



# Semiconductor metasurfaces in strong optical fields



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Department of Electrical Engineering and Computer Science  
University of California, Irvine

Sponsors and collaborators:



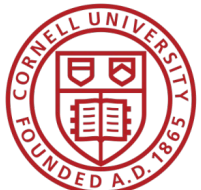
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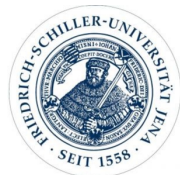
Agency for  
Science, Technology  
and Research

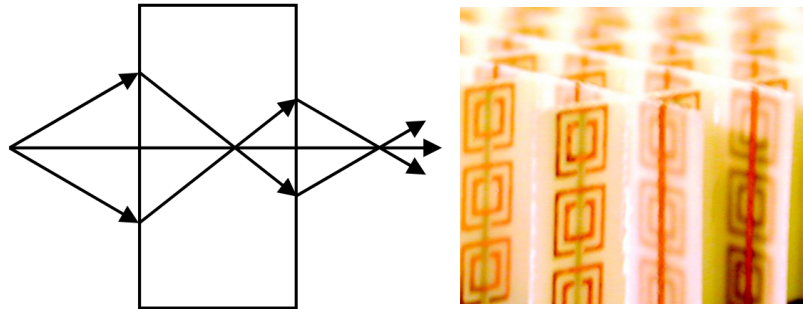


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Sandia  
National  
Laboratories

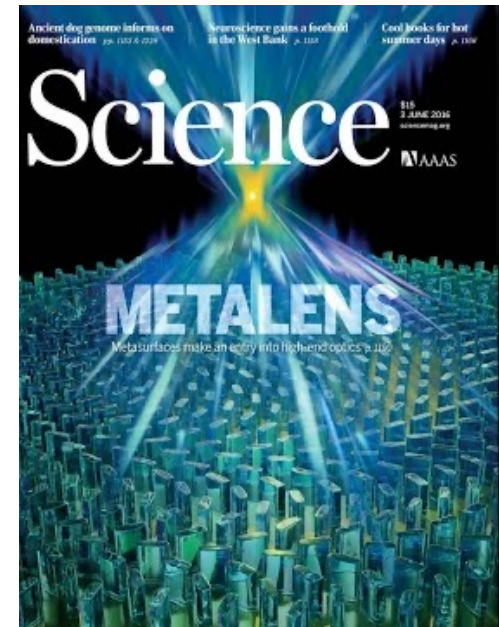
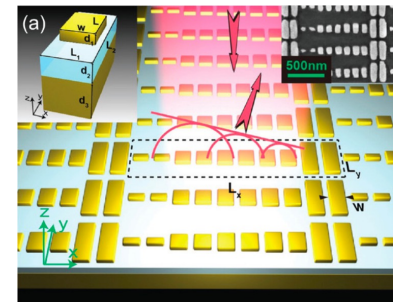
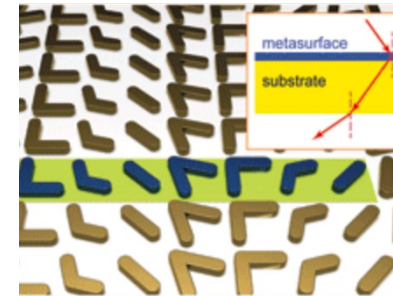
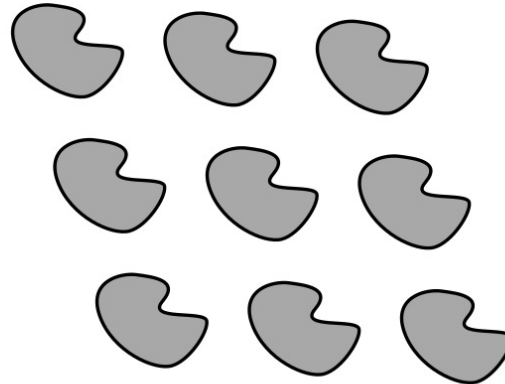




## Negative refraction and LHM

Veselago, Pendry, Shelby, Smith, Schultz, Lezec, Shalaev, X Zhang, Soukoulis, Sihvola, Tretyakov, Fan

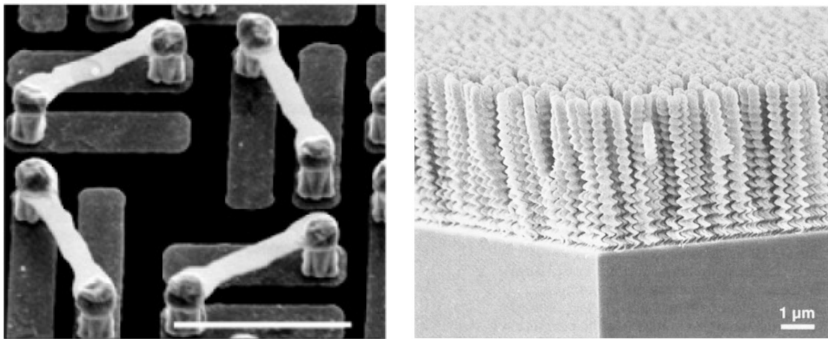
$$\mathbf{P}(\mathbf{r}) = \epsilon_0 \chi(\mathbf{r}) \mathbf{E}(\mathbf{r})$$



## Metasurface-based devices

Capasso, Alu, Yu, Kivshar, Shalaev, Boltasseva, Brongersma, Fan, Maier, Belov, Simovski, Zentgraf, Tsai, Bozhevolnyi, Neshev, Cai, Faraon, Staude, Brener, many others

$\chi(\mathbf{r})$  defines it all



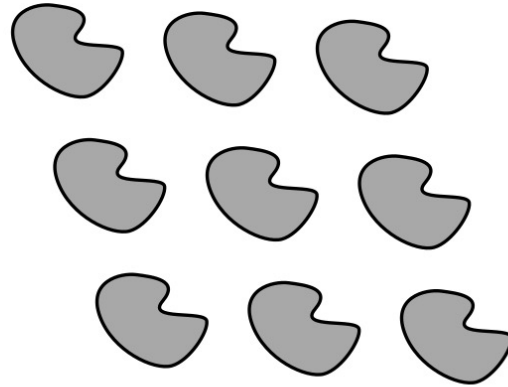
## Polarization and chirality

Pendry, Lakhtakia, X Zhang, Zheludev, He, Wegener, Pertsch, Soukoulis, Ozbay, HT Chen, S Zhang



# Metamaterials: tailored nonlinear and spatio-temporal response

$$\mathbf{P}(\mathbf{r}) = \varepsilon_0 \chi(\mathbf{r}) \mathbf{E}(\mathbf{r})$$

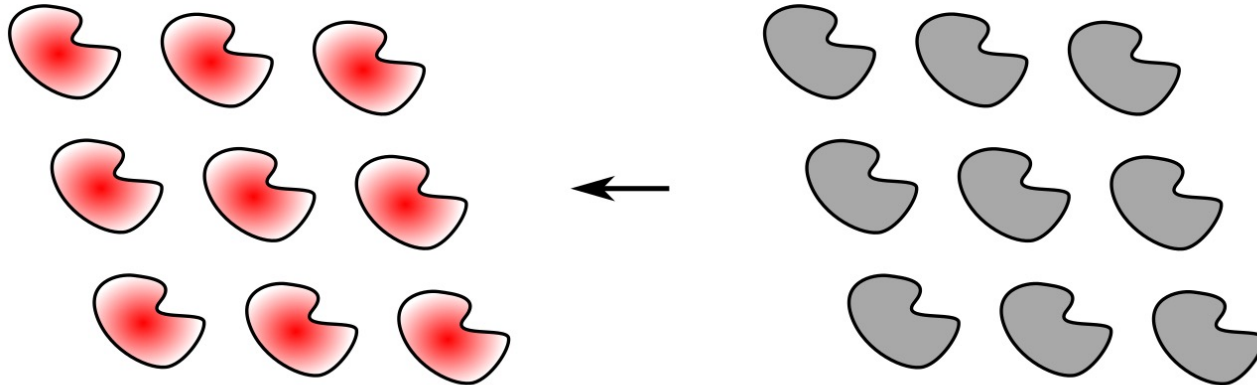




# Metamaterials: tailored nonlinear and spatio-temporal response

$$\mathbf{P}(\mathbf{r}) = \varepsilon_0 \sum \chi^{(n)}(\mathbf{r}) \mathbf{E}^n(\mathbf{r})$$

$$\mathbf{P}(\mathbf{r}) = \varepsilon_0 \chi(\mathbf{r}) \mathbf{E}(\mathbf{r})$$



Nonlinear metamaterials

*Nano Letters* **14**, 6488 (2014)

*ACS Photonics* **2**, 578 (2015)

*Nano Letters* **15**, 6985 (2015)

*Nano Letters* **16**, 4857 (2016)

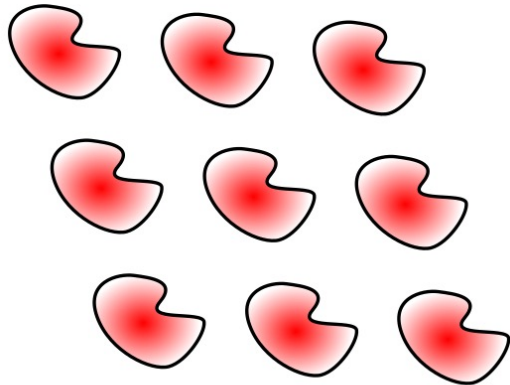
*Nature Communications* **8**, 17 (2017)

*Nature Communications* **10**, 1345 (2019)



# Metamaterials: tailored nonlinear and spatio-temporal response

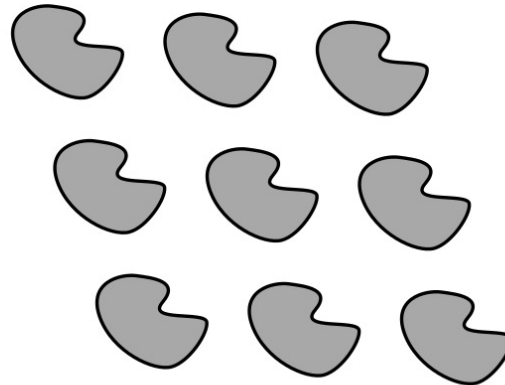
$$\mathbf{P}(\mathbf{r}) = \varepsilon_0 \sum \chi^{(n)}(\mathbf{r}) \mathbf{E}^n(\mathbf{r})$$



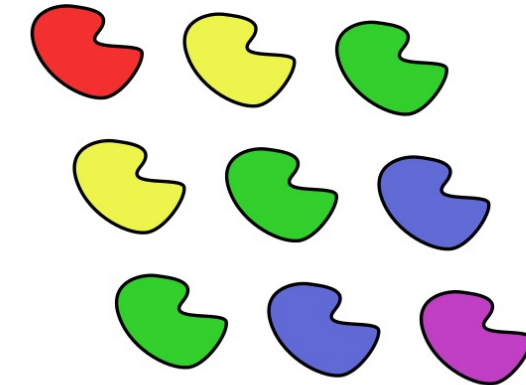
Nonlinear metamaterials

*Nano Letters* **14**, 6488 (2014)  
*ACS Photonics* **2**, 578 (2015)  
*Nano Letters* **15**, 6985 (2015)  
*Nano Letters* **16**, 4857 (2016)  
*Nature Communications* **8**, 17 (2017)  
*Nature Communications* **10**, 1345 (2019)

$$\mathbf{P}(\mathbf{r}) = \varepsilon_0 \chi(\mathbf{r}) \mathbf{E}(\mathbf{r})$$



$$\mathbf{P}(\mathbf{r}, t) = \varepsilon_0 \int d\mathbf{r}' \int dt' \chi(\mathbf{r}, t, \mathbf{r}', t') \mathbf{E}(\mathbf{r} - \mathbf{r}', t - t')$$

