ON-CHIP RAMAN SPECTROSCOPY: BACKGROUND CHALLENGES AND THEIR MITIGATION

Kristof Reynkens

PhD defence 05/10/21

GHENT UNIVERSITY

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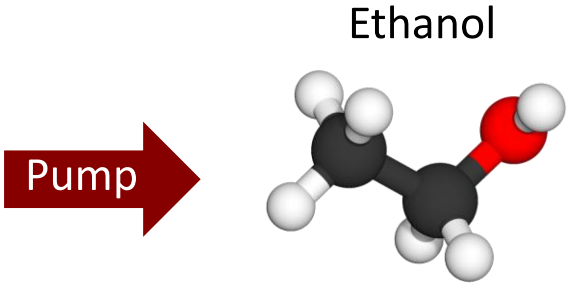
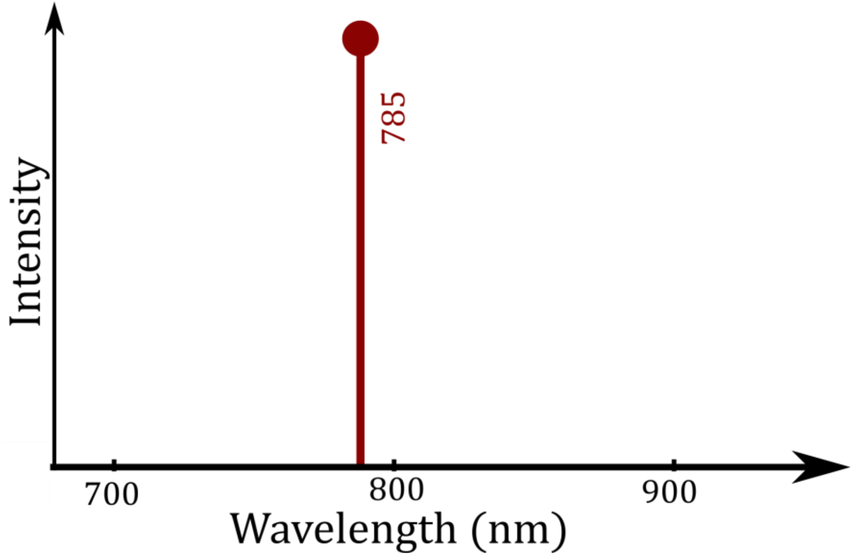
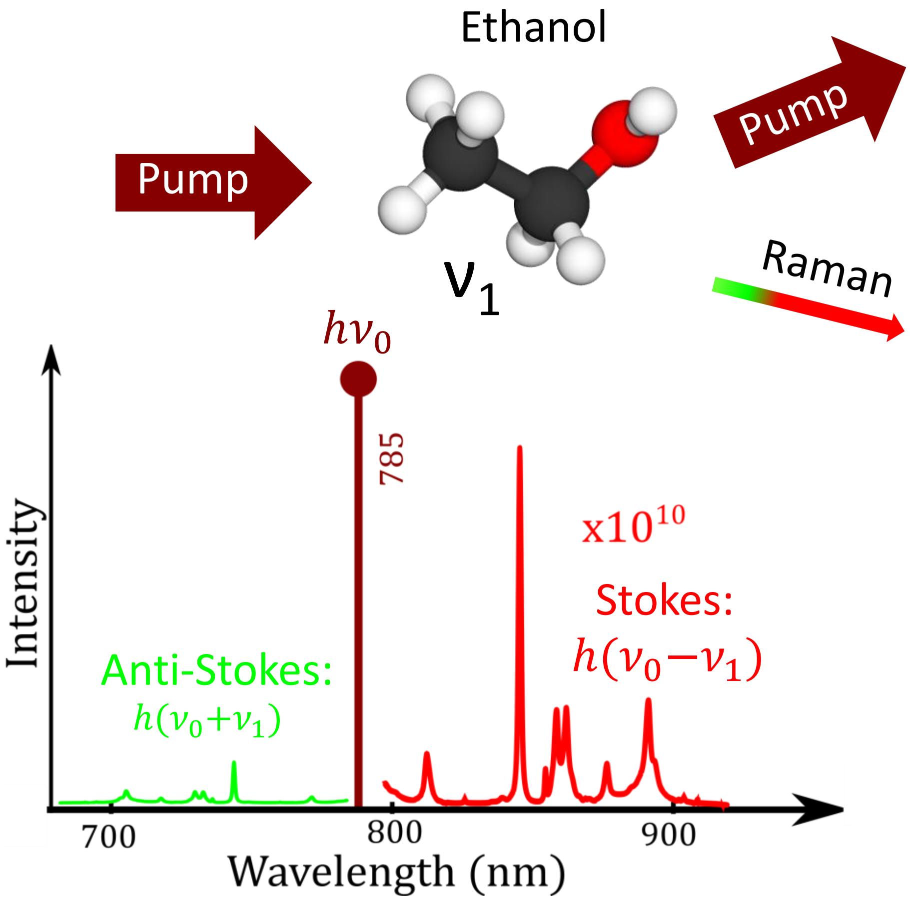
ON-CHIP RAMAN SPECTROSCOPY



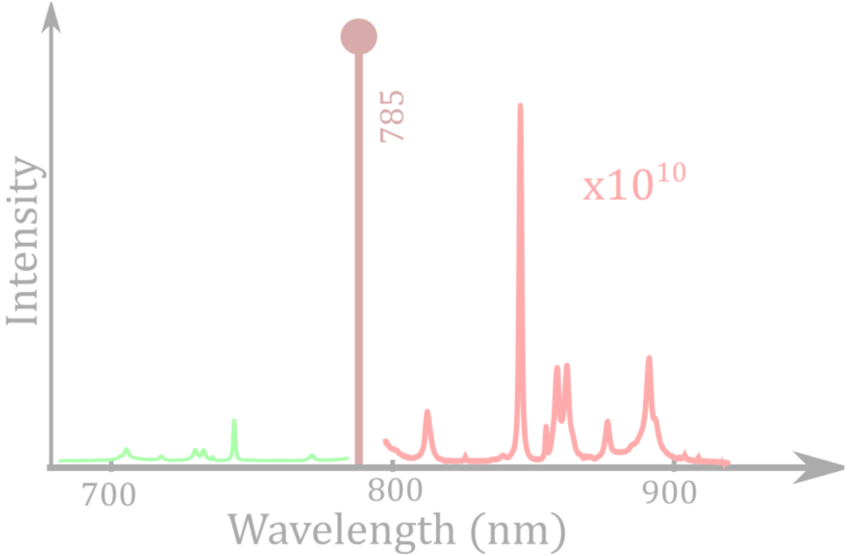


On-chip Raman spectroscopy



C.V. Raman

ON-CHIP RAMAN SPECTROSCOPY



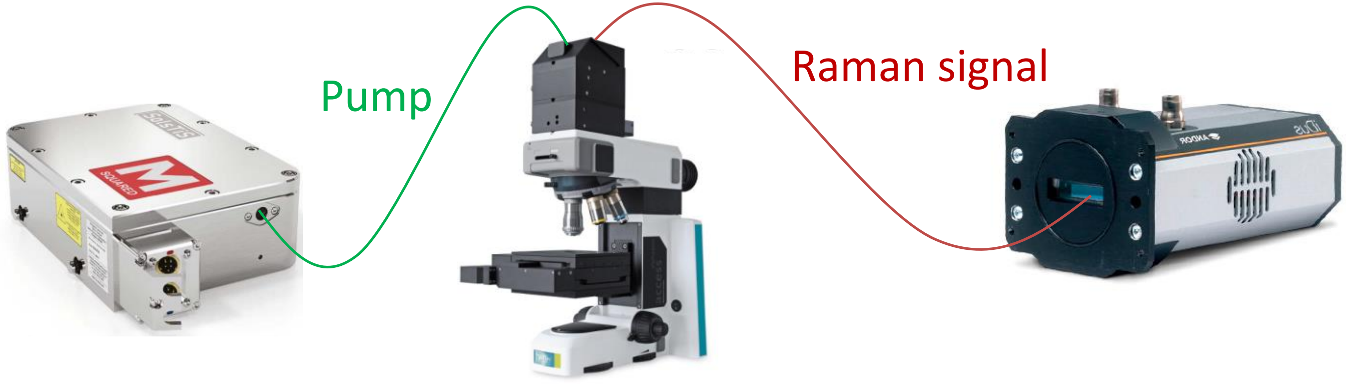
- ✓ Non-invasive, label-free
- ✓ Distinctive molecular fingerprint (multiple analytes)

- ✗ Inherently weak

  ➔Lab environment

RAMAN SPECTROSCOPIC SYSTEM IN THE LAB

$1\ \mathrm{m}^3$ | 100 000 €

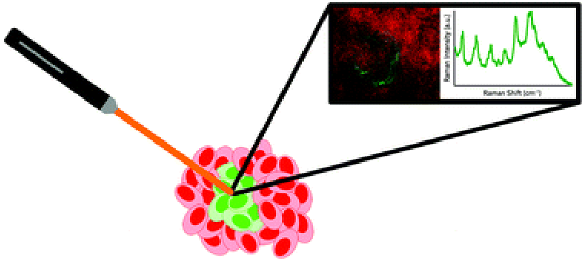


Pump

Raman signal

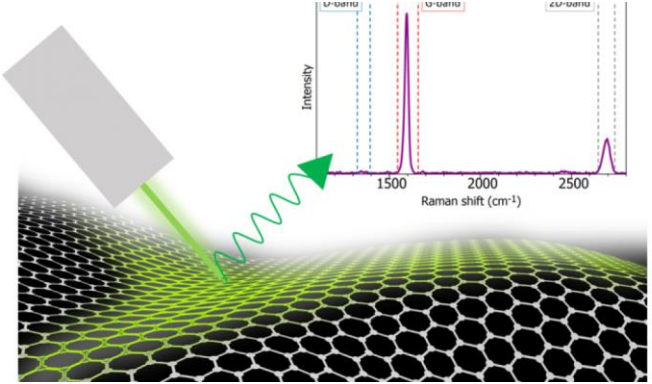
NON-DESTRUCTIVE MATERIAL ANALYSIS, BUT …

Cancer diagnostics:



Austin et al, 2016

Semiconductor R&D:



Flack et al, 2021

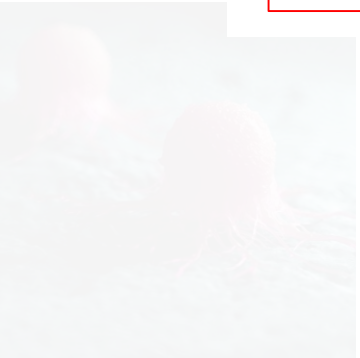
Pigment analysis:

Philip IV (*Felipe IV*) by Velázquez (1623)

Gutiérrez-Neira et al, 2013

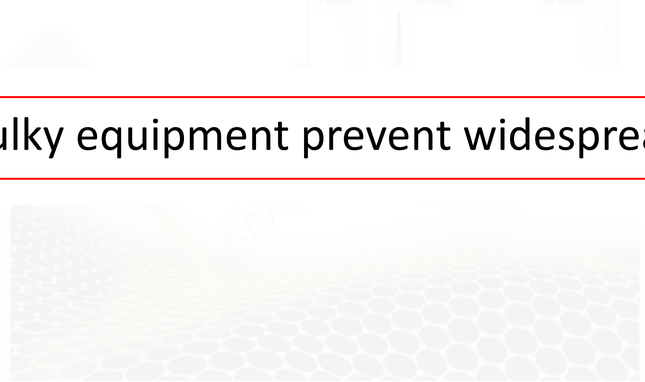
NON-DESTRUCTIVE MATERIAL ANALYSIS, BUT …

Cancer diagnostics:



