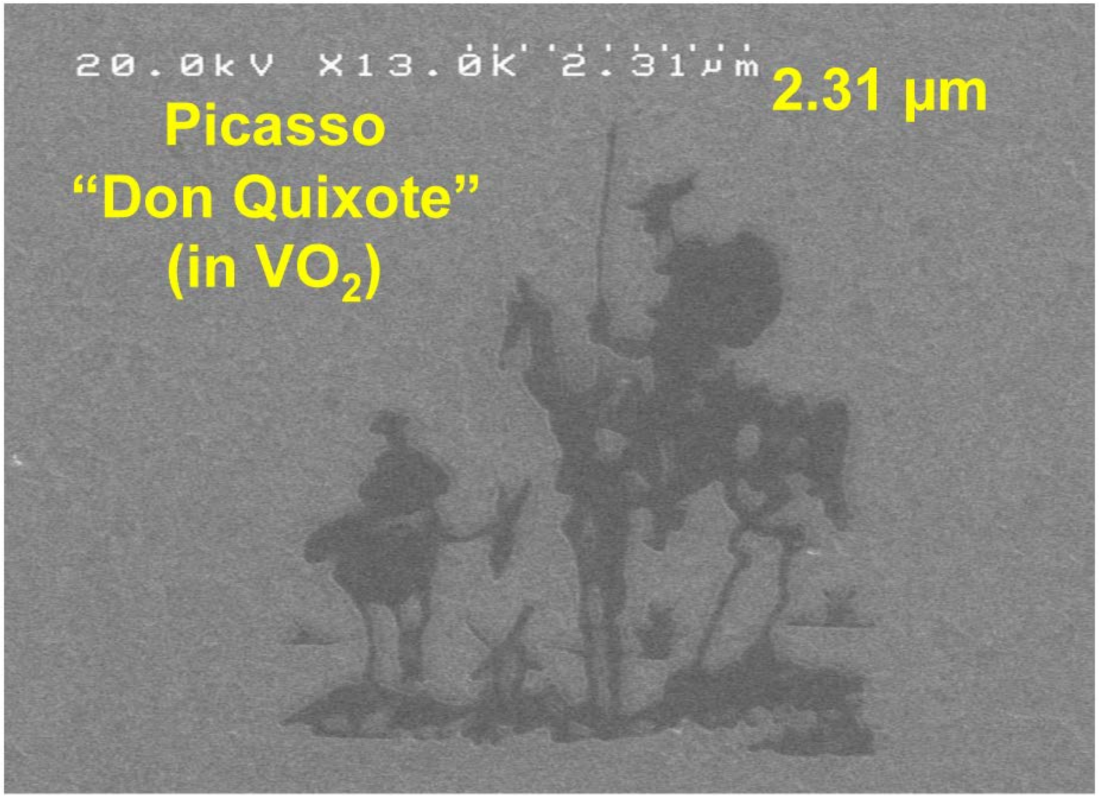


- Metallic monoclinic (mM) phase confirmed in 2014
- Seen in photoelectron spectroscopy, ultrafast electron diffraction and strain-stabilized films
- No significant structural information available as of today …
- ... so stay tuned!

Quo vadis?

- VO_2 is a model phase-changing material for optical control of plasmonic and photonic devices
  - Low-power, ultrafast switching is required for applications
  - Designed nanoscale morphology may be a key enabler
  - Time-dependent dielectric function is known unknown!
  - Better thin-film synthesis is critical to making real devices
- Ultrafast switching is a very real possibility ... Especially in the near infrared where “off” time is on ps scale
- Much interesting VO_2 physics can be probed in hybrid plasmonic nanostructures and silicon photonics devices
- Measuring electronic and structural dynamics as function of thin-film synthesis, processing, size and structural properties is crucial!

So the quest continues …



"The legitimate purpose of research can only be, to make two questions grow where there was only one before."

(Thorsten Veblen, 1927)

Thanks to funding agencie$$ and the heavy lifters!

- **Vanderbilt**
  - Krishen Appavoo (BNL)
  - Joyeeta Nag (IBM)
  - Judson Ryckman (Intel)
  - Bin Wang (Okla State)
  - Sokrates Pantelides
- **Alabama-Birmingham**
  - Nathaniel Brady
  - David Hilton
- **Los Alamos-Sandia**
  - Rohit Prasankumar
  - Mina Seo
- **FHI-Berlin**
  - Daniel Wegkamp
  - Simon Wall (IFCO)
  - Julia Stähler
  - Martin Wolf
- **Konstanz**
  - Bernhard Meyer
  - Alexander Grupp
  - Alexej Pashkin
  - Alfred Leitenstorfer
- **Regensburg**
  - Huber, Plankl, Eisele …
  - Tyler Cocker
  - Rupert Huber

Alexander von Humboldt
Stiftung/Foundation