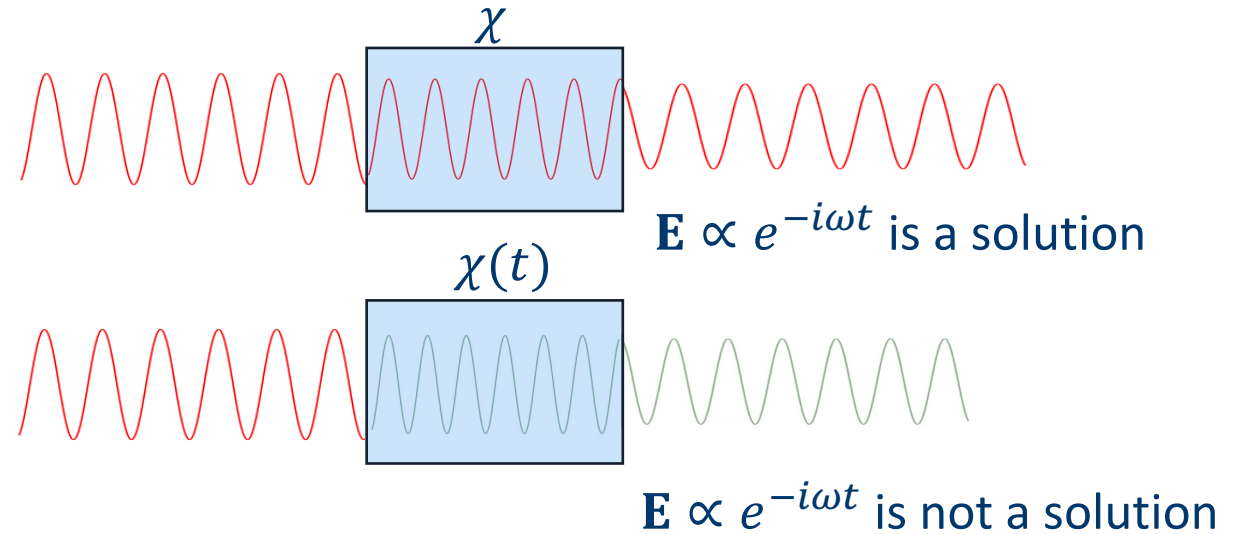
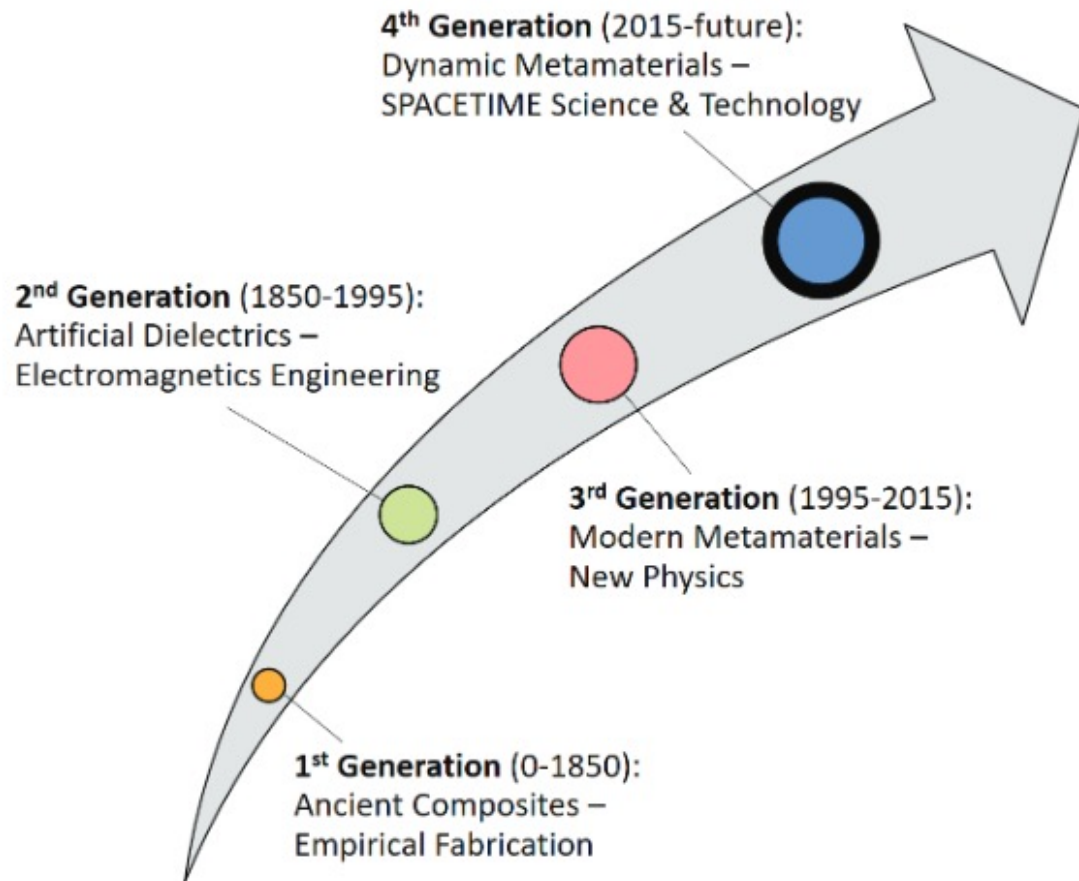


Time-variant metamaterials

*Nature Communications* **10**, 1345 (2019)  
*Optica (Memorandum)* **6**, 1441 (2019)  
*Physical Review A* **100**, 063847 (2019)  
*Nano Letters* **20**, 7052 (2020)  
*APL Materials* **9**, 060701 (2021)



# Time-variant metasurfaces

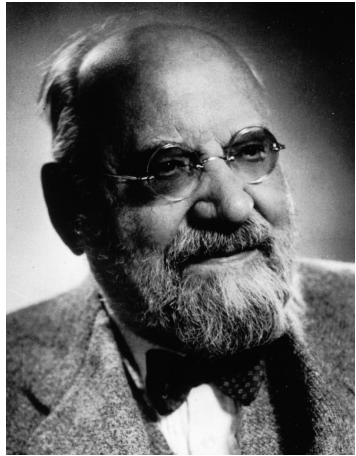


Tretyakov, Boyd, Pendry, Engheta, Alu, Segev, Shadrivov, Shvets, Huidobro, Boltasseva, Shalaev, Brongersma, Kinsey, Halevi, Khurgin, Caglayan, Faccio, Nassar, Narimanov, Monticone, Sapienza, Fleury, Rodriguez, Lurie, Ramezani, Ramaccia many others

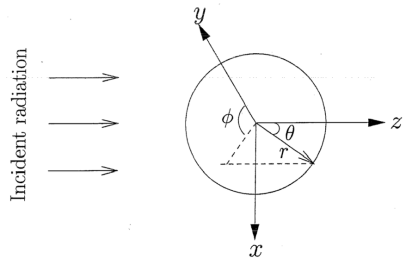
## Experiments in time-variant metasurfaces

*Nano Letters* **20**, 7052 (2020)

*Nature Communications* **10**, 1345 (2019)

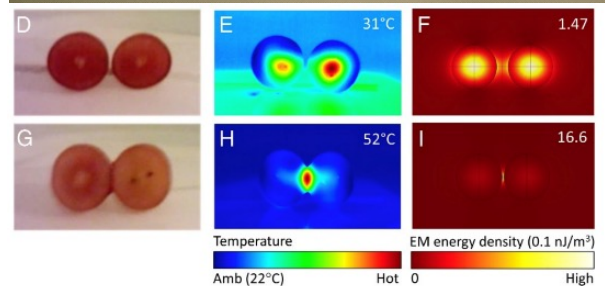
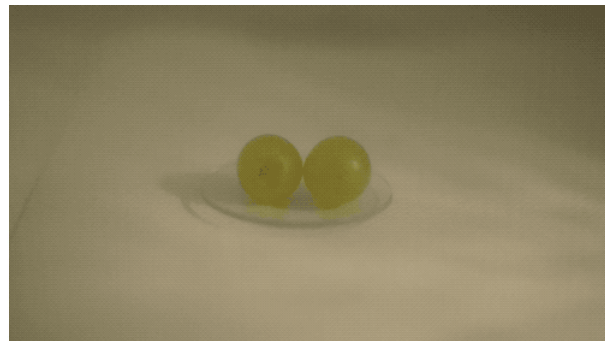
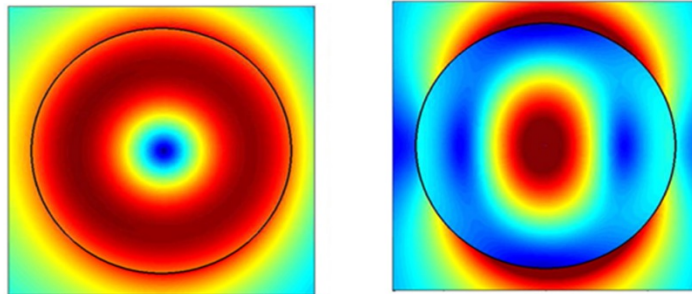


Gustav Mie  
(1869 – 1957)



$|\mathbf{E}|^2$  maps

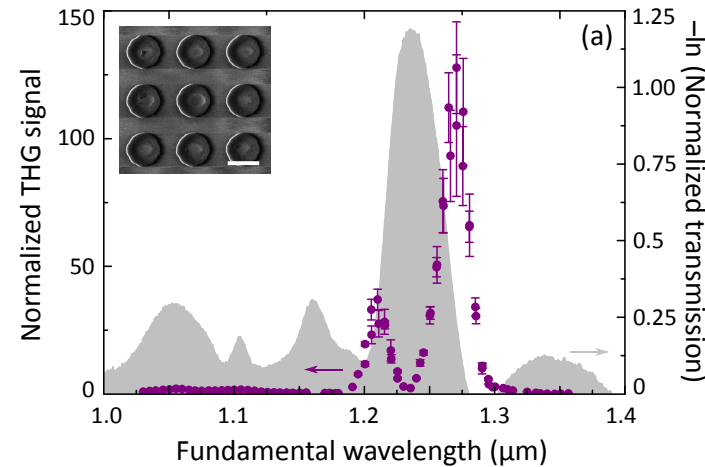
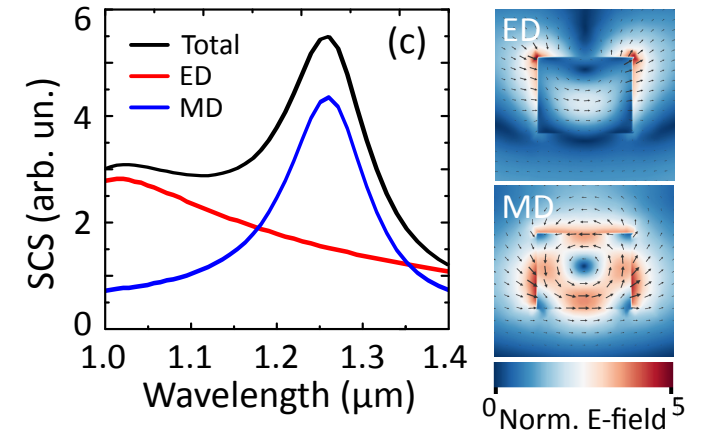
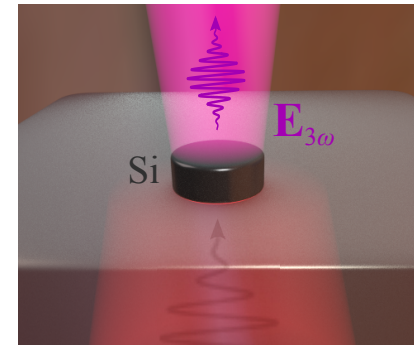
(1) Magnetic dipolar (2) Electric dipolar



Khattak et al., *PNAS* **116**, 4000 (2019)

$$\tilde{P} = \chi^{(1)} \tilde{E}(t) + \chi^{(2)} \tilde{E}^2(t) + \chi^{(3)} \tilde{E}^3(t) + \dots$$

$$\tilde{E}(t) \propto e^{i\omega t} \quad \propto e^{2i\omega t} \quad \propto e^{3i\omega t}$$



10<sup>2</sup> enhancement  
vs bulk silicon

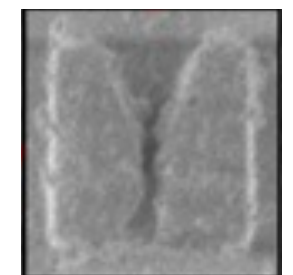
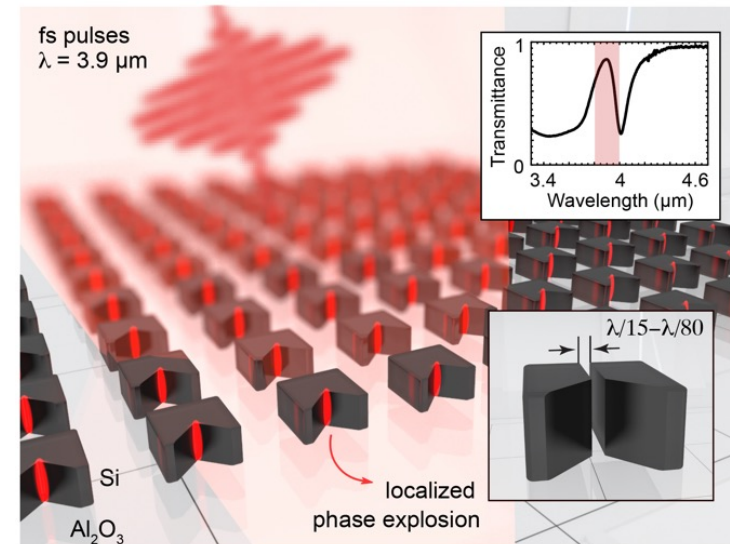
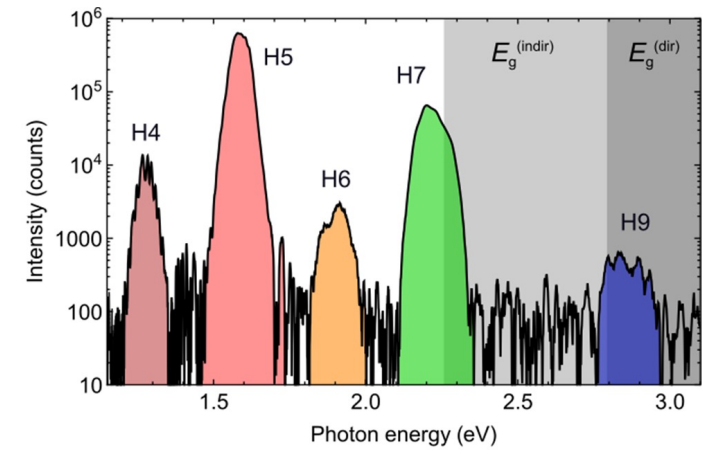
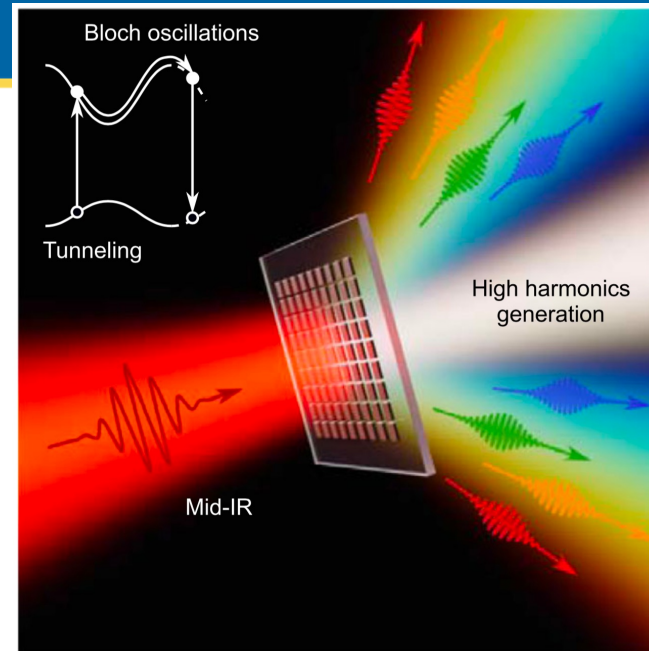
Shcherbakov et al.,  
*Nano Lett.* **14**, 6488 (2014)

Bohren, C. F. & Huffman, D. R.  
*Absorption and Scattering of Light by Small Particles*  
Wiley Inter-Science, 1998.