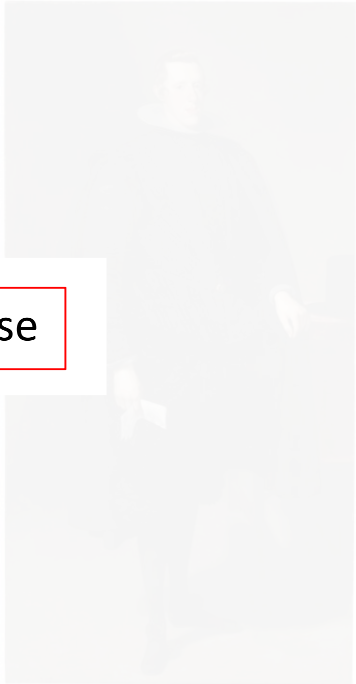
Austin LA et al, 2016

Semiconductor R&D:



Flack A et al, 2021

Pigment analysis:

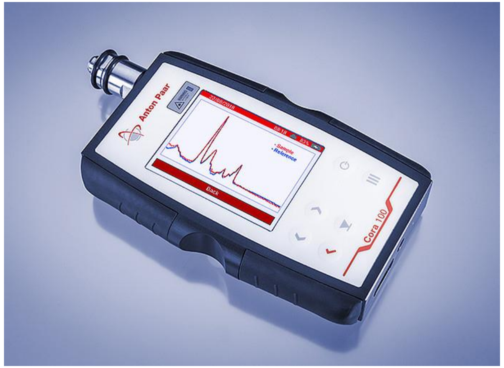


Philip IV (*Felipe IV*) by Velázquez (1623)

Gutiérrez-Neira et al, 2013

Costly and bulky equipment prevent widespread use

HANDHELD RAMAN DEVICES: ON-SITE APPLICATIONS



- ✓ Rapid, on-site
- ✗ Reduced performance
  - ➔ Complex samples e.g. blood
- ✗ Rather costly

Material ID

NanoRam®, 2021

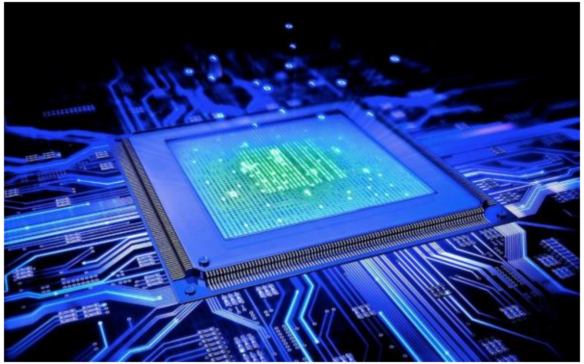
Drug detection

Kranenburg et al, 2020

Food-quality monitoring

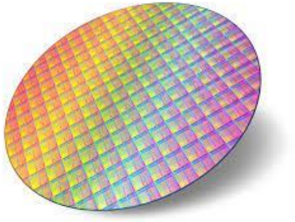
Martín-Gómez et al, 2021

ON-CHIP RAMAN SPECTROSCOPY

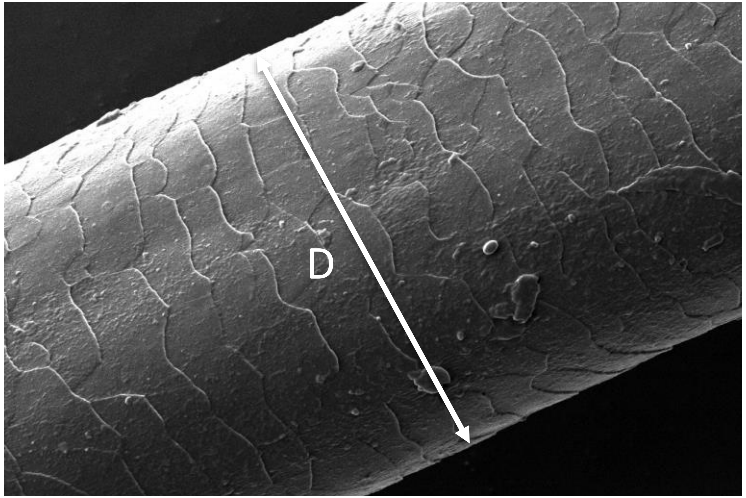


A set of electronic circuits on a small flat piece of silicon:

- Transistors (mini electrical current switches)
- Microchip: billions of transistors
- CMOS technology:
    - High Yield
    - Compact
    - Mature technology
    - Low cost in volume

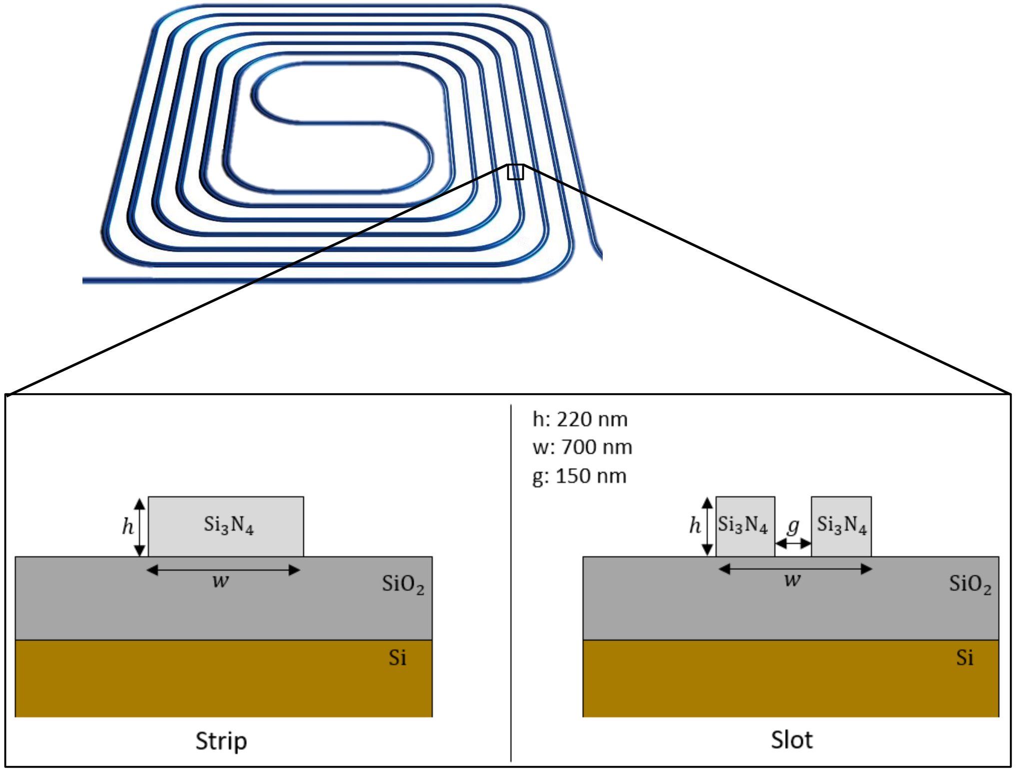


NANOTECHNOLOGY: 1 NM



Diameter (D) of a human hair: $\sim 50{,}000 - 100\ 000\ nm$

SILICON PHOTONICS: HIGH INDEX CONTRAST WAVEGUIDES



h: 220 nm
w: 700 nm
g: 150 nm

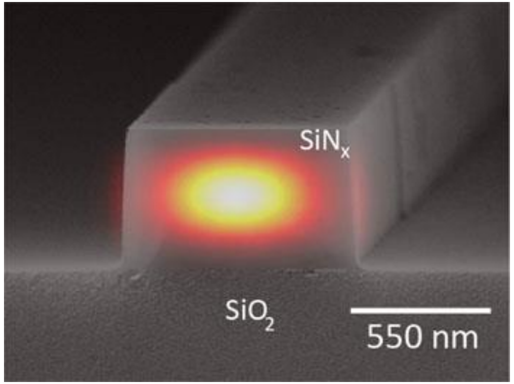


Image by P. Wuytens

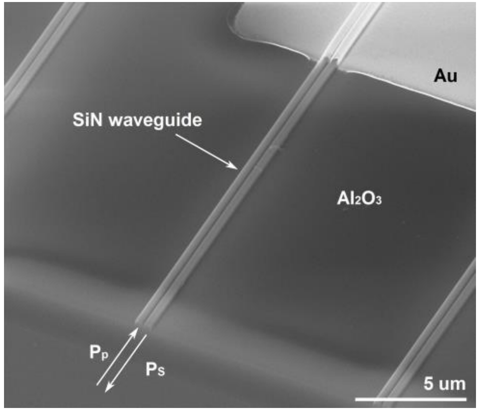
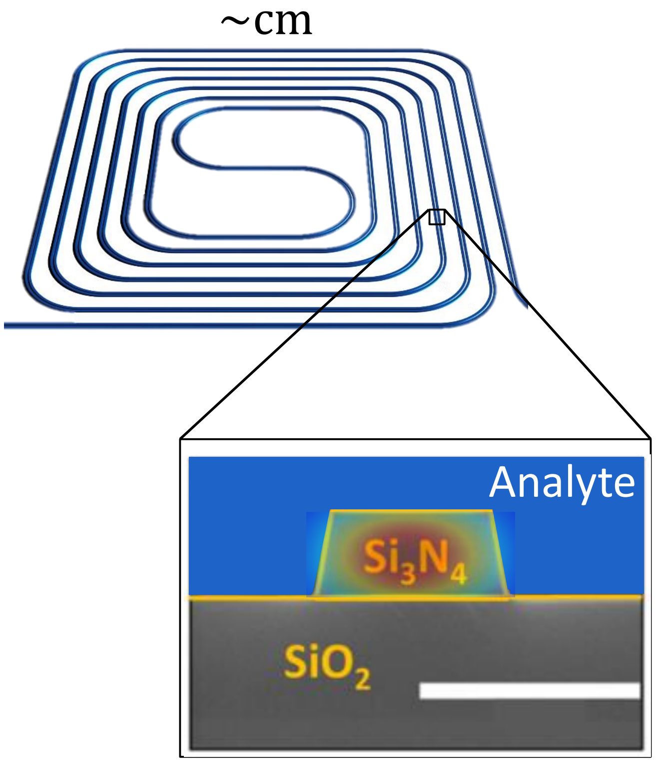
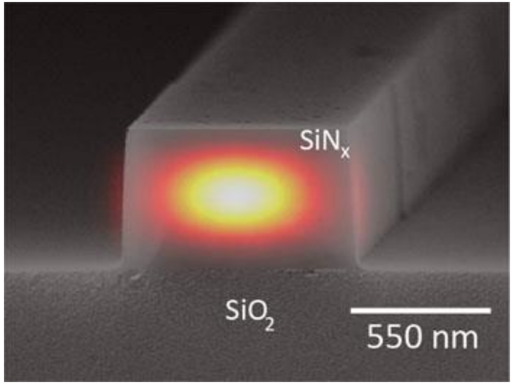