# Outline

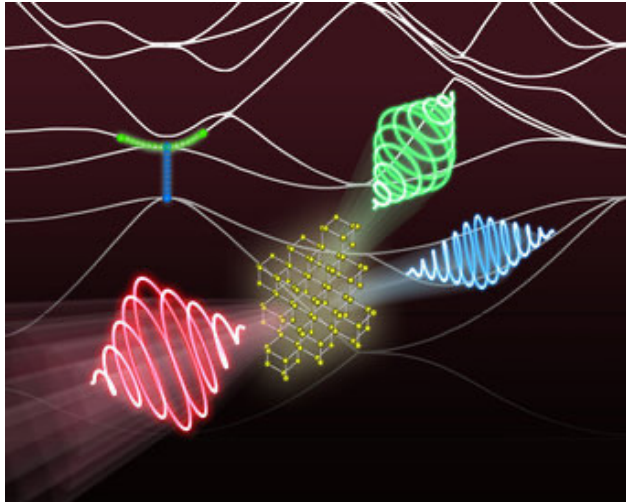
- Strong fields:  
high harmonics generation
- Very strong fields:  
laser damage and nanomachining



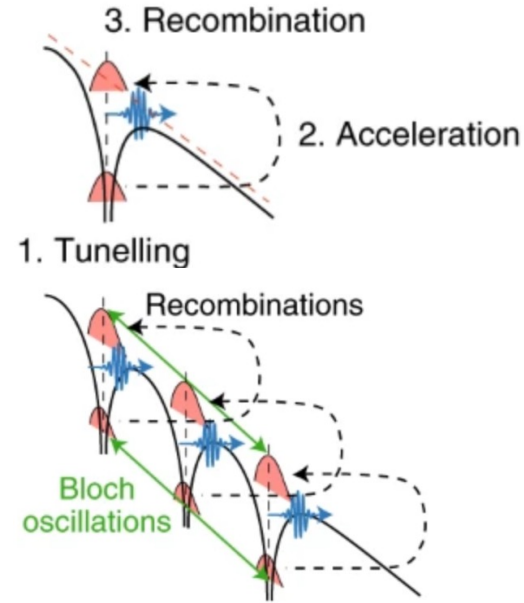




# High harmonic generation

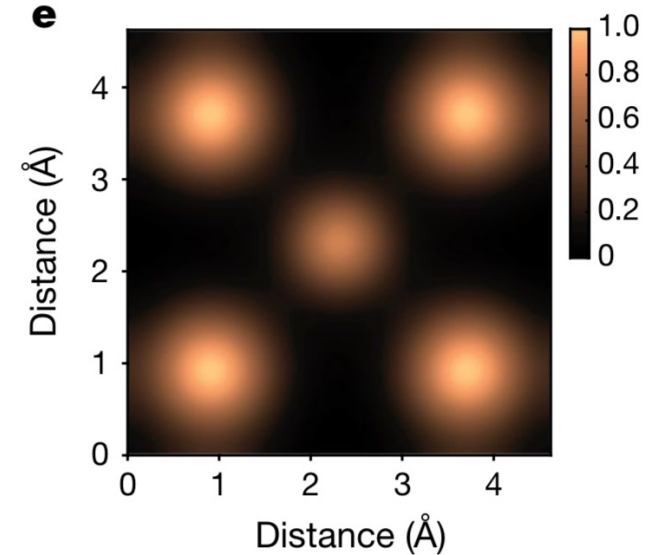
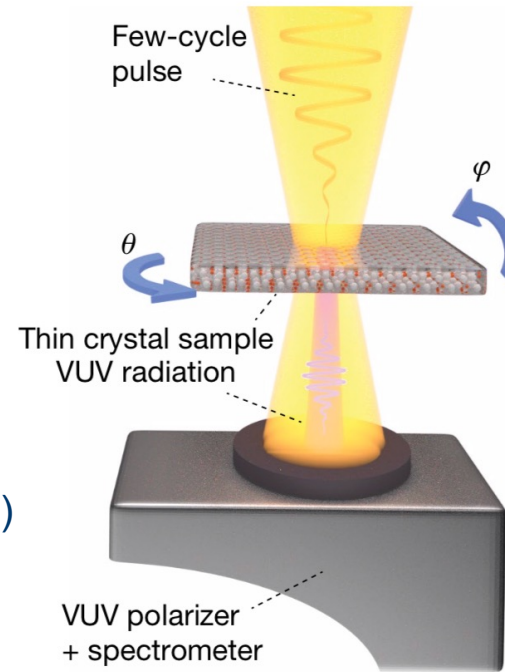


© Nicolas Tancogne-Dejean  
+ Joerg M. Harms, MPSPD

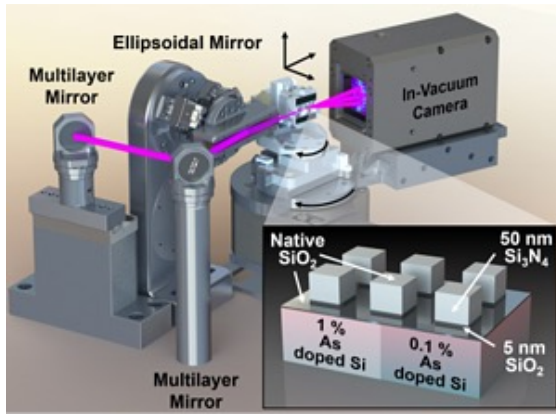


Ghimire & Reis,  
*Nature Physics* **15**, 10–16 (2019)

## Pushing the resolution limits



## XUV sources for photolithography



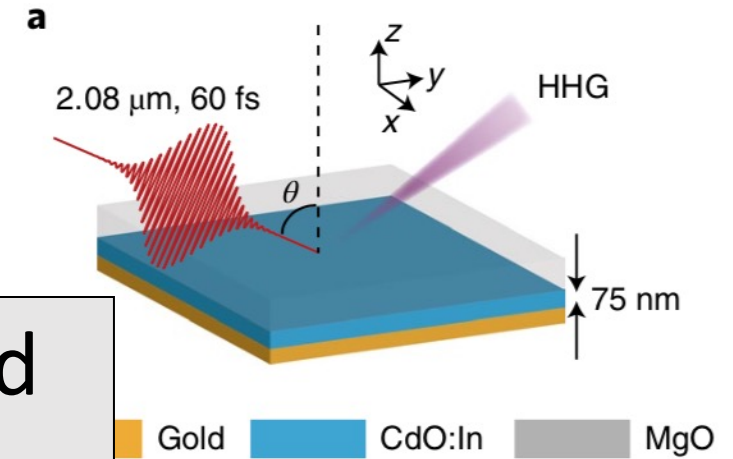
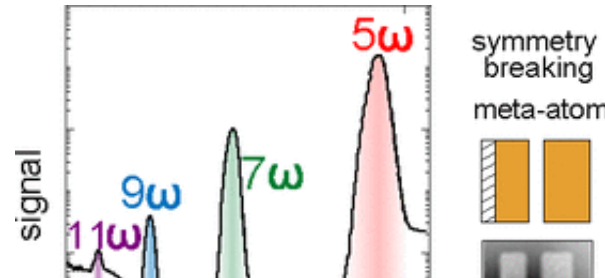
### Article

## Laser picoscopy of valence electrons in solids

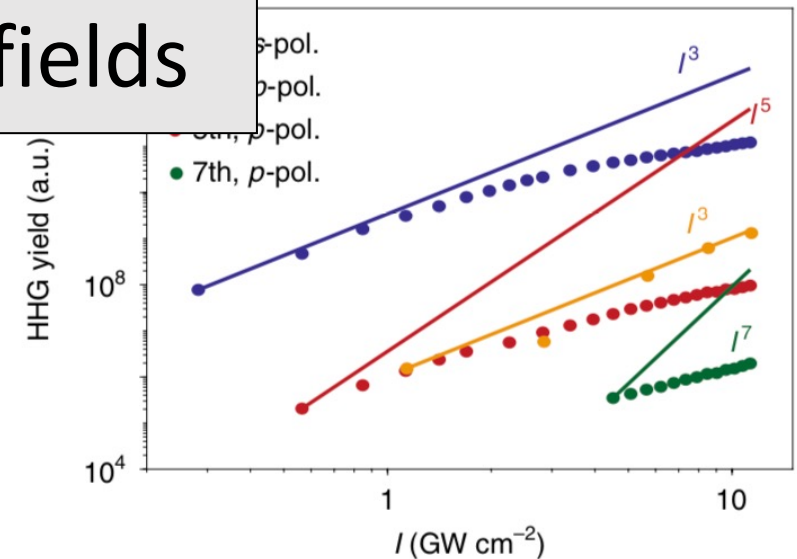
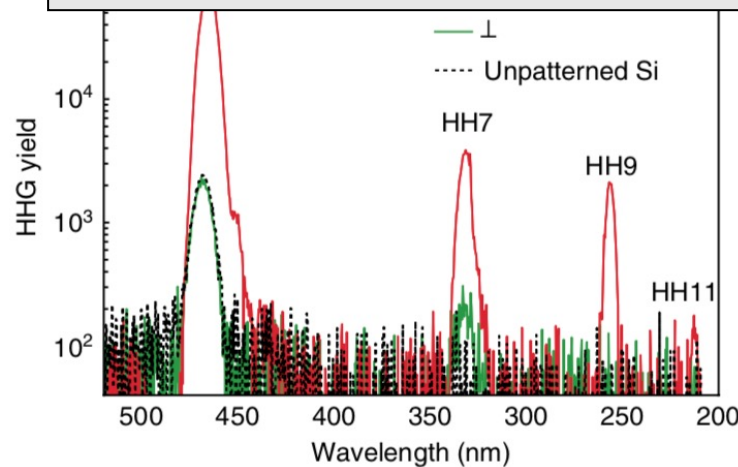
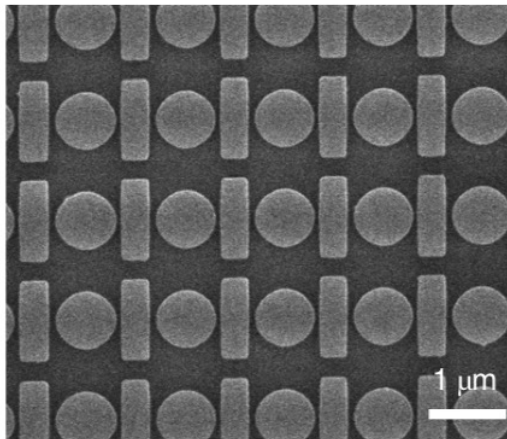


# HHG in nanostructures

- Nanostructured solids are natural candidates for HHG
- No phase matching issues
  - Enhanced local field

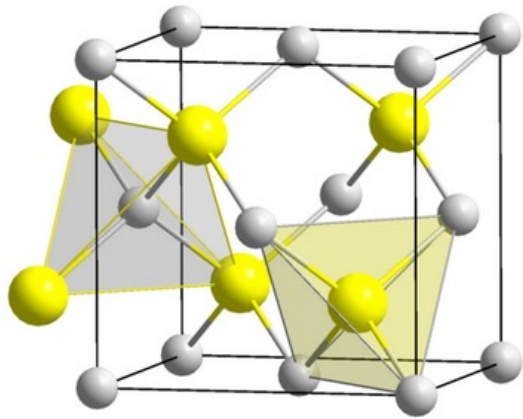


- Even harmonics are limited
- High reabsorption
- Hard to access strong fields



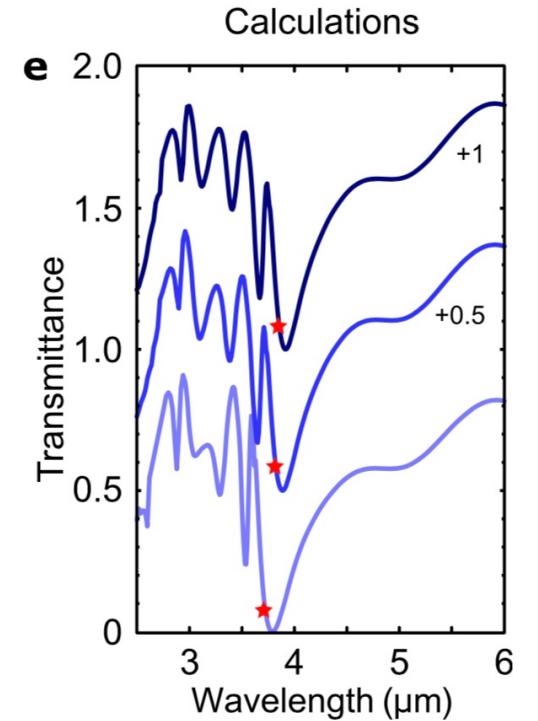
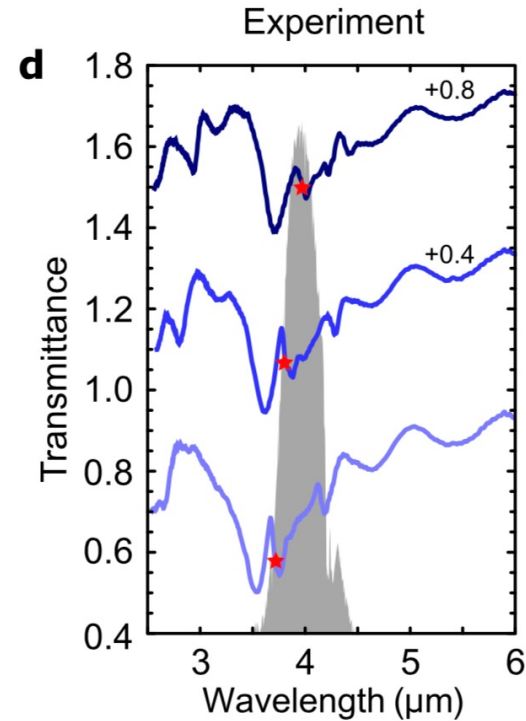
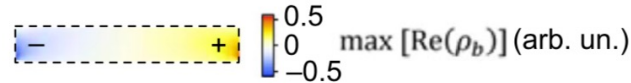
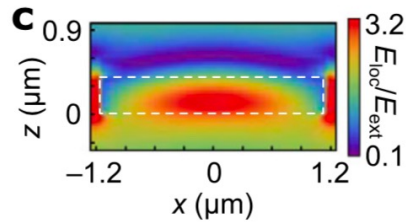
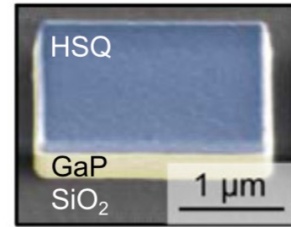
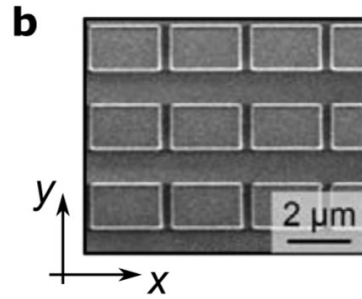


# GaP — material of choice



■ Ga<sup>3+</sup> ■ P<sup>3-</sup>

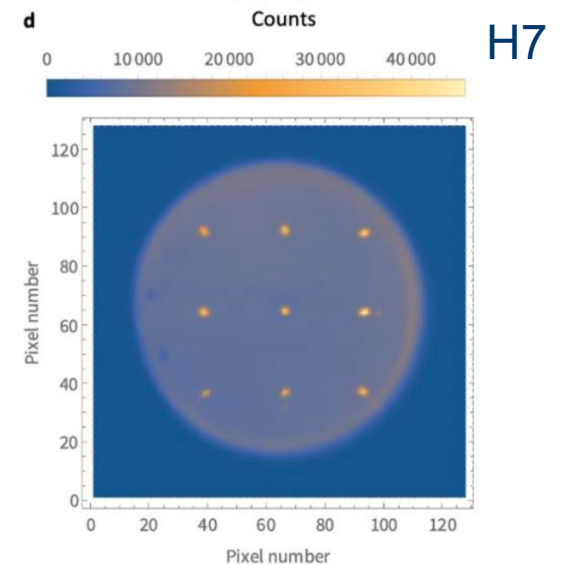
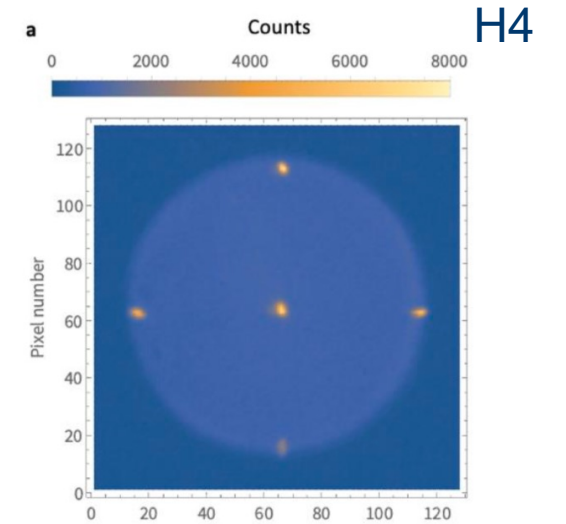
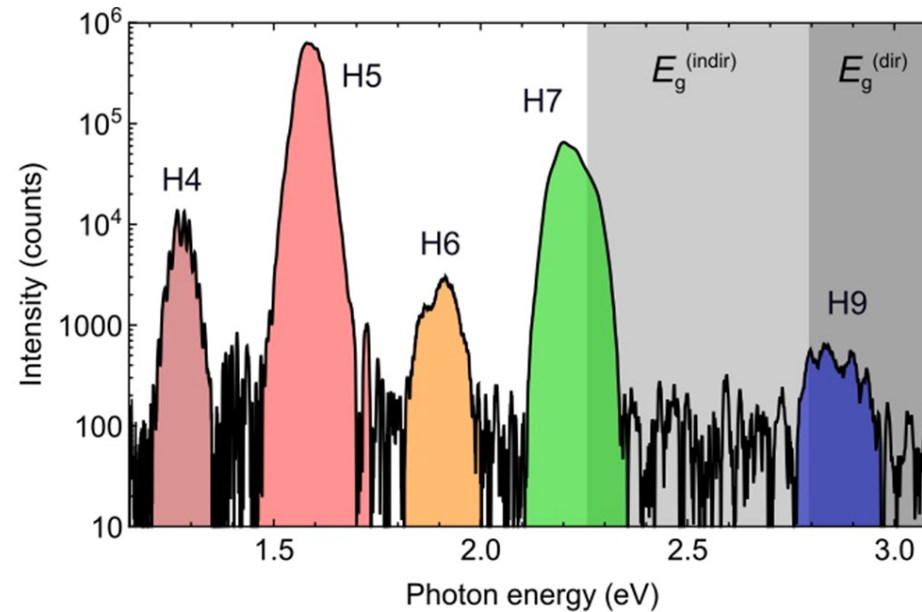
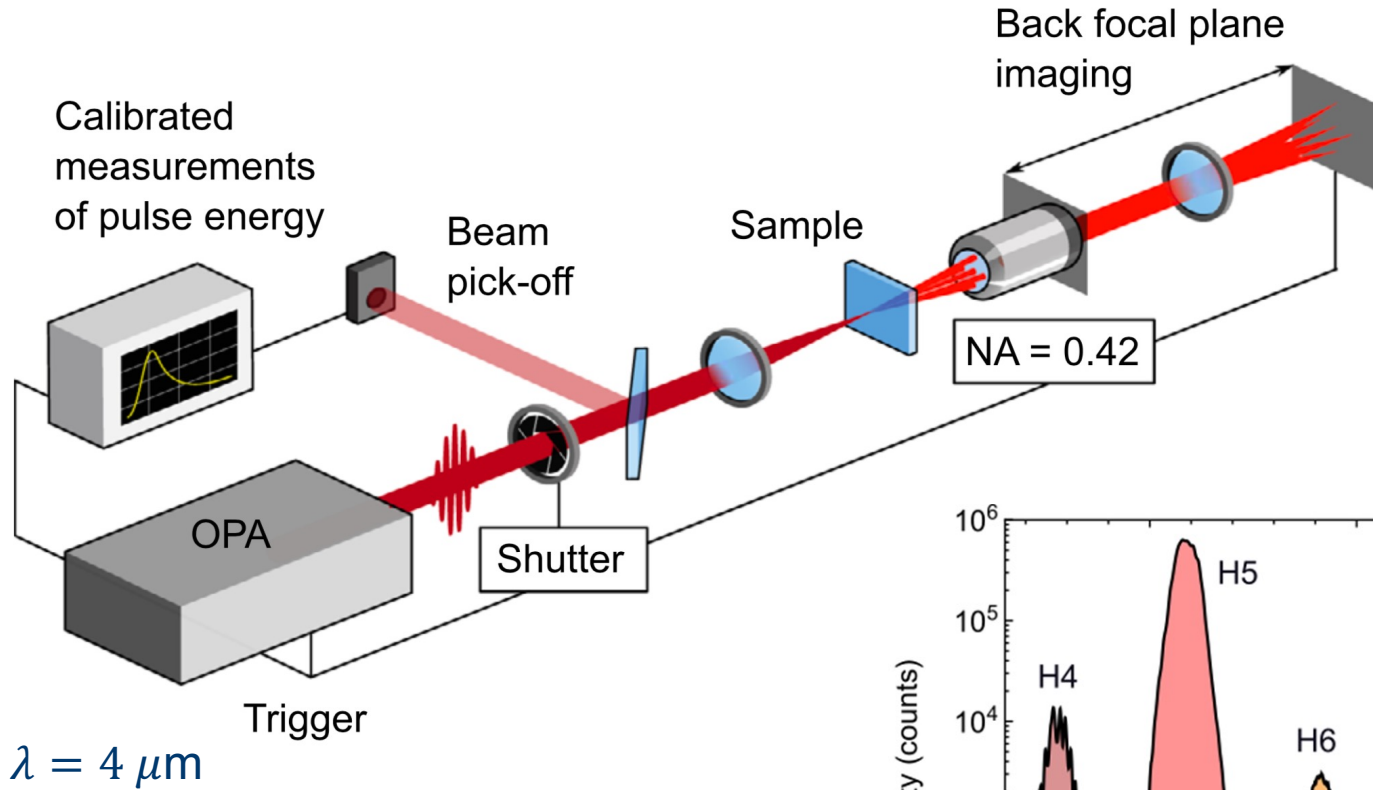
- Noncentrosymmetric
- Good tradeoff between  $E_{bg}$  and refractive index



Fabrication — Arseniy Kuznetsov's group, A\*Star (Singapore)



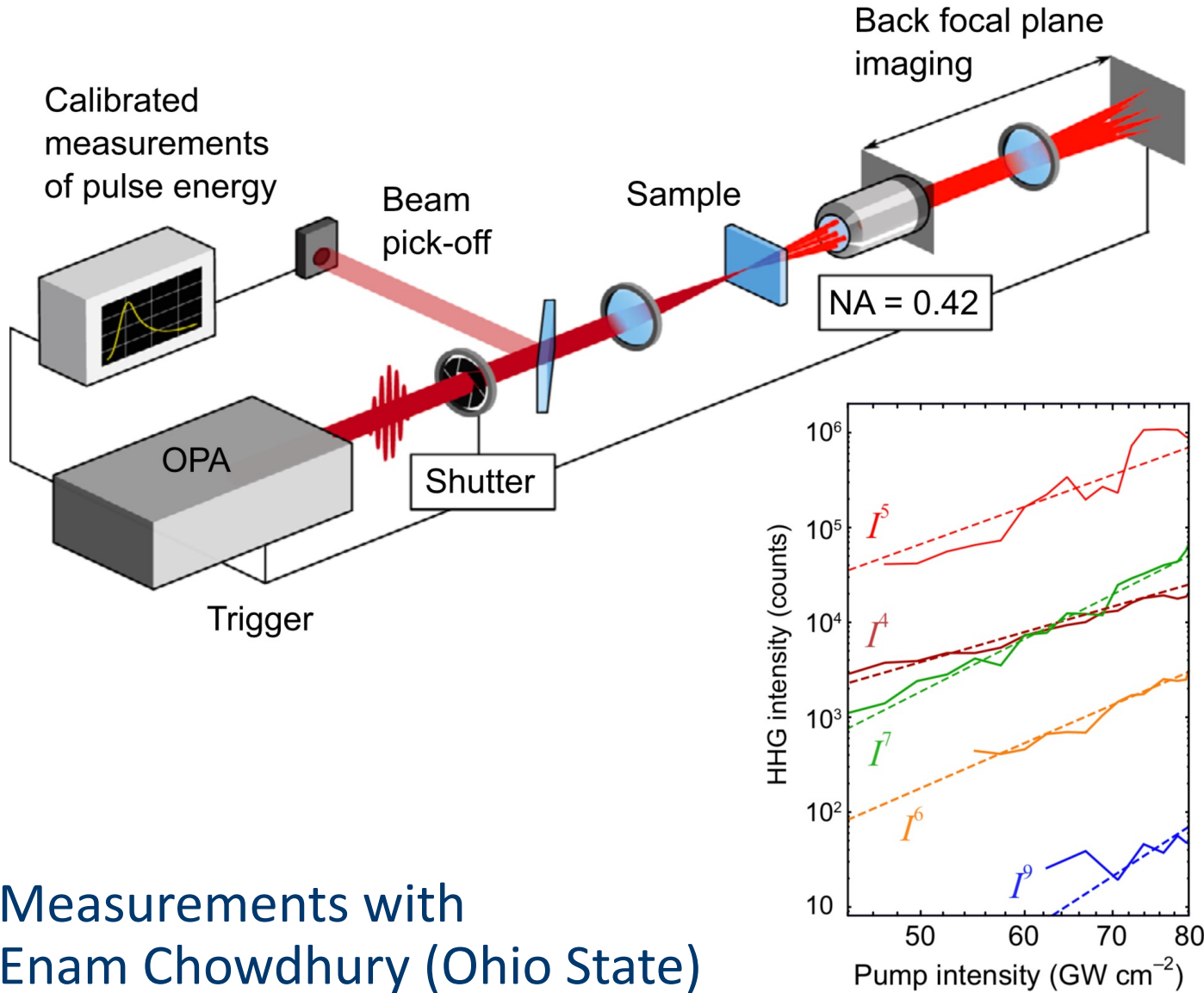
# HHG measurements



Measurements with  
Enam Chowdhury (Ohio State)