Image by N. Turk

SILICON PHOTONICS: HIGH INDEX CONTRAST WAVEGUIDES



Evanescent field enhancement & long interaction lengths



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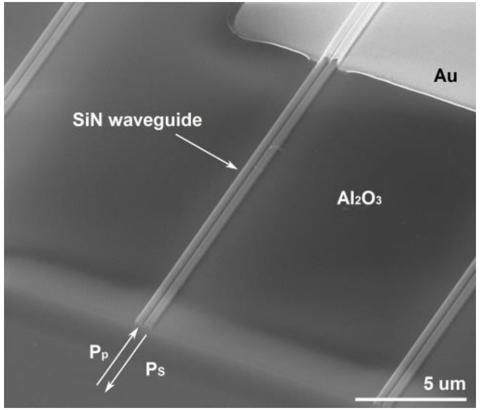
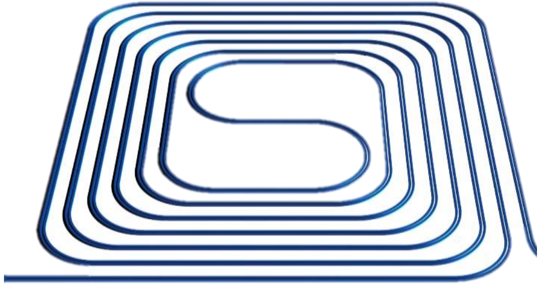
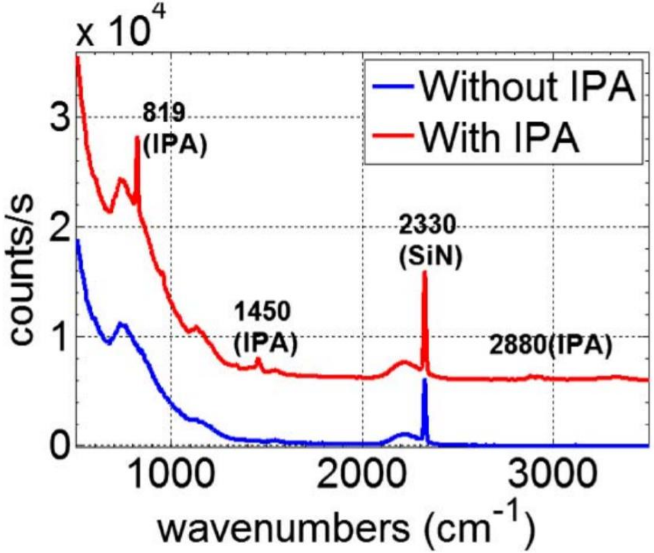


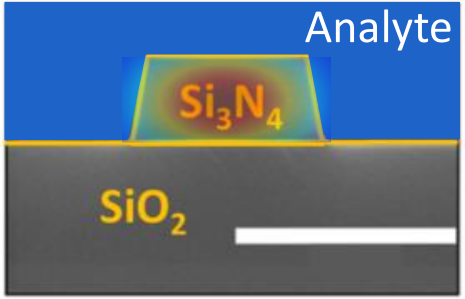
Image by N. Turk

INTEGRATED RAMAN SENSOR: NWERS

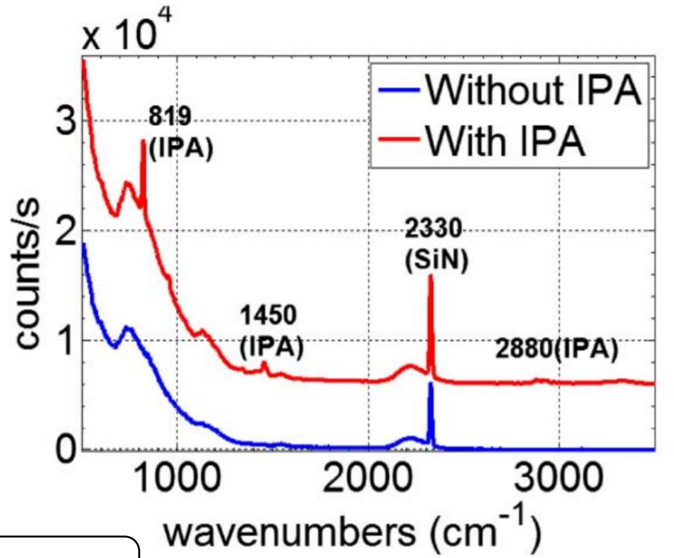




Dhakal et al., 2014



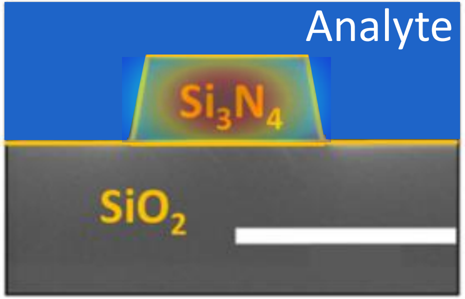
Integrated Raman sensor: NWERS



Dhakal et al., 2014

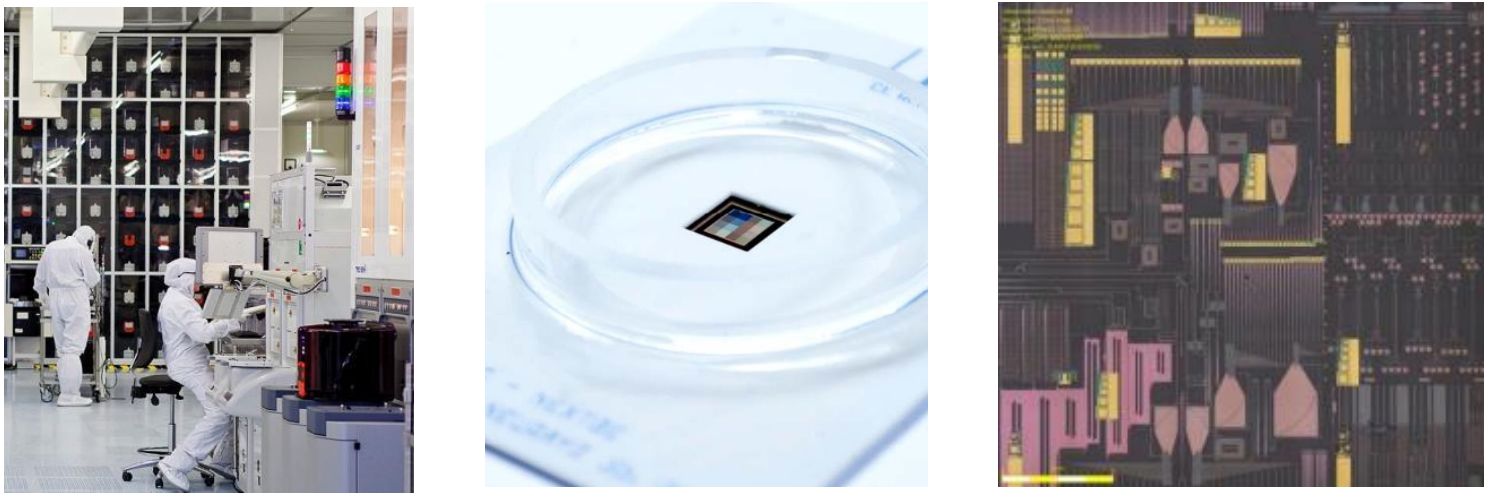
✓ Boosted Raman signal vs confocal Raman microscope (performance)

▪ Monolayers	Dhakal et al, 2016
▪ Bulk liquids	Evans et al, 2016
▪ Gases	Holmstrom et al, 2016



SILICON PHOTONICS FOR INTEGRATED RAMAN SENSING?

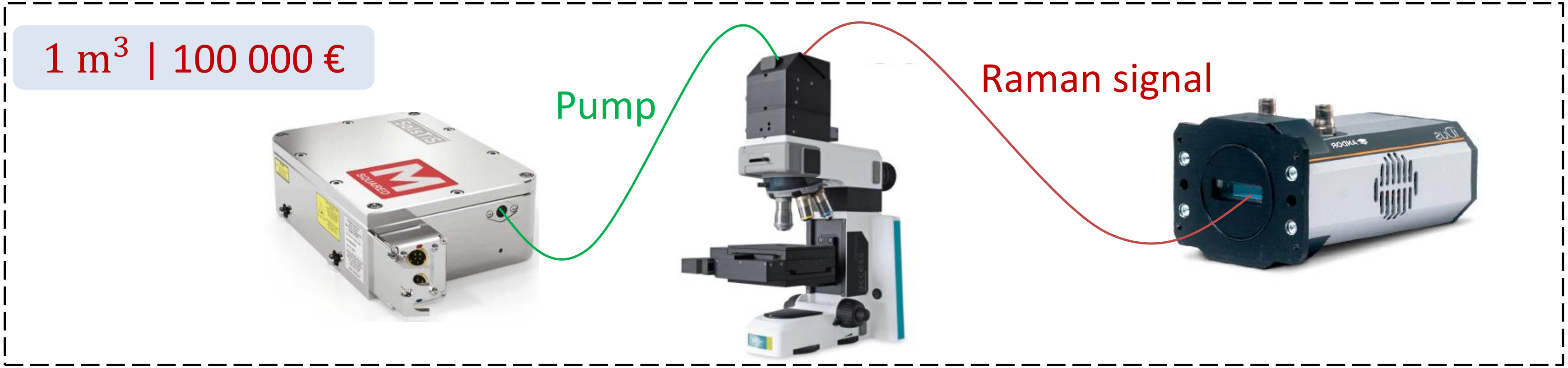
The implementation of high-performance optical components with standard semiconductor technology (CMOS) in an integrated chip



Pictures, courtesy of imec

Enabling complex photonic functionalities on a compact chip at very low cost

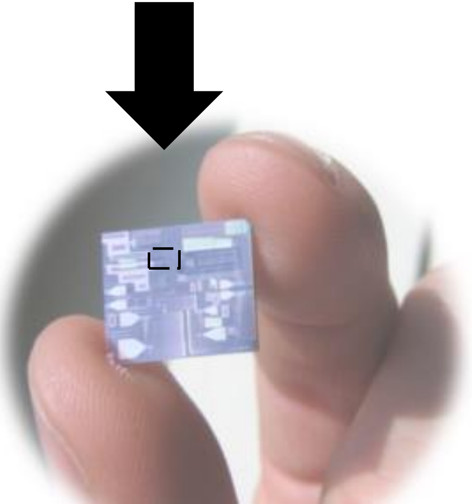
FULLY-INTEGRATED RAMAN SPECTROSCOPIC SYSTEM



1 m^3 | 100 000 €

Pump

Raman signal

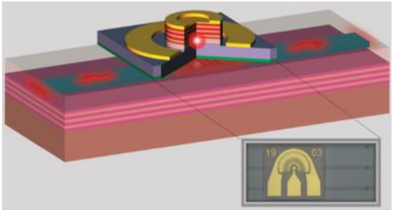


FULLY-INTEGRATED RAMAN SPECTROSCOPIC SYSTEM

1 m^3 | 100 000 €

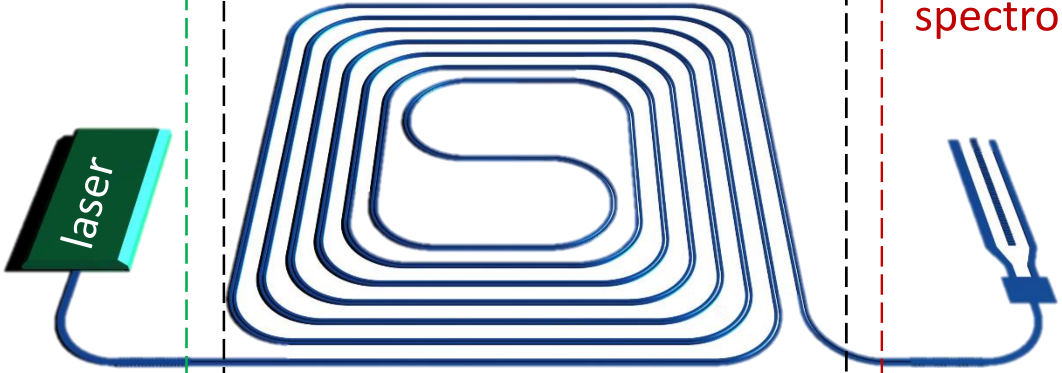
1 mm^2 | 10 €

Integrated pump laser

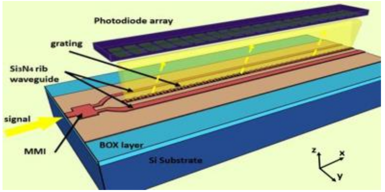


Kumari et al, LPR 2018

Integrated Raman sensor



Integrated detector and/or spectrometer



Nie et al.,OE 2017

ON-CHIP RAMAN SPECTROSCOPY: HIGH PERFORMANCE & ON-SITE

Selective detection of medically relevant molecules in a complex environment :

- In-vitro diagnostics
- Point-of-care applications
- Drug development

=> Lab-on-a-chip

Dhakal et al, 2016

Coucheron et al, 2019

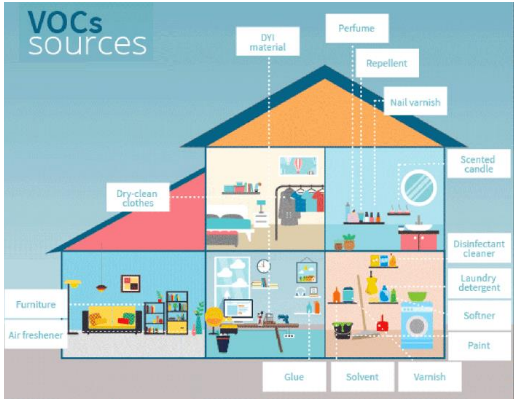
Turk et al, 2020

(Use once) biosensor at home

Bed-site (drug) monitoring

ON-CHIP RAMAN SPECTROSCOPY: ON-SITE & HIGH PERFORMANCE

Environmental-quality monitoring



Zhao et al., 2020

Detecting chemical warfare agents

Tyndall et al, 2018

SILICON PHOTONICS: REFRACTIVE INDEX SENSING

Antelope DX

“Clinical lab performance with the ease-of-use of a pregnancy test at a consumer price tag.”

REMINDER

NWERS: Nanophotonic Waveguide-Enhanced Raman Spectroscopy

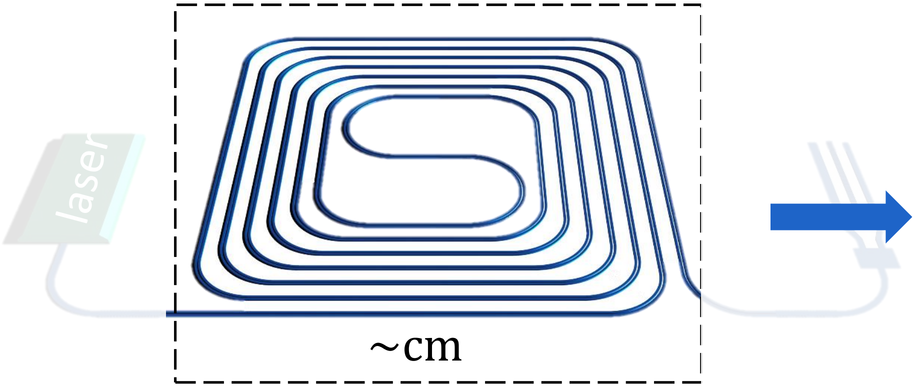
SERS: Surface-Enhanced Raman Spectroscopy

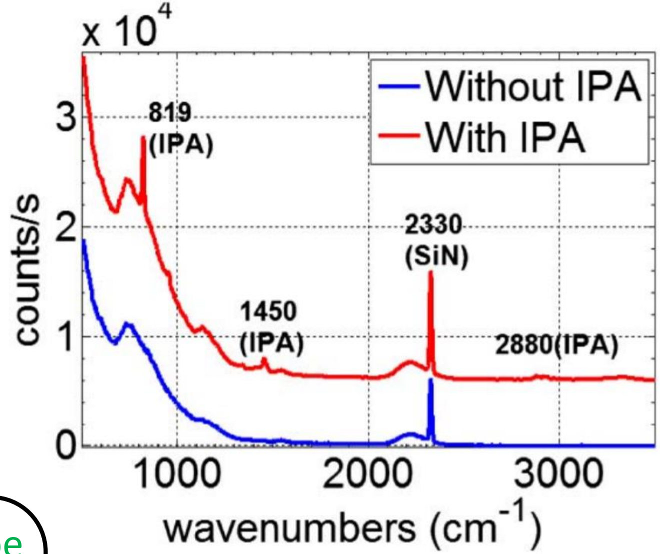
SRS: Stimulated Raman Spectroscopy

SE-SRS: Surface-Enhanced Stimulated Raman Spectroscopy

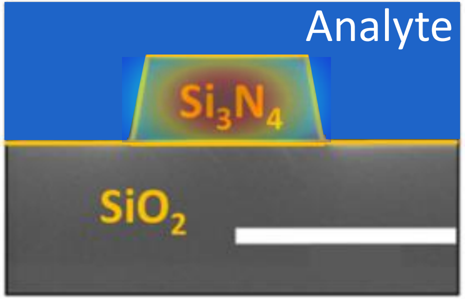
SE-CARS: Surface-Enhanced Coherent anti-Stokes Raman Spectroscopy

INTEGRATED RAMAN SENSOR: NWERS





Dhakal et al., 2014



- ✓ Boosted Raman signal vs confocal Raman microscope
- ✗ Photon background
  - ➔ Limits detection sensitivity (SBR)
- ✗ Deeply-cooled detectors
  - ➔ Difficult to integrate

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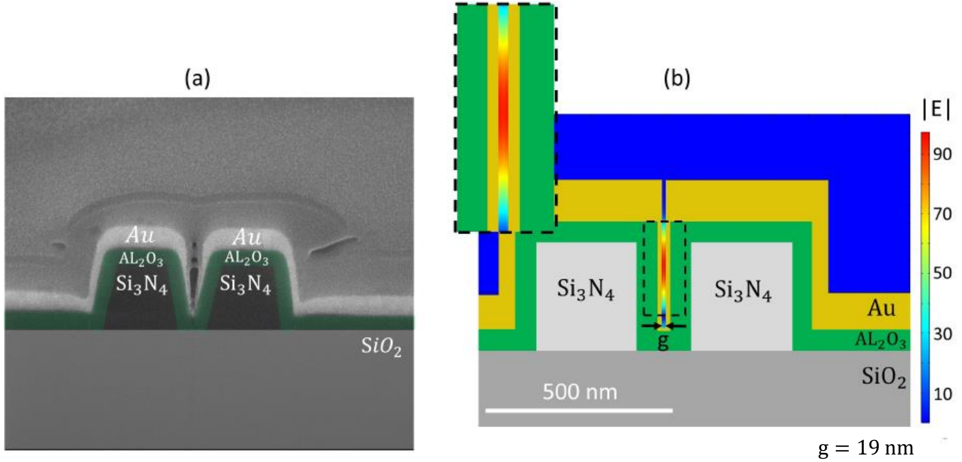
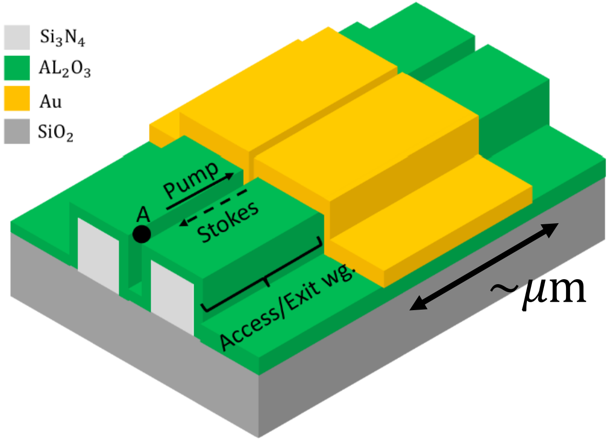
SERS ON THE NANOPLASMONIC SLOT





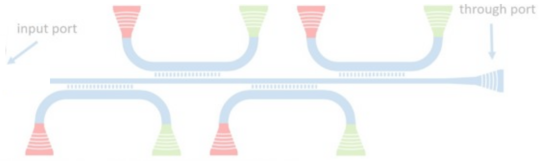
Raza et al., 2018

- ✓ Equivalent Raman signal to NWERS
- ✓ Reduced photon background $\left(\frac{Bg_{plas}}{Bg_{slot}} \sim 0{,}1\right)$

➔ Short interaction length ($\sim\mu$m vs. cm):

- ✗ Deeply-cooled detectors

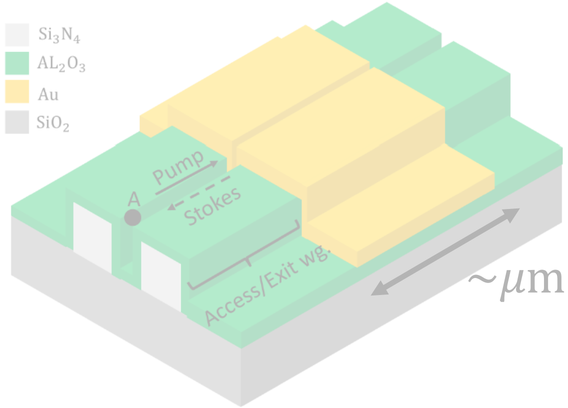
INTEGRATING THE NANOPLASMONIC SLOT



Pump rejection filter | Nie *et al., 2019*

Arrayed Waveguide Grating |*Martens et al., 2015*

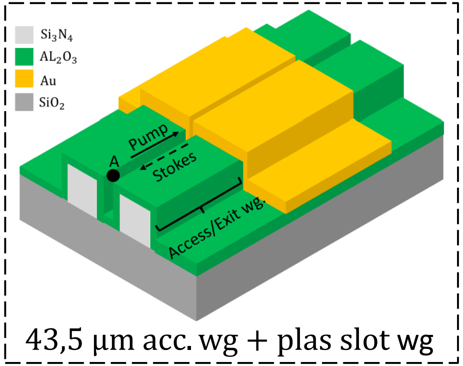
Analyzing circuit: $\sim$mm



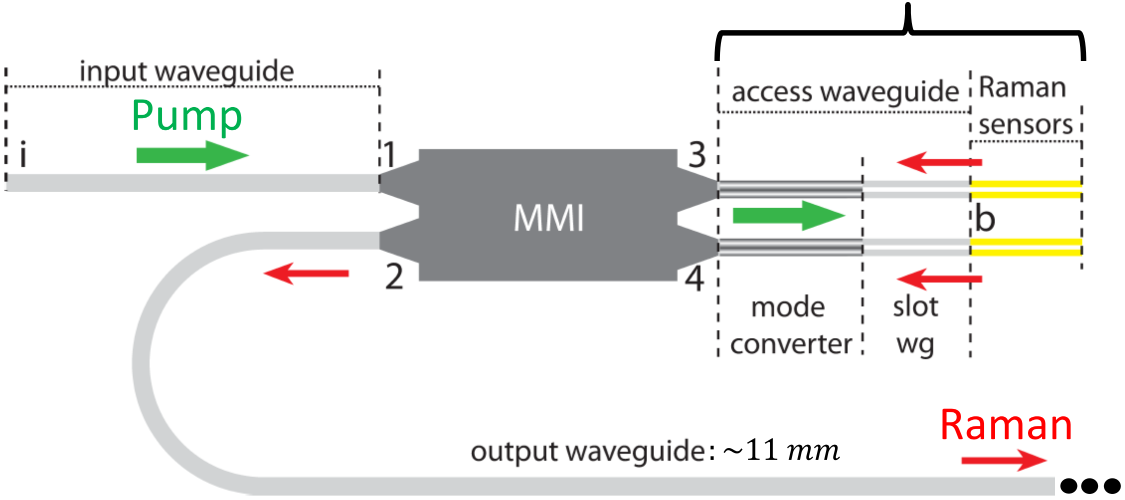
Raman sensor: $\sim\mu$m

Avoid additional background contributions of analyzing circuit !