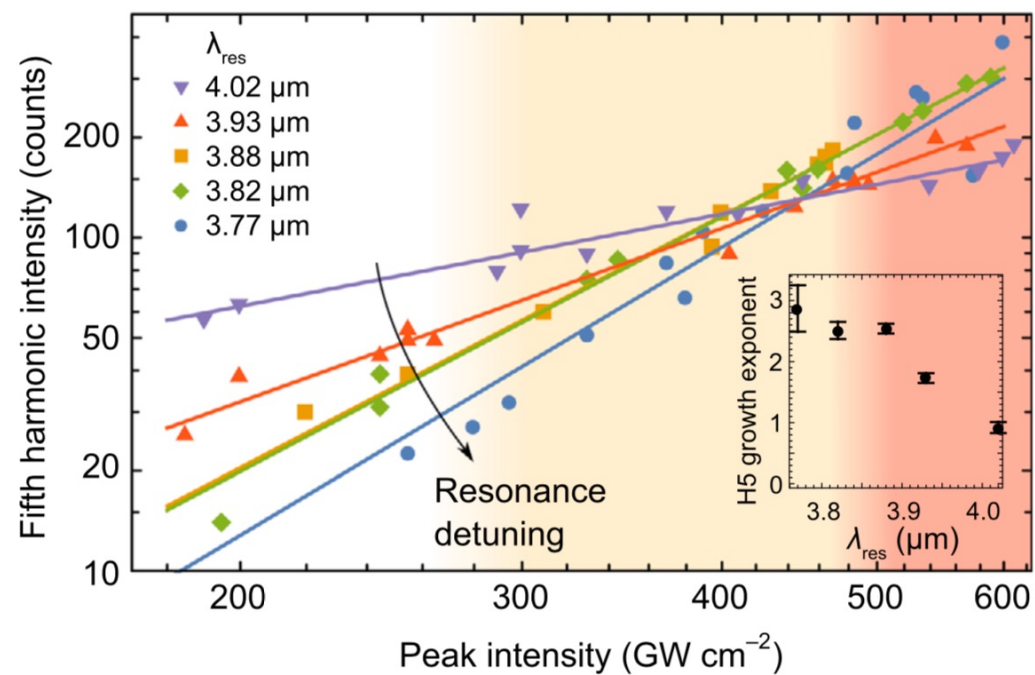
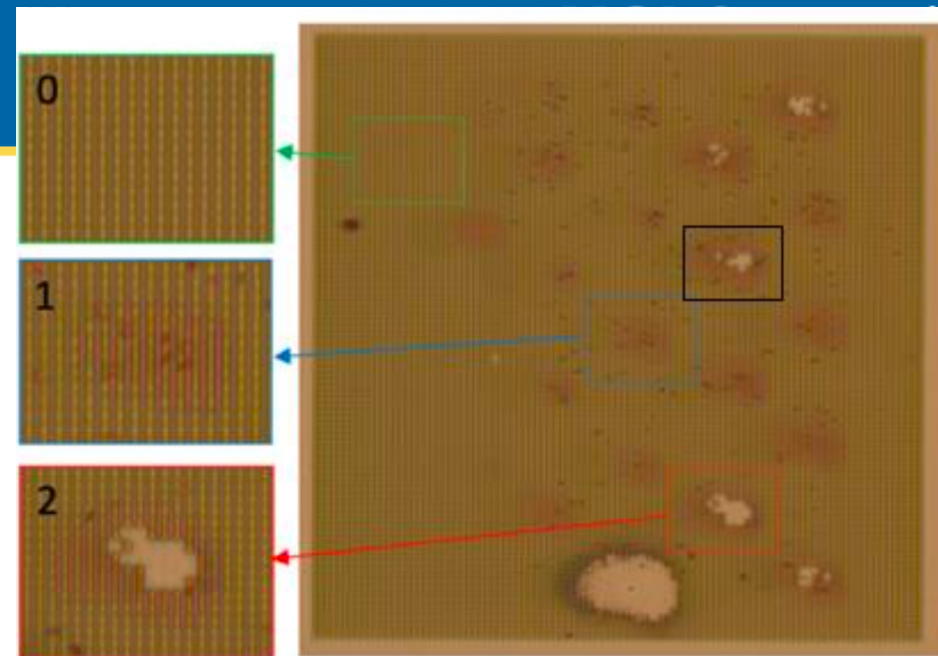
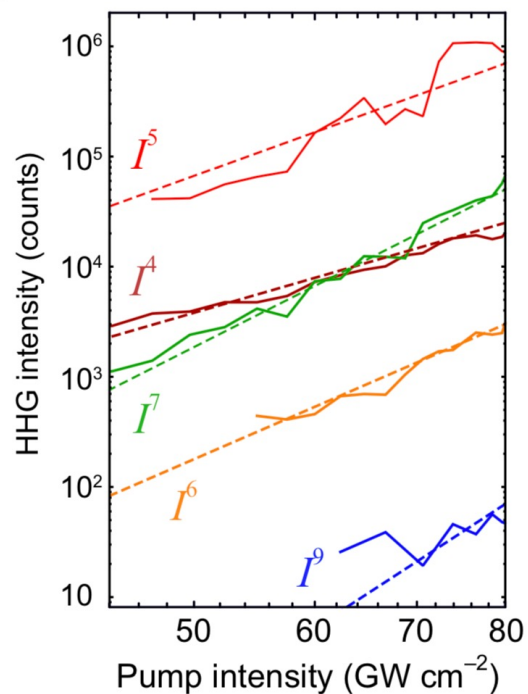
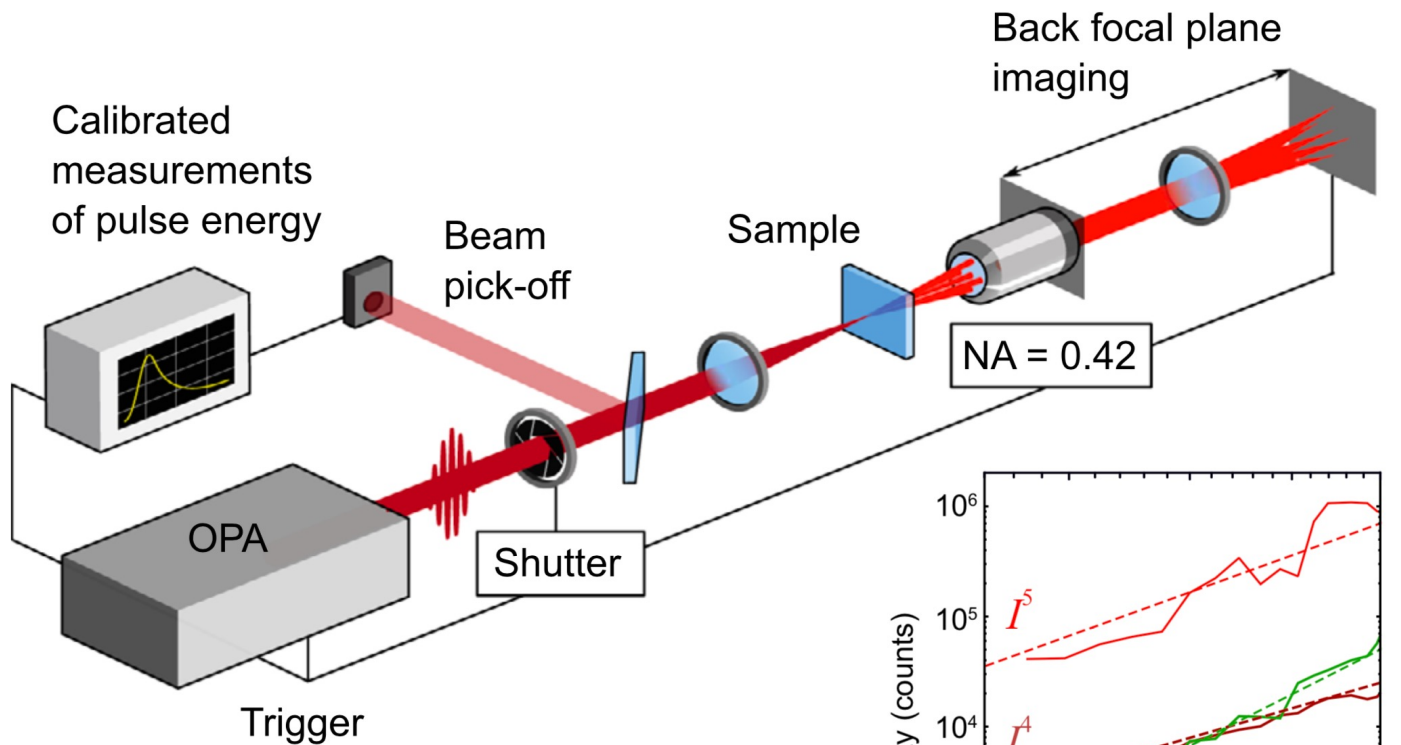
# Single-shot HHG



Measurements with  
Enam Chowdhury (Ohio State)



# Single-shot HHG

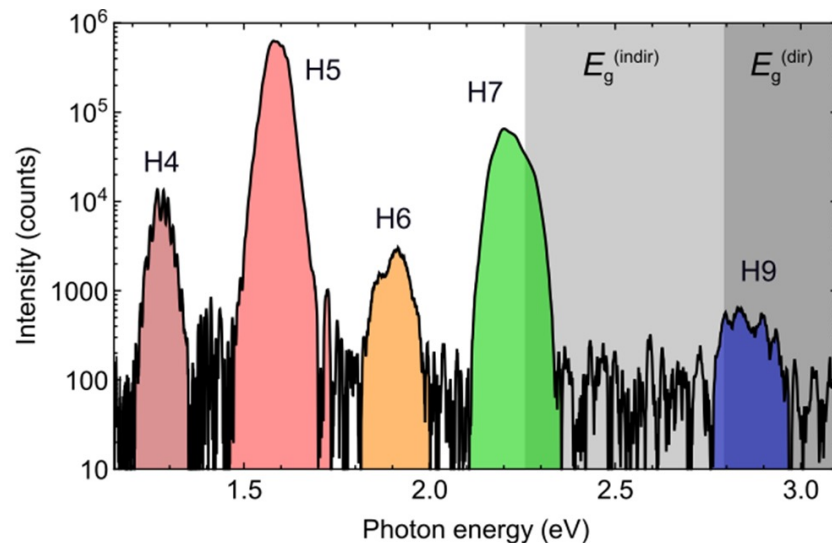
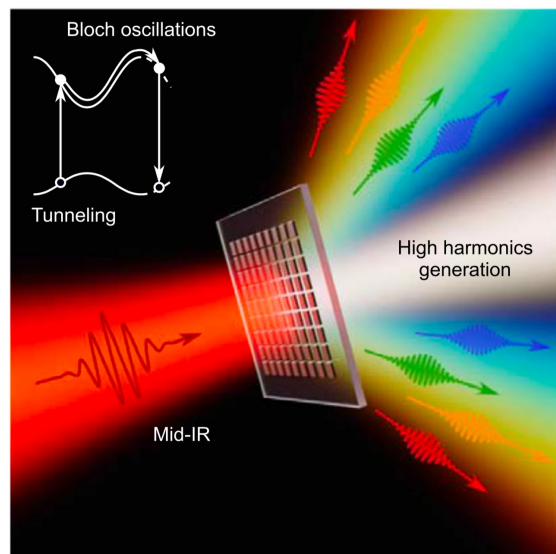


Measurements with  
Enam Chowdhury (Ohio State)



# Conversion efficiencies

Material	Harmonic order	Conversion efficiency	Efficiency per 1 $\mu\text{m}$ thickness
GaP metasurface [this work]	5 (SP)	$1.4 \times 10^{-6}$	$3.5 \times 10^{-6}$
	7 (MP)	$2 \times 10^{-9}$	$5 \times 10^{-9}$
ZnO [8]	5	$3 \times 10^{-5}$	$10^{-7}$
	7	$6 \times 10^{-6}$	$2 \times 10^{-8}$
Periodically poled LiNbO <sub>3</sub> [9]	5	$10^{-2}$	$4 \times 10^{-7}$
	7	$10^{-2}$	$4 \times 10^{-7}$
Si metasurface [10]	5	$5 \times 10^{-9}$	$2.2 \times 10^{-8}$
ENZ material [11]	5	$10^{-8}$	$1.3 \times 10^{-7}$
	7	$10^{-10}$	$1.3 \times 10^{-9}$



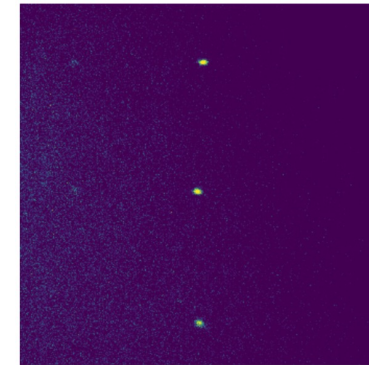
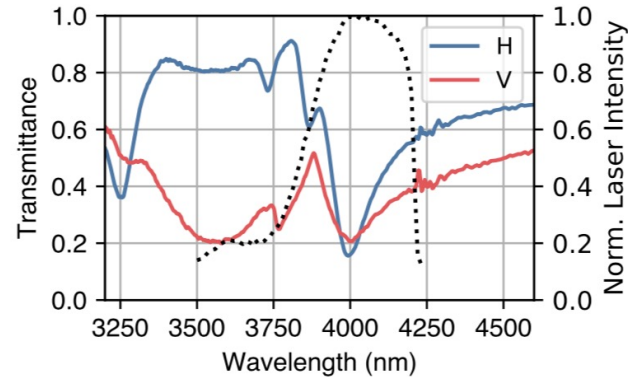
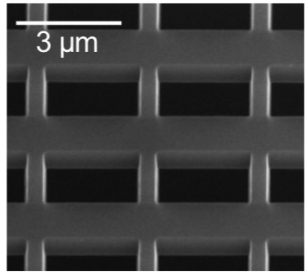
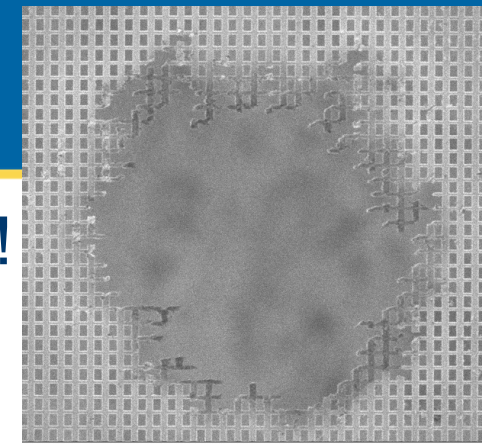
Shcherbakov et al.,  
*Nature communications* **12**, 4185 (2021)



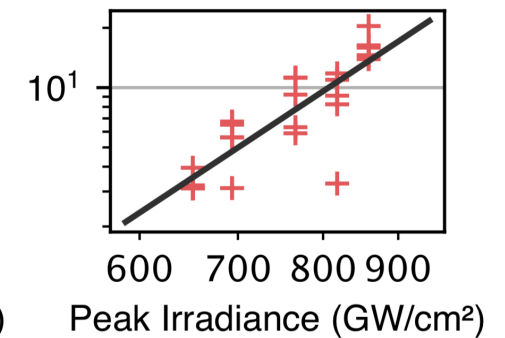
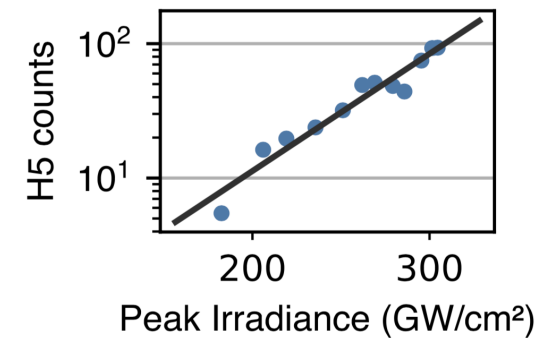
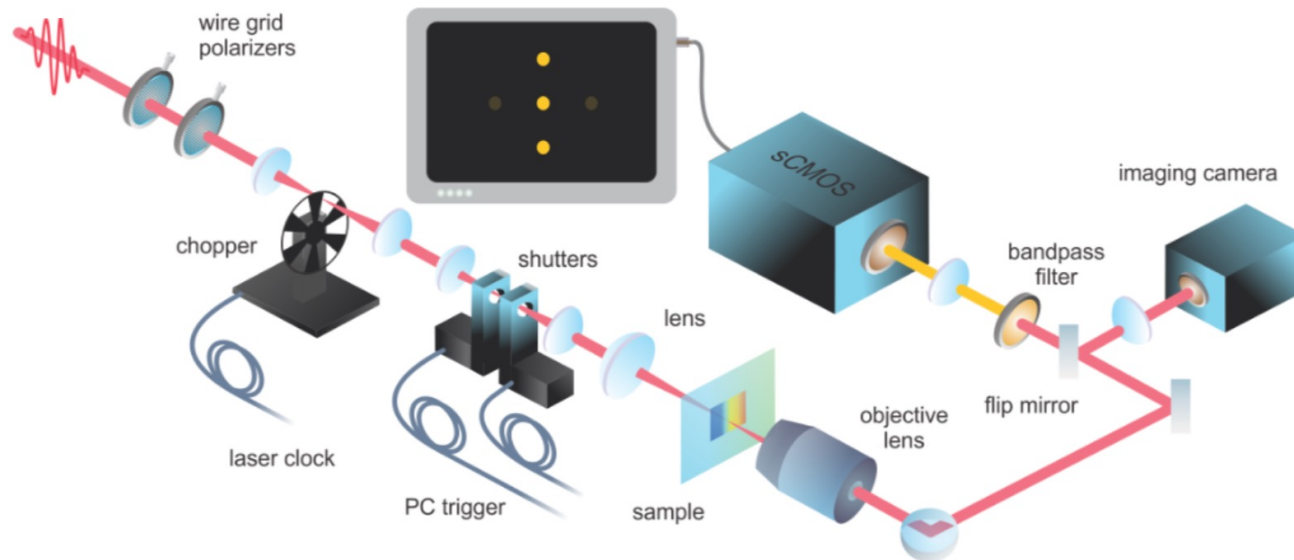
# SiC free-standing membranes

Samples: Free-standing membrane SiC metasurface  
Daniil Lukin, Jelena Vuckovic

Pulse train = damage!



Single shot  
back focal plane H5



$$I(5\omega) \sim I^5(\omega)$$

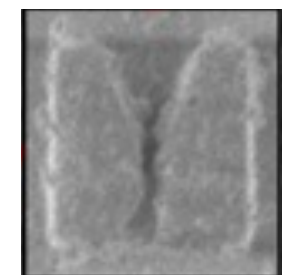
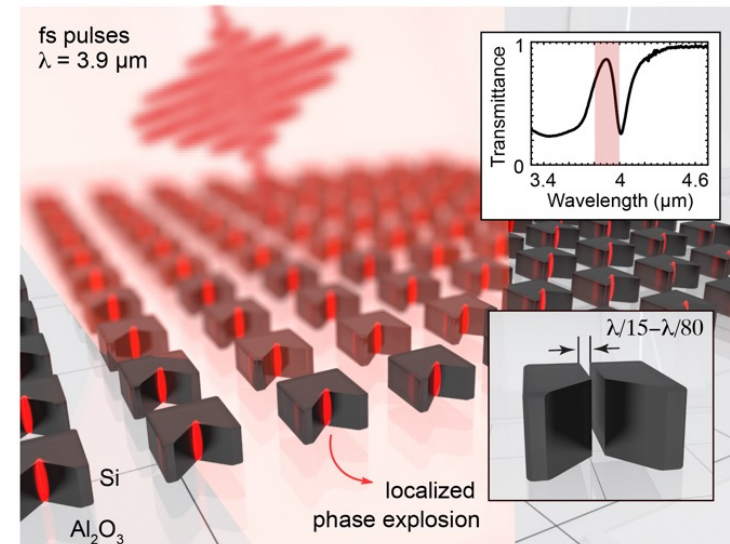
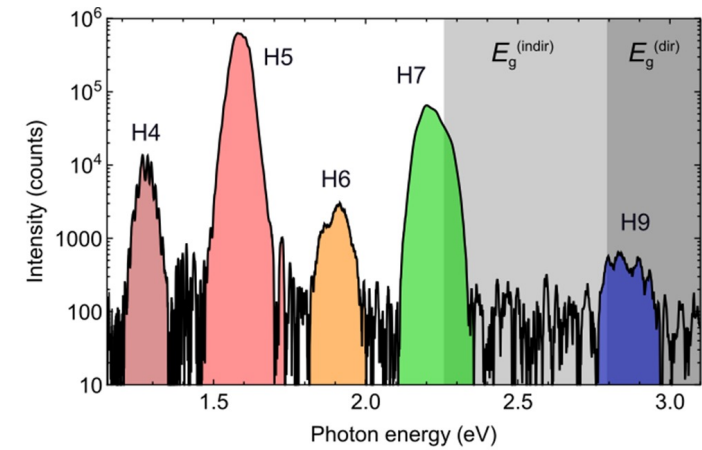
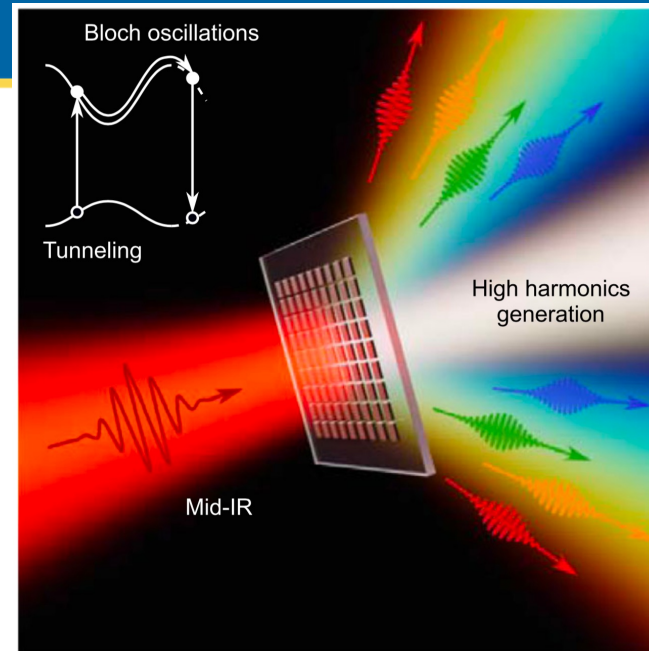
Measurements: Shvets group, Fishman group, Shcherbakov group





# Outline

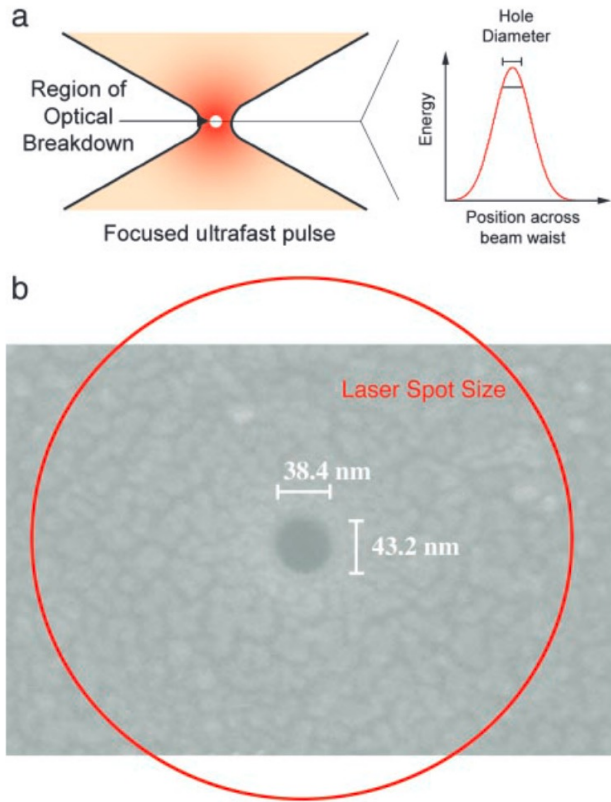
- Strong fields:  
high harmonics generation
- Very strong fields:  
laser damage and nanomachining





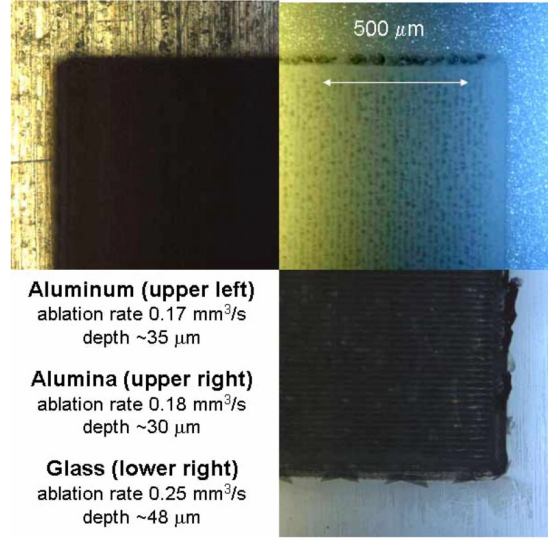
# Laser Nanomachining

## Single-pulse

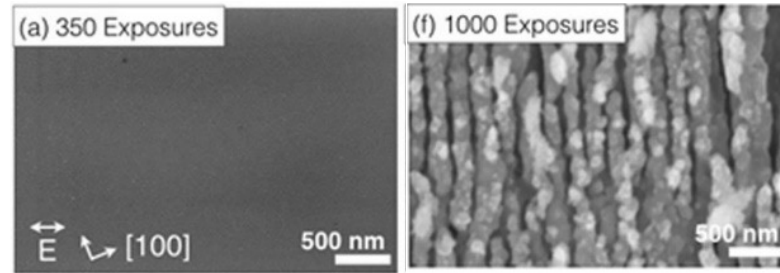


Joglekar et al., *PNAS* **101** 5856 (2003)

## Multi-pulse



Shah et al., *Optics Express* **14**, 12546 (2006)