MMI-NANOPLASMONIC SLOT CONFIGURATION

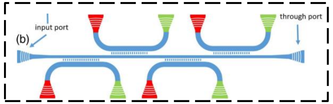


43,5 μm acc. wg + plas slot wg



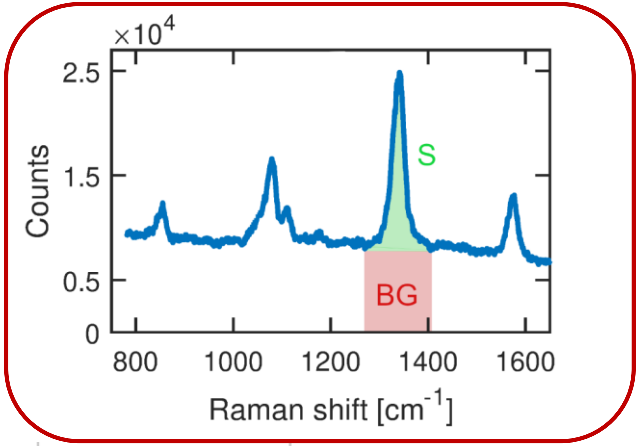
MMI
$\lambda_{central} = 830\ nm$
$BW_{3dB} = 220\ nm$
W x L= 8,9 x 112,5 μm^2

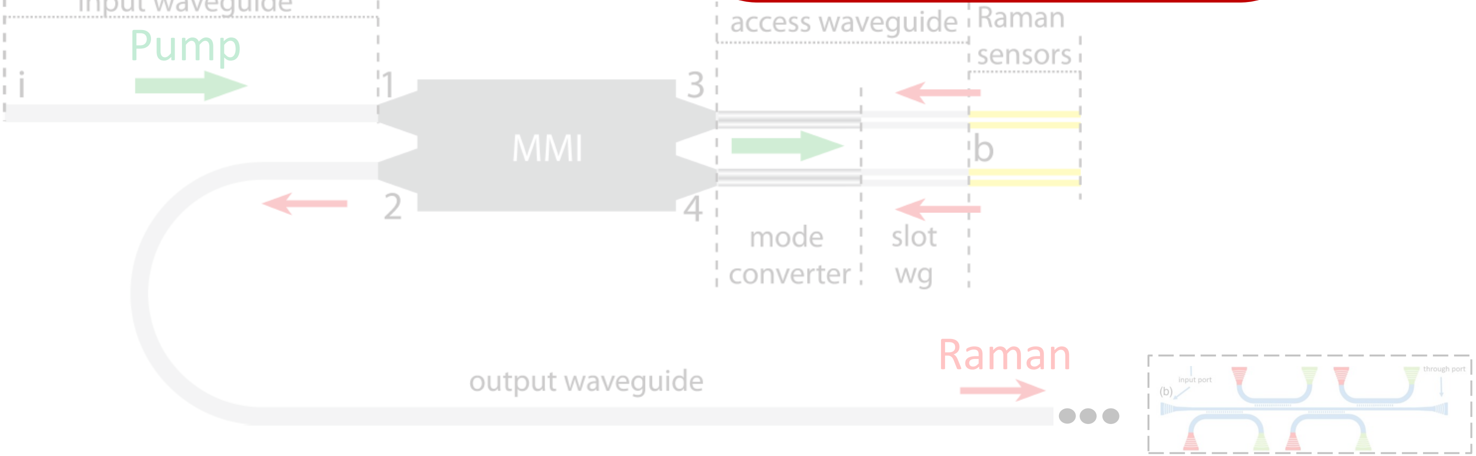
$\lambda_{pump} = 785\ nm$



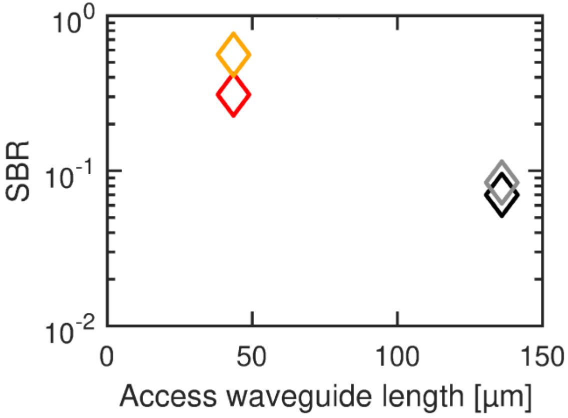
MMI-NANOPLASMONIC SLOT CONFIGURATION

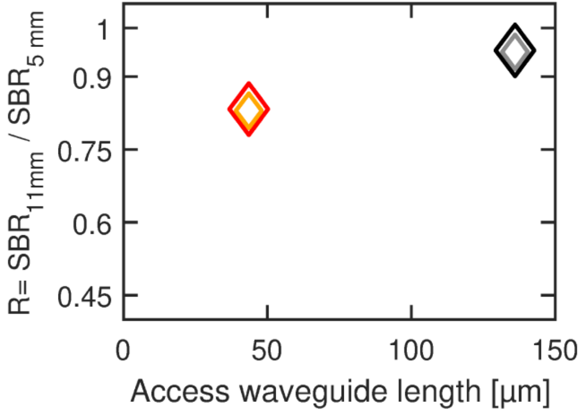
- Raman signal efficiently captured

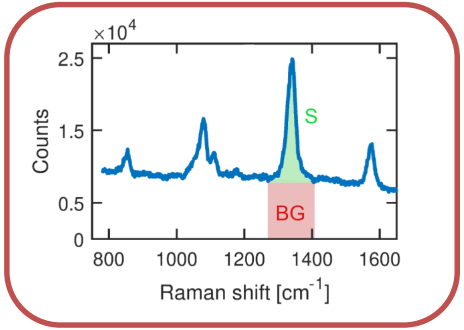


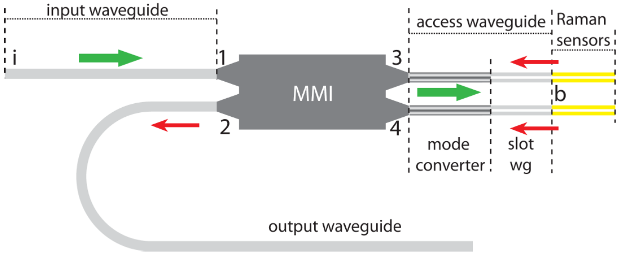


ORIGIN OF THE EXTRA BACKGROUND



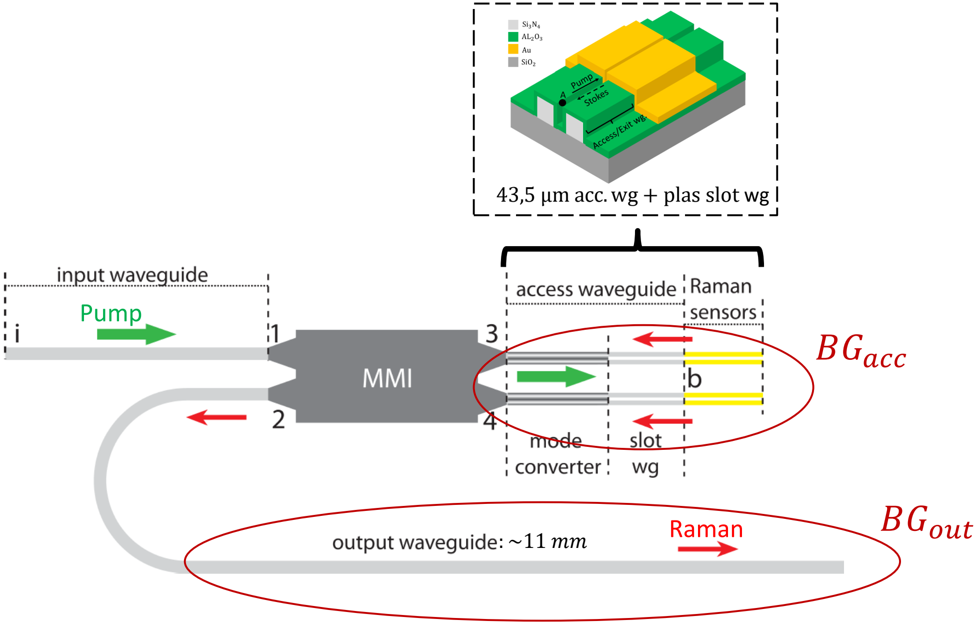






ORIGIN OF THE EXTRA BACKGROUND

- Raman signal efficiently captured
- Equivalent BG contribution output WG and SERS sensor (MMI no strong BG contribution)



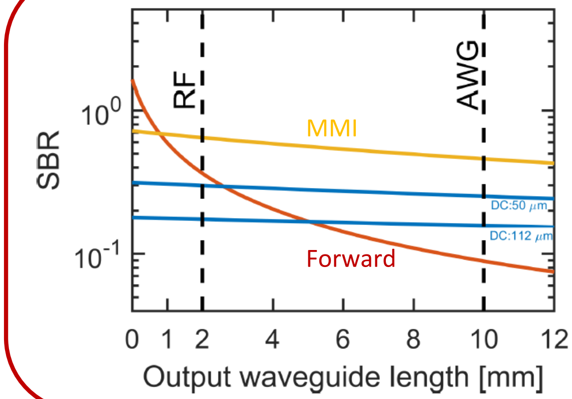
$R_{\text{plas}} = -27\ \text{dB}$

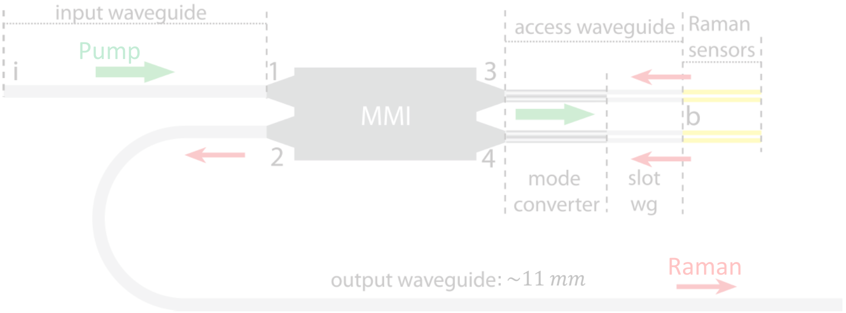
$$\Rightarrow \frac{\text{BG}_{\text{out}}}{\text{L}_{\text{out}}} = \frac{1}{173}\,\frac{\text{BG}_{\text{acc}}}{\text{L}_{\text{acc}}}$$