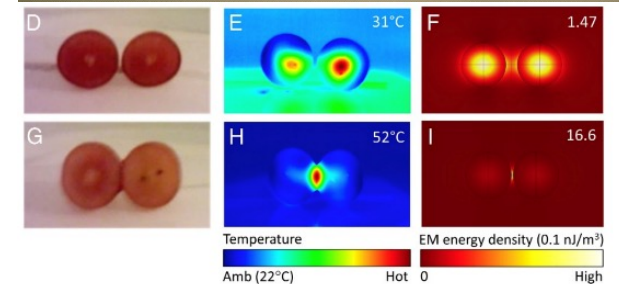
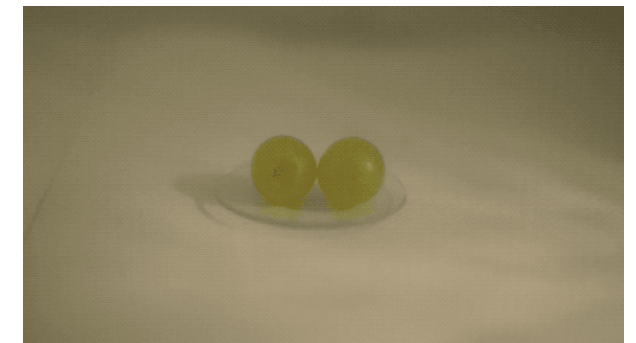
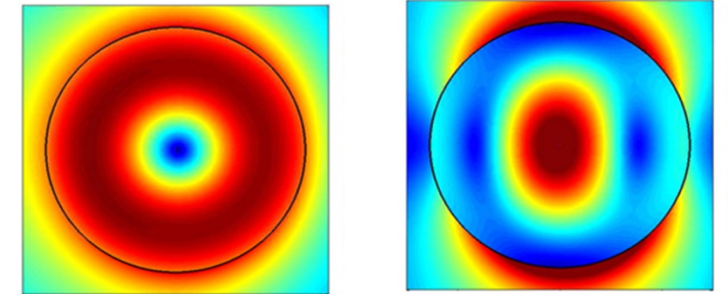


Abere et al., *Journal of Applied Physics* **126**, 143102 (2019)

## $|E|^2$ maps

(1) Magnetic dipolar

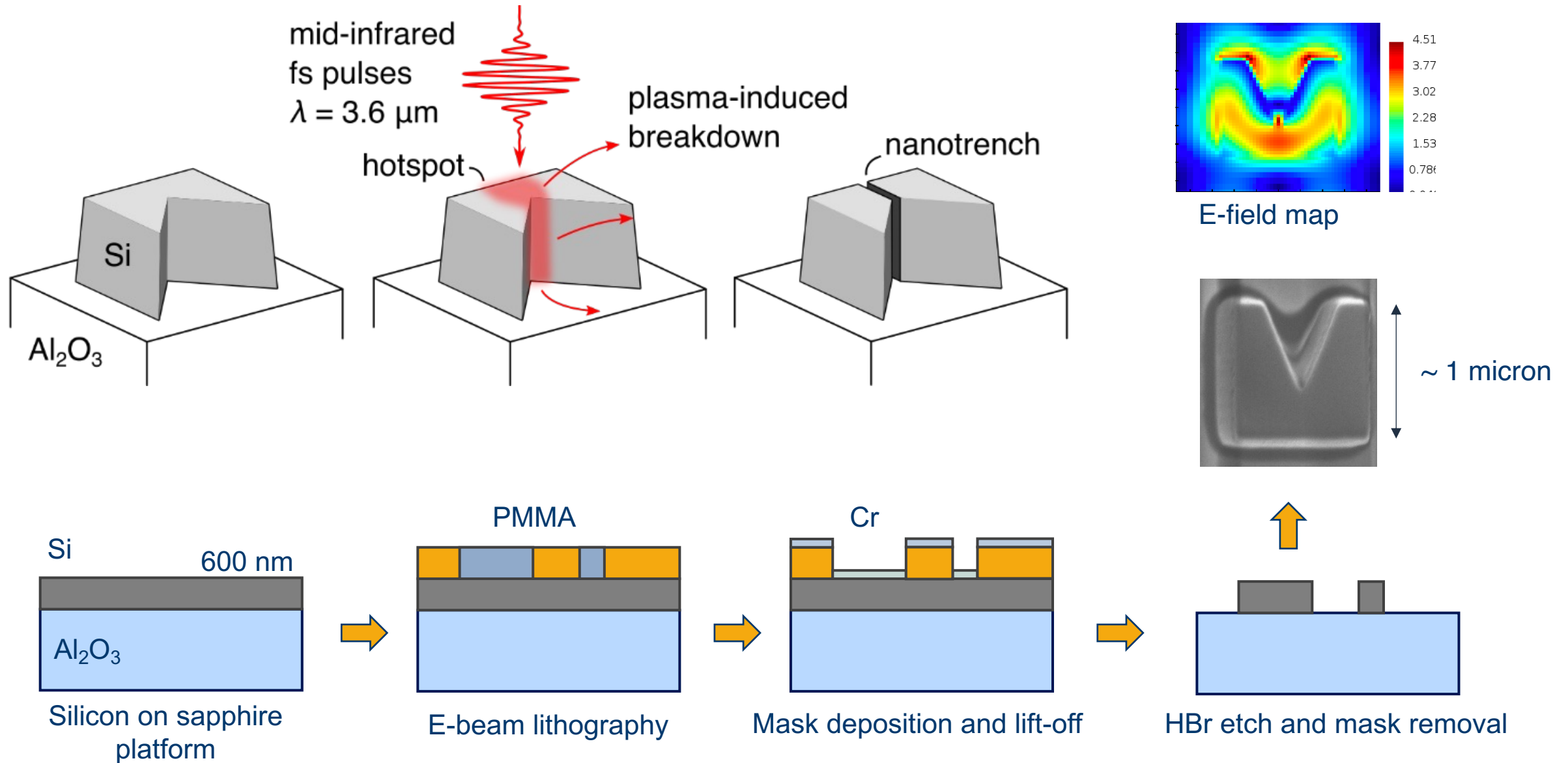
(2) Electric dipolar

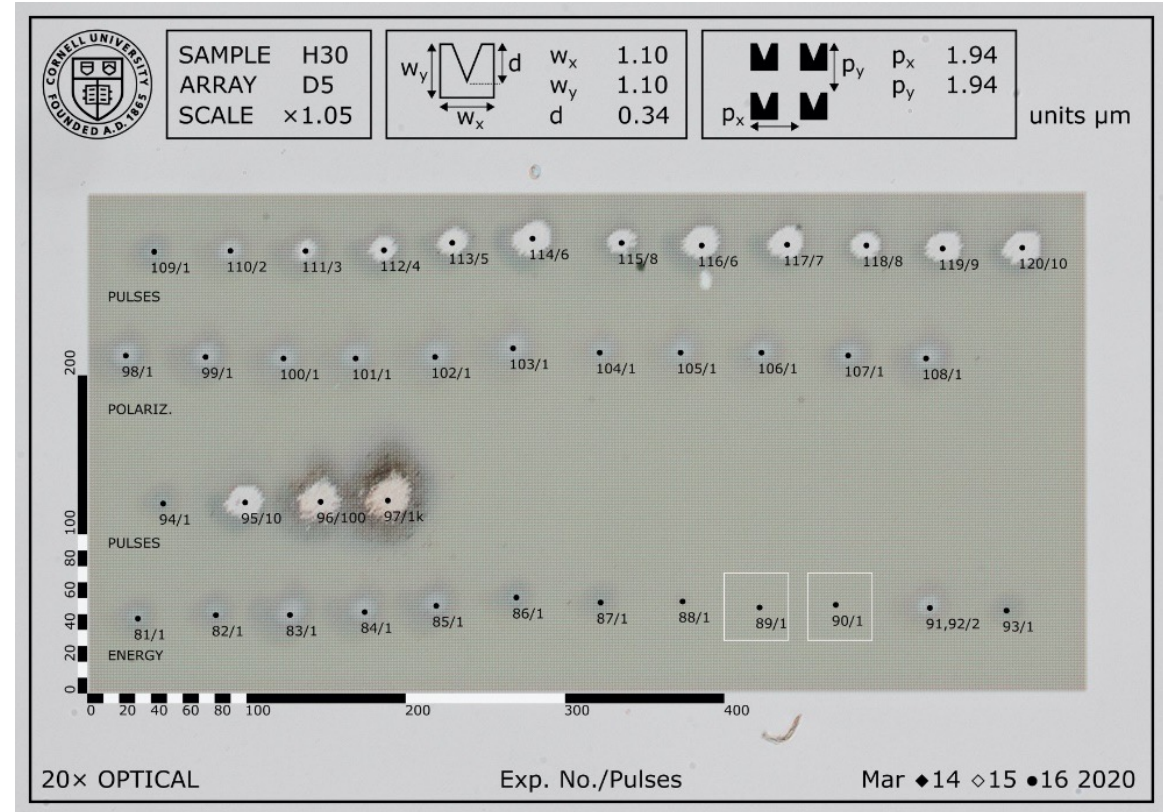
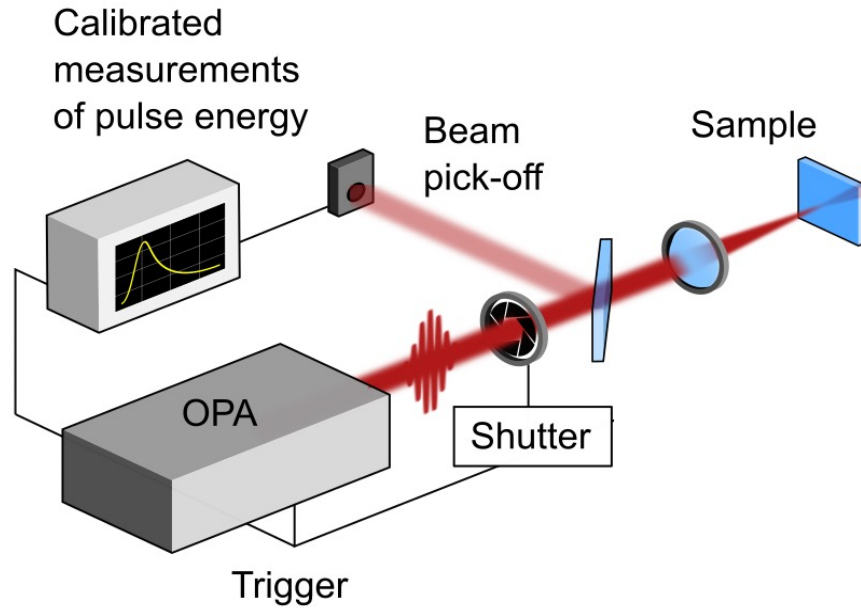


Khattak et al., *PNAS* **116**, 4000 (2019)



# Resonators for deep-subwavelength machining

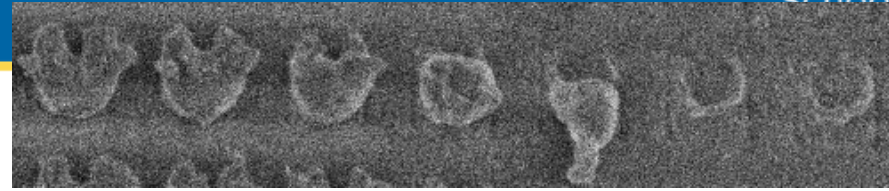




Variations in pulse energy, number and polarization



# Damage Type 1: Single-pulse Above Threshold



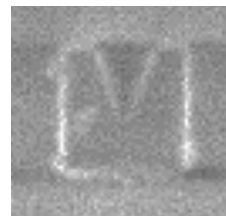
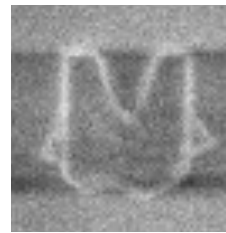
Outside  $\longrightarrow$  Beam center

“Wings”

“Blob”

Detachment

E-field



Beam center

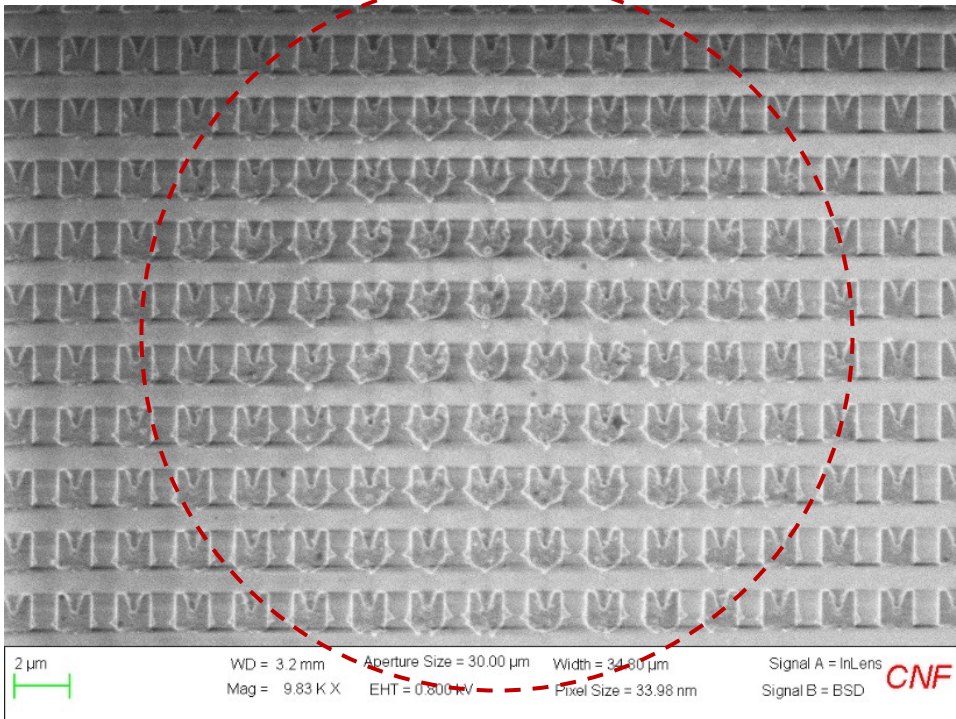
Damage threshold:  $< 0.05 \text{ J/cm}^2$

Typical mid-IR Si single-pulse damage threshold:  $\sim 0.25 \text{ J/cm}^2$

Werner et al., *Scientific Reports* 9, 19993 (2019)