PERFORMANCE VS. OTHER CONFIGURATIONS

- Raman signal efficiently captured
- Equivalent BG contribution output WG and SERS sensor (MMI no strong BG contribution)
- MMI-configuration performs better than DC- and forward-configuration

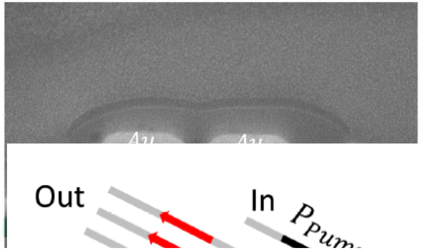
  => Most (incoherent) BG photons not efficiently collected

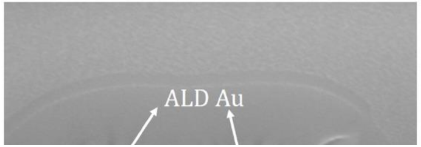


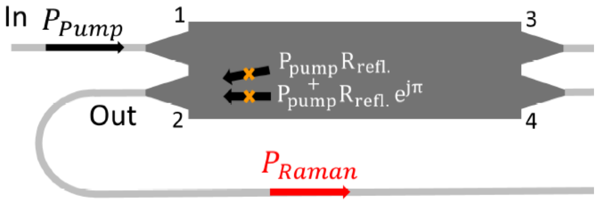


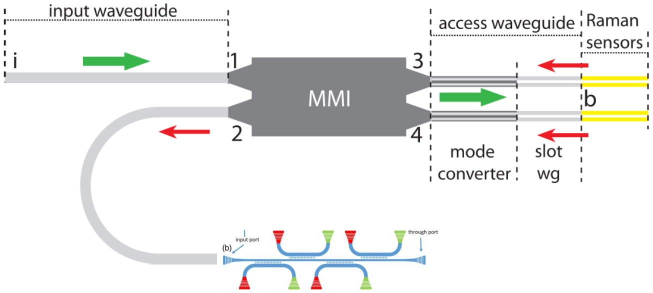
OUTLOOK

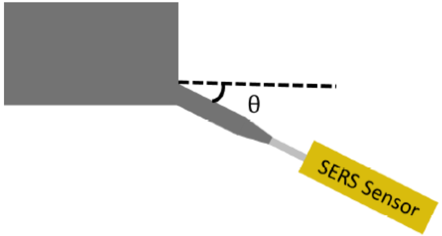
- Better control the plas. slot reflection by using ALD gold
- Engineer the MMI such that it acts as a wavelength division multiplexer
- Combine the MMI with a RF or LF on th





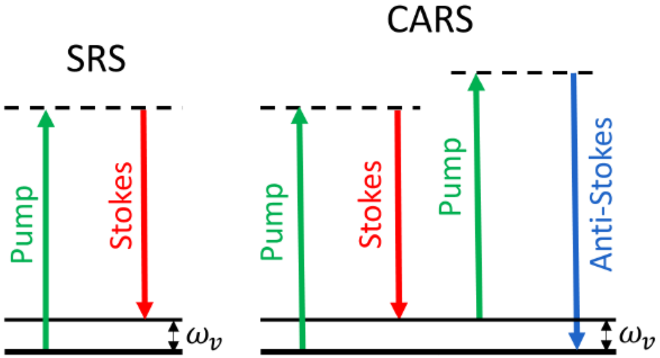


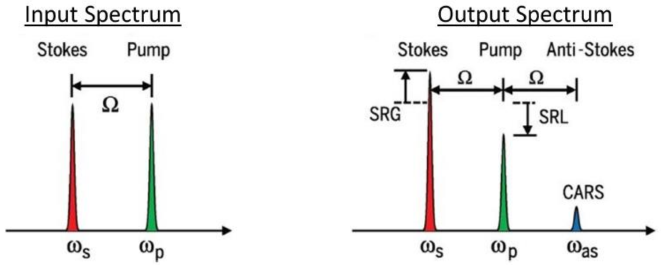


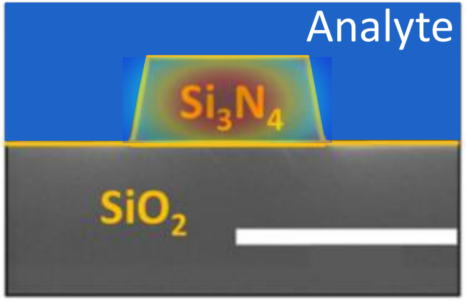


GET RID OF DEEPLY-COOLED DETECTION?

SRS on the dielectric (SiN) strip waveguide:



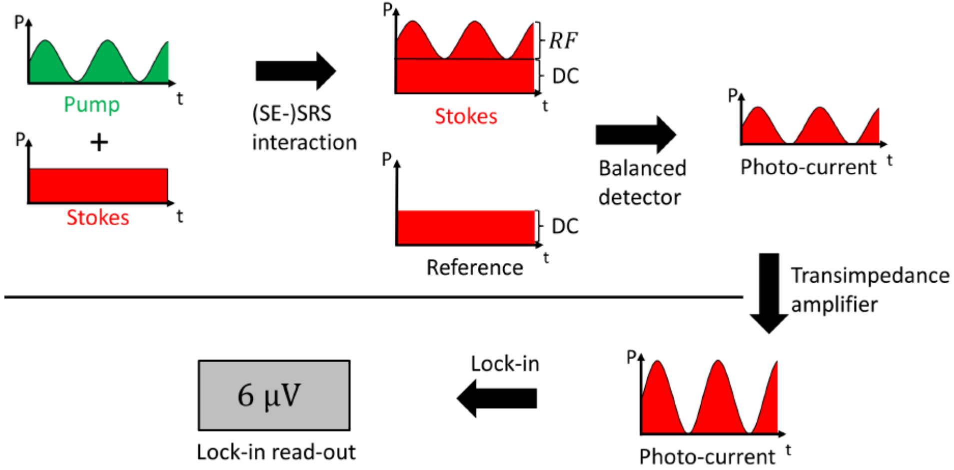


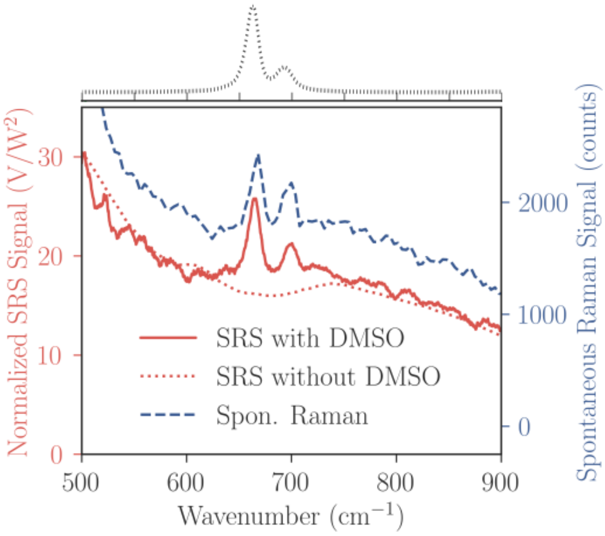


GET RID OF DEEPLY-COOLED DETECTION?

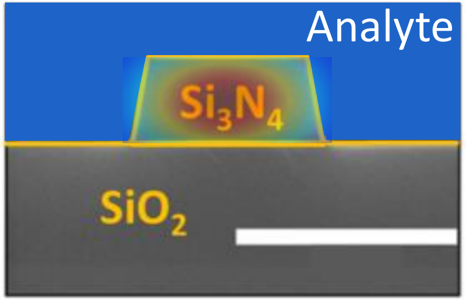
SRS on the dielectric (SiN) strip waveguide:

- SRS signal 10^5 stronger than spontaneous signal
  - ✓ Room-temperature detection
  - \- More complex detection scheme (lock-in)
- Long interaction length (~mm)





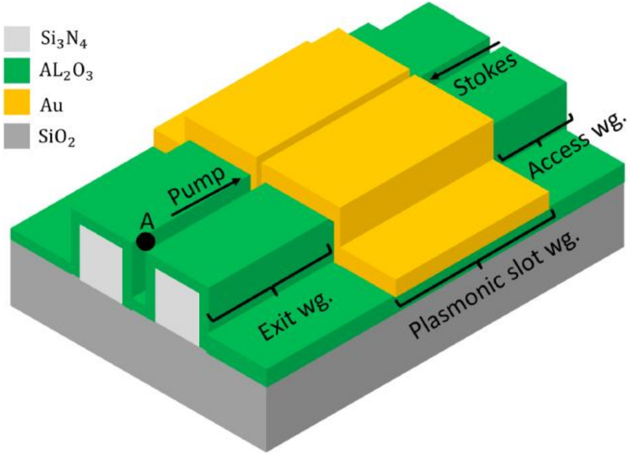
Zhao et al., 2018



GET RID OF DEEPLY-COOLED DETECTION?

SE-SRS on the plasmonic slot waveguide:

- SE-SRS signal 10^3 stronger than SERS on the plasmonic slot [NTP monolayer]
  - ✓ Room-temperature detection
  - \- More complex detection scheme (lock-in)
- SBR of SE-SRS 10^3 better than SRS [NTP monolayer]
  - ✓ Photon background



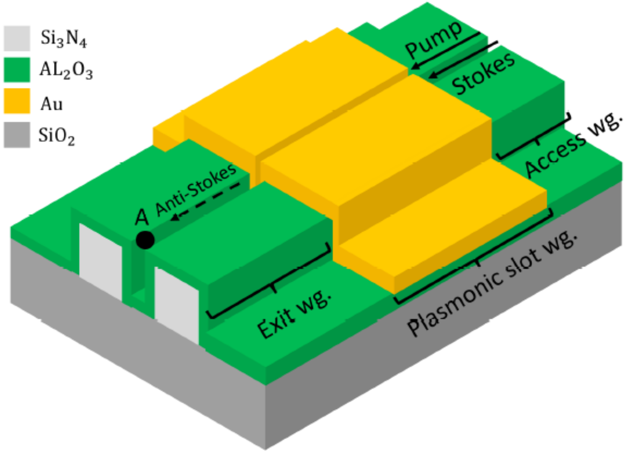
GET RID OF DEEPLY-COOLED DETECTION?

SE-CARS on the plasmonic slot waveguide:

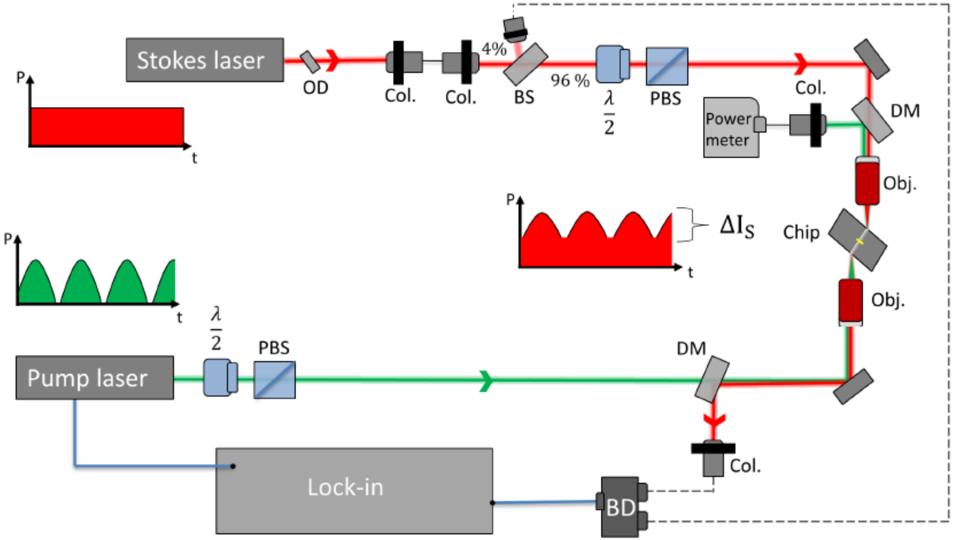
- Comparable performance as SE-SRS [NTP monolayer]
  - ✓ Filters + Single pixel detector
  - \- Pulsed laser source
- Low-concentration analytes:
  - ✗ $\frac{P_{SECARS}}{P_{SESRS}} \sim 10^{-3}$ $\left(\frac{SBR_{SECARS}}{SBR_{SESRS}} \sim 10^{-2}\right)$

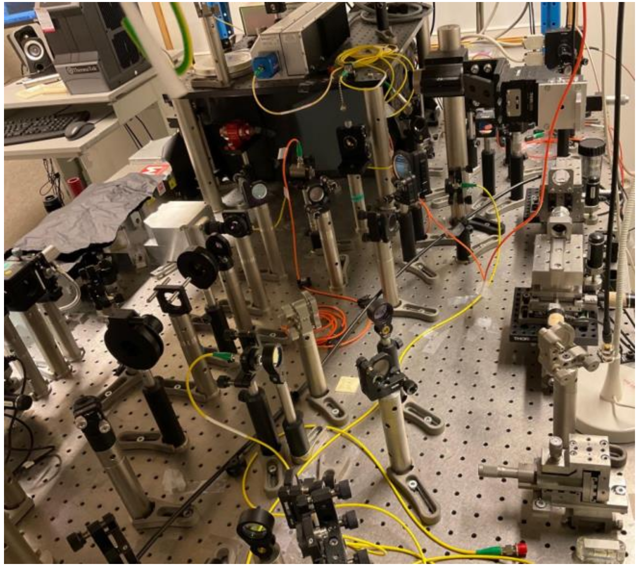
=> Quadratic dependence on conc. (vs. linear)

=> Detection of biologically relevant analytes (lab-on-a-chip)



SETUP FOR ON-CHIP SE-SRS





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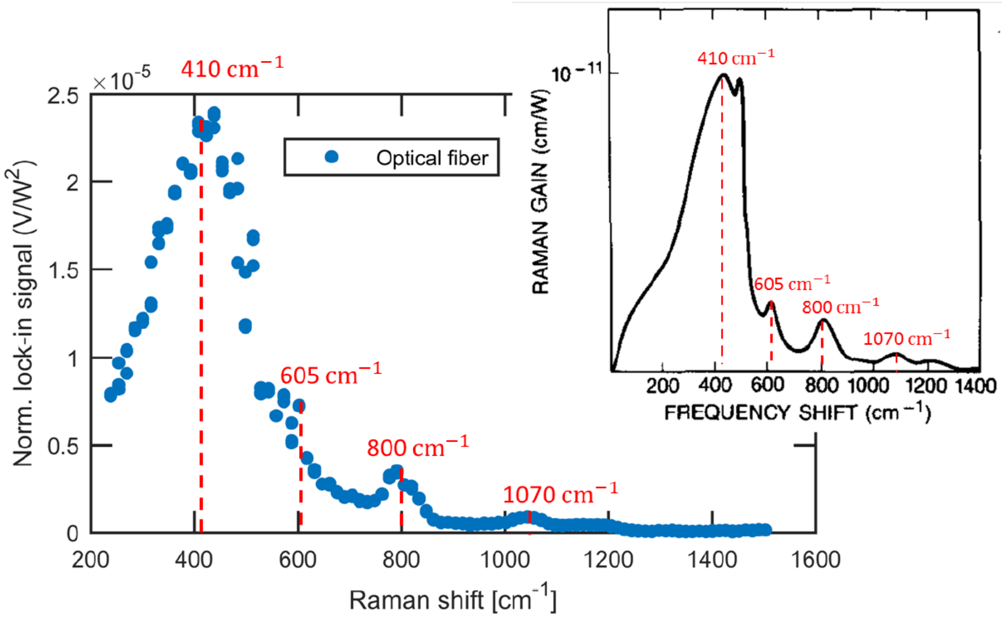
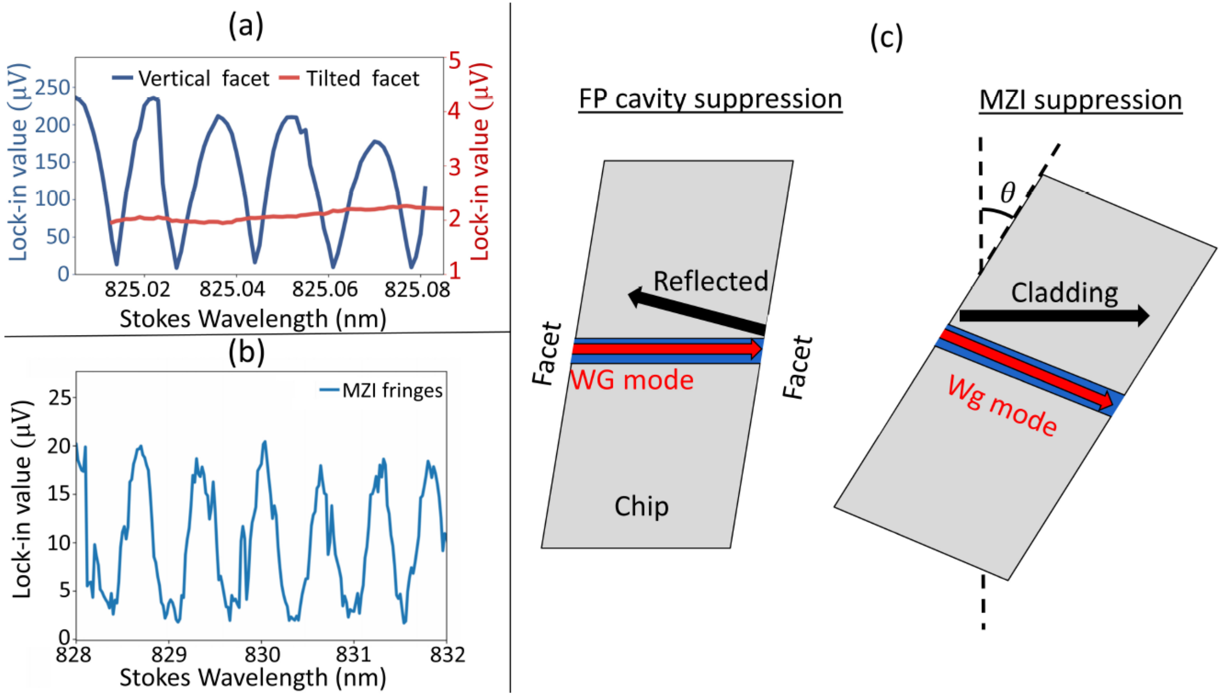
SRS ON A OPTICAL FIBER

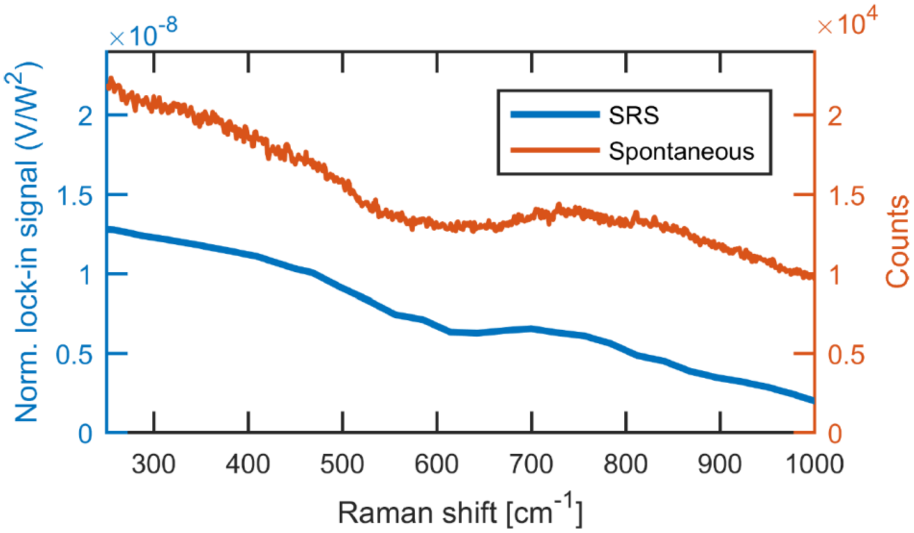


SRS on a Dielectric slot waveguide

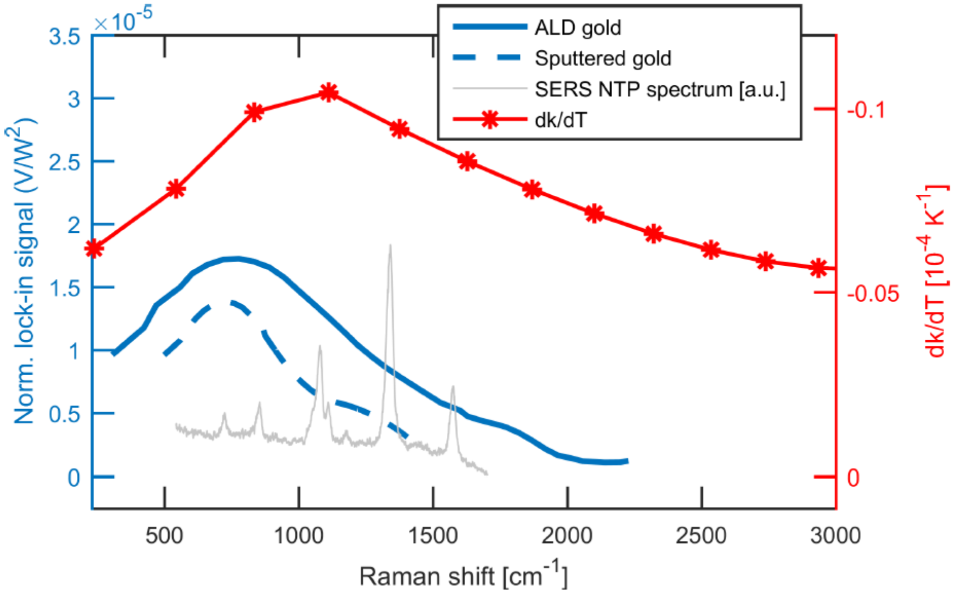
- Laser back-reflections on the chip facet (laser instability)
- Suppress cavities and stray light paths (Kerr-induced):



SRS on a Dielectric slot waveguide



CHALLENGING EXPERIMENTAL DEMONSTRATION SE-SRS



- At first unclear origin spurious SRS signal (XPM, TPA, ..)
    - MZI & FPC (plas. slot) refuted from calc.
    - No fringes on detailed meas. + same chip as dielectric slot
- Thermo-optic effect two orders of magnitude stronger than Raman response
- Overlap with spectral variation of thermo-extinction coefficient of gold (dk/dT)

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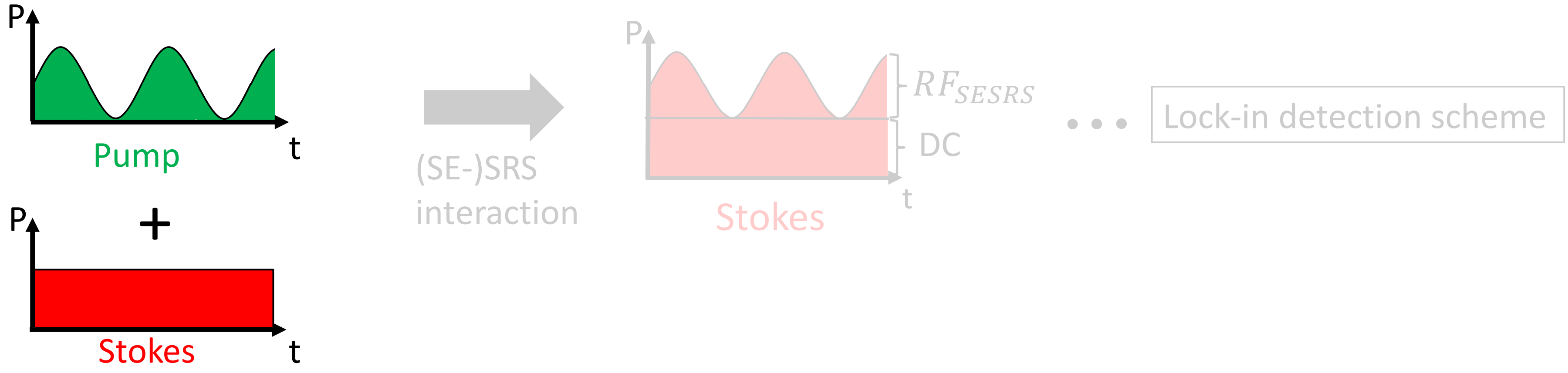


Pump

Stokes

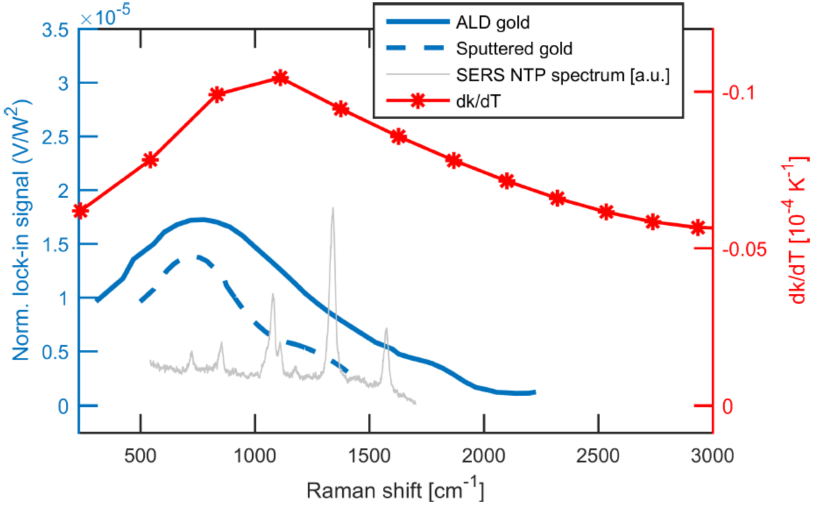
(SE-)SRS interaction

Stokes

Lock-in detection scheme

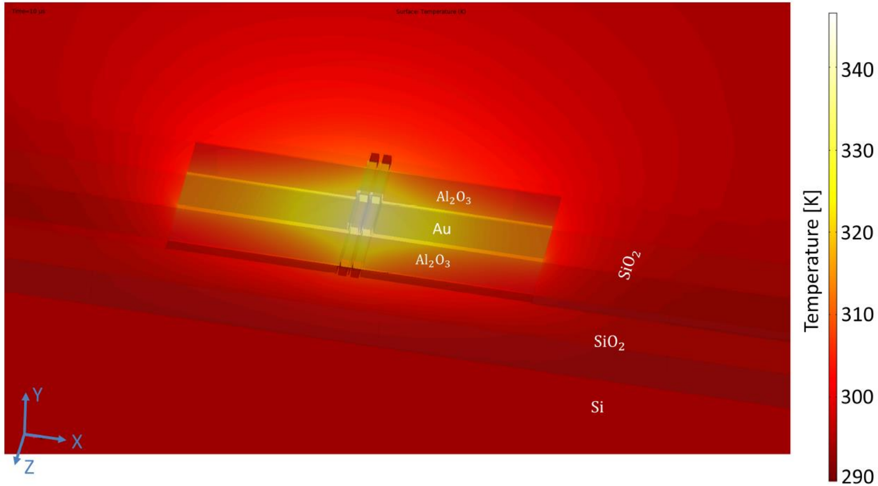
THERMAL ORIGIN OF SPURIOUS SRS SIGNAL

- Clear decline spurious signal with mod. frequency [0,15-5 MHz]
    - Slow effect such as a thermal effect
    - XPM, TPA, TA considerable faster



THERMAL ORIGIN OF SPURIOUS SRS SIGNAL

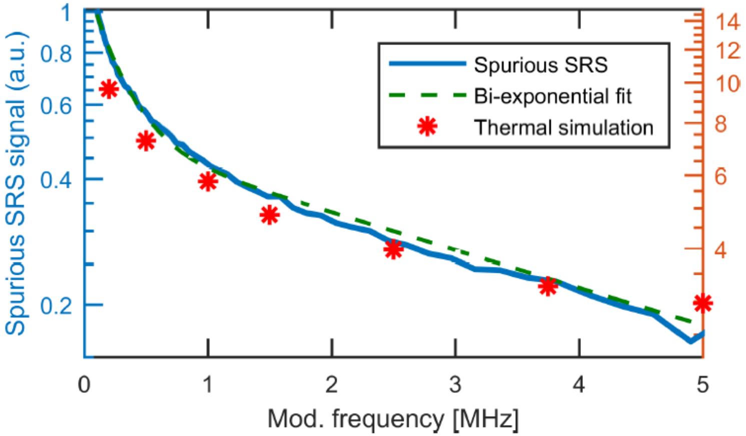
- Clear decline spurious signal with mod. frequency (slow)
- Strong spurious BG due to the dk/dT
    - Only mild temperature increase ($\Delta T_{Exp} = 2{,}5\ \mathrm{K}$) needed in plasmonic slot
    - Parasitic FPC due to dn/dT not relevant
    - ΔT_{Exp} Confirmed by 3D thermal simulations

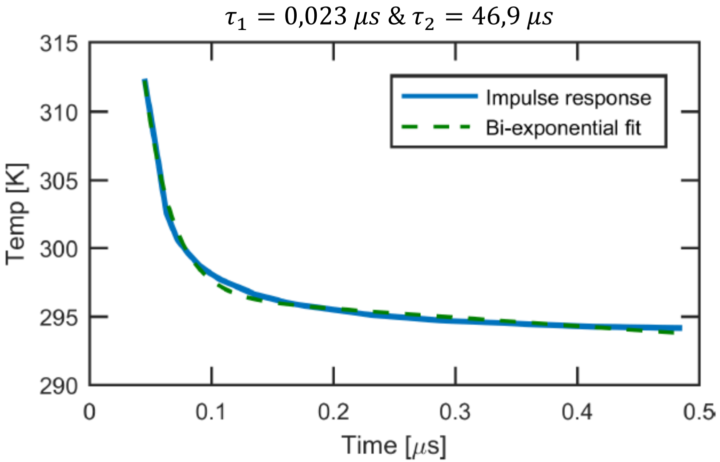


THERMAL ORIGIN OF SPURIOUS SRS SIGNAL

- Clear decline spurious signal with mod. frequency (slow)
- Strong spurious BG due to the dk/dT
- Thermal simulations recreate bi-exponential fit ($\Delta\mathrm{T}_{\mathrm{Sim}}$ in slot):

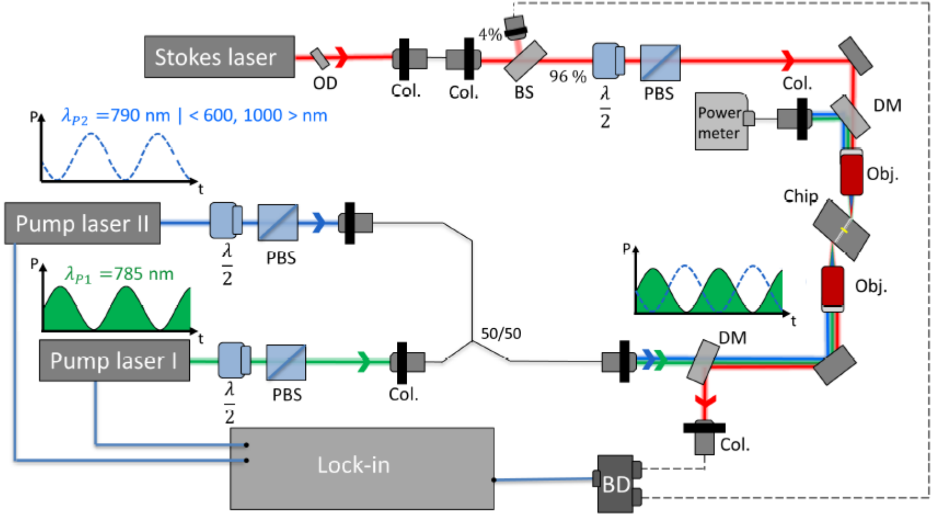
➔ Two heat phenomena: faster heat flow gold vs. surrounding materials





MITIGATION STRATEGIES:

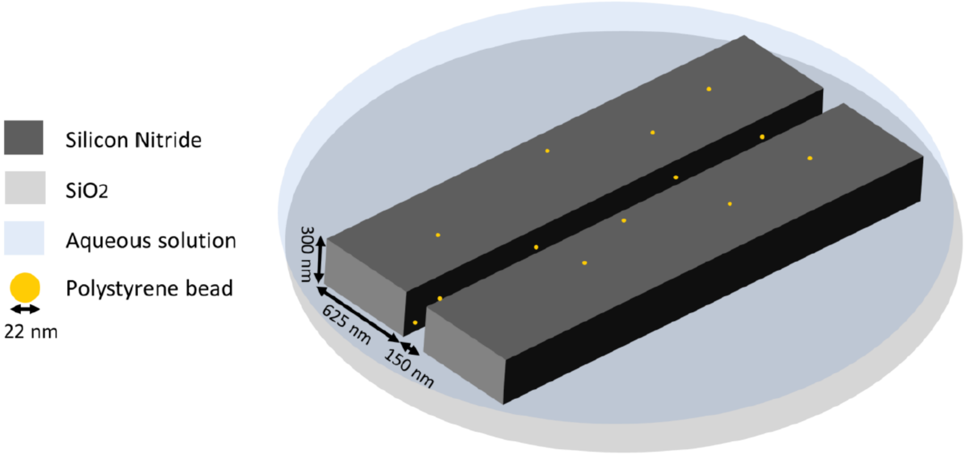
- Increase modulation frequency (100s of MHz)
- Increase heat capacity gold layer to lower thermal response
- Remove thermal modulation:



Probing acoustic vibrations of (viral) NPs using PIC

Towards an integrated virus sensor (low-frequency CARS):

- Estimated signal five orders of magnitude above noise floor
  - ✗ No observable signal
- Non-ideal particle behavior ?
  - Long list refuted hypothesis
  - Discontinued after two years



PHD CONCLUSIONS

- The MMI-plasmonic slot configuration allows for the integration of the SERS sensor with an analyzing circuit without a large BG penalty
- SE-SRS on the nanoplasmonic slot tackles the low signal generation and photon background issues of NWERS
- The spurious background present in SE-SRS measurements is of thermal origin and can be mitigated in the future
- Measuring the acoustic vibrations of (viral) nanoparticles using PICs is challenging!