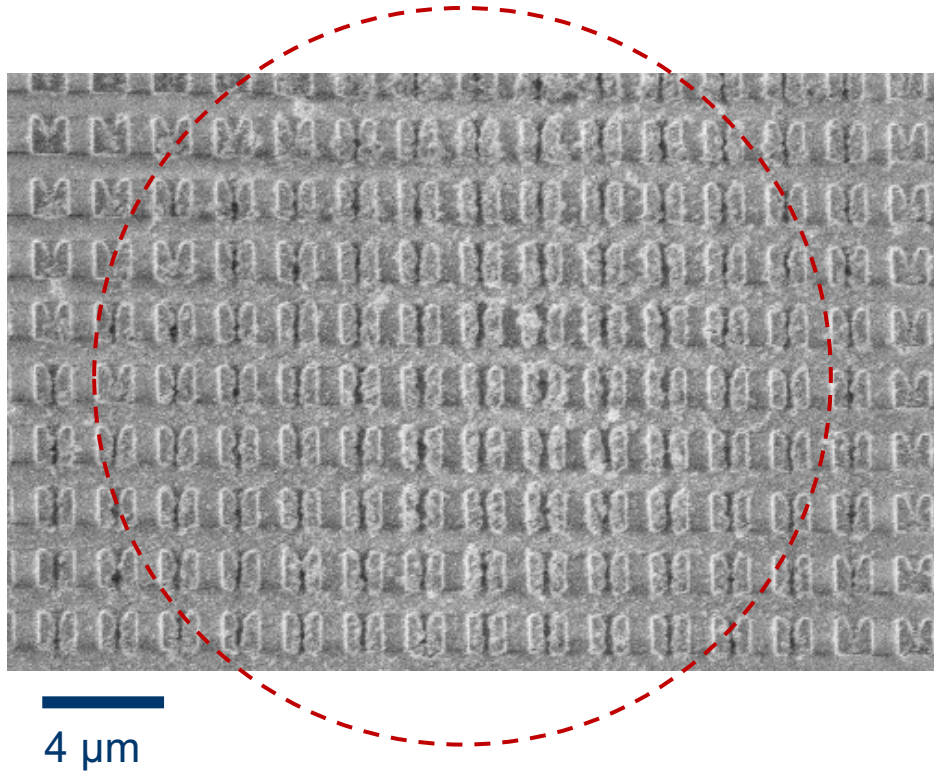
Outside

Beam FWHM

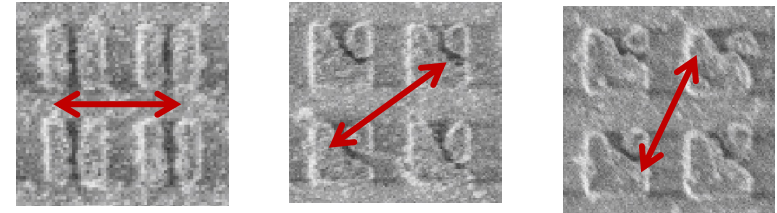




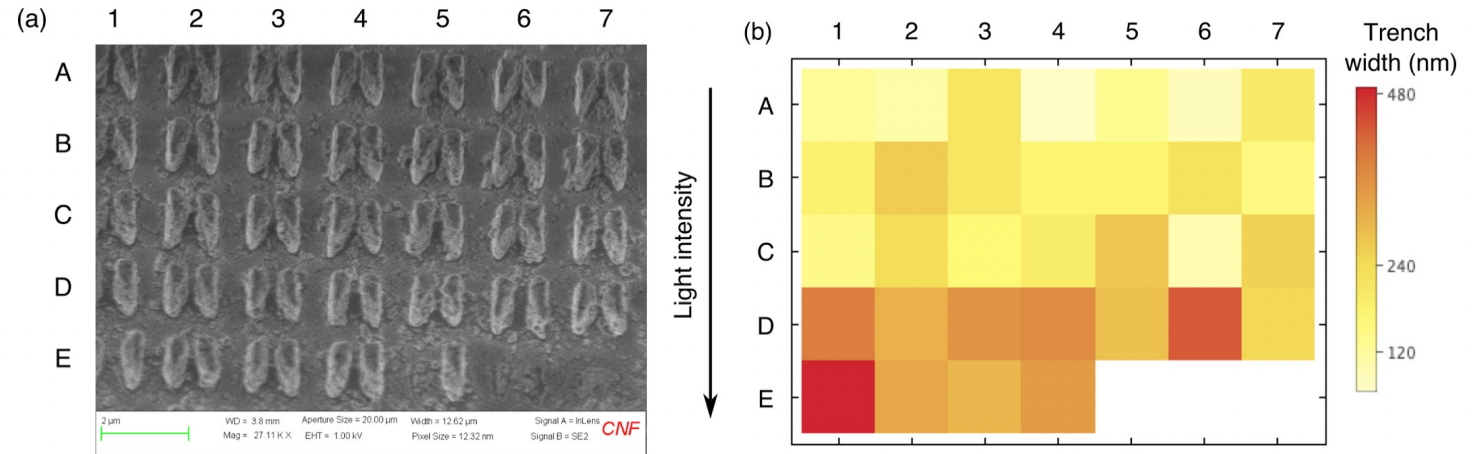
# Damage Type 2: Sub-threshold "Trenching"



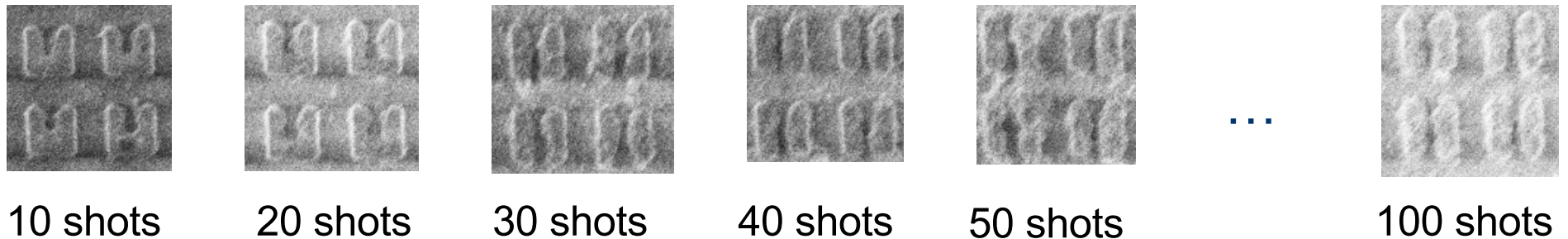
Trench control by polarization



Trench control by intensity



Trench control by number of shots





# Dense plasma formation and machining

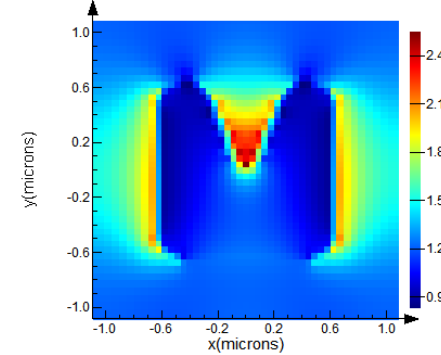
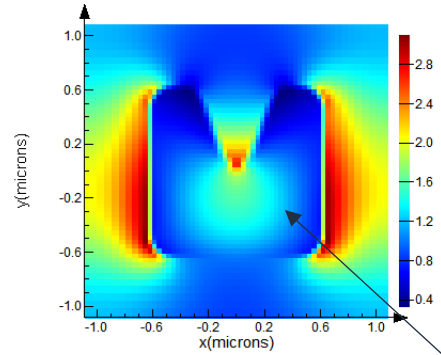
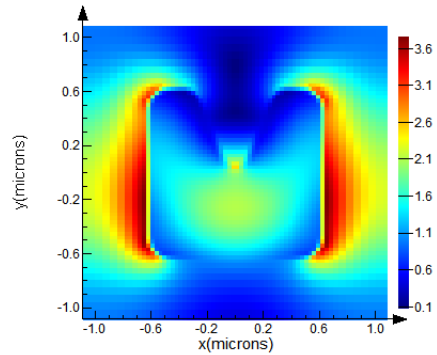
$$\varepsilon = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}, \quad \varepsilon_\infty = 12, \quad \gamma^{-1} = 10 \text{ fs}, \quad \omega_{p,crit} = \omega\sqrt{\varepsilon_\infty} = 1.6 \cdot 10^{15} \text{ s}^{-1}$$

$$\omega_p = 10^{14} \text{ s}^{-1}$$

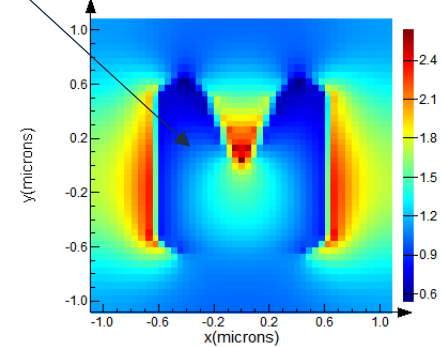
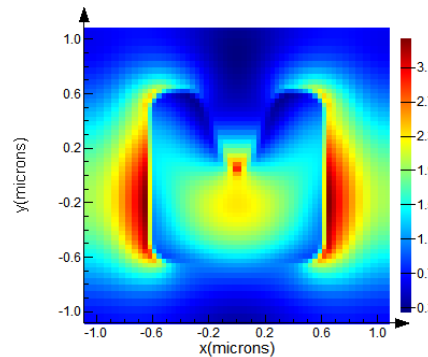
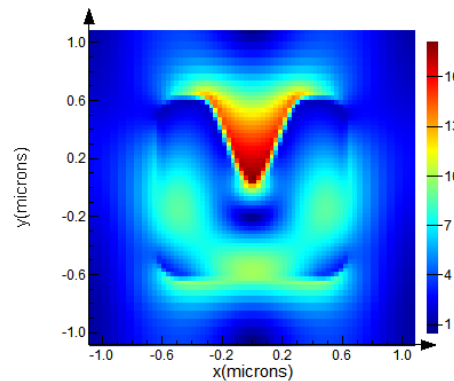
$$\omega_p = 10^{15} \text{ s}^{-1}$$

$$\omega_p = 1.5 \cdot 10^{15} \text{ s}^{-1}$$

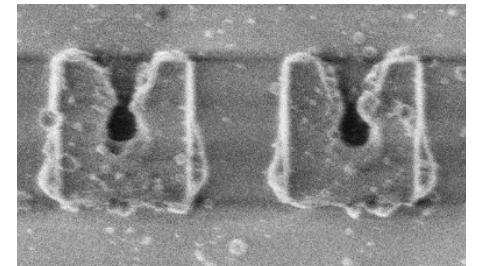
Off resonance



On resonance



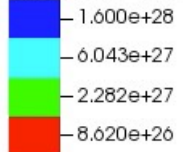
Near-threshold



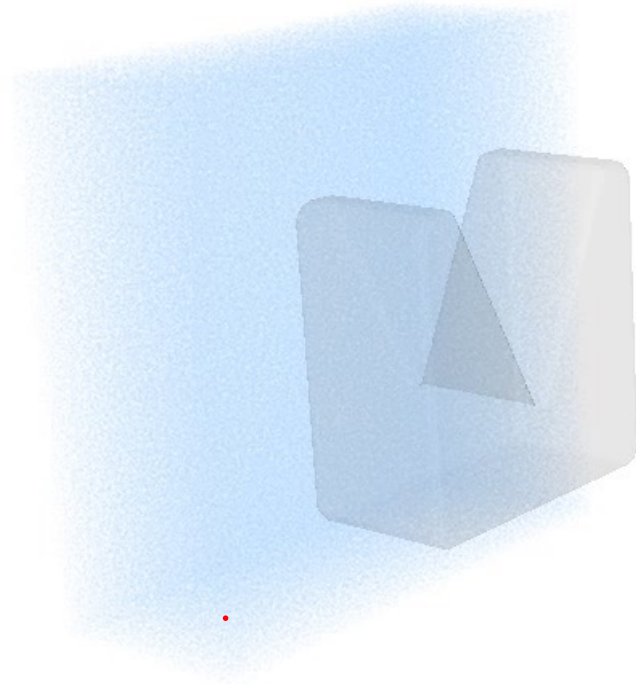
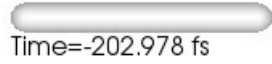


# Particle-in-cell simulations

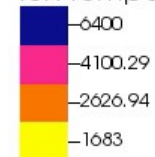
Electron density (1/m<sup>3</sup>)



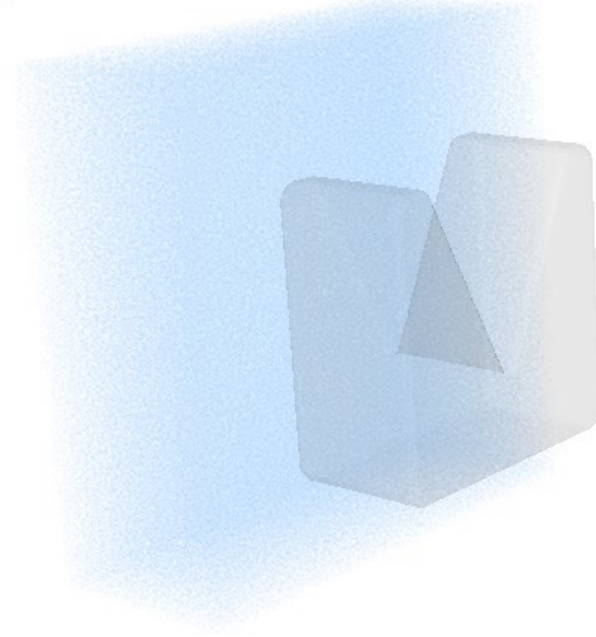
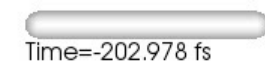
Max: 0.000  
Min: 0.000



Ion temperature (K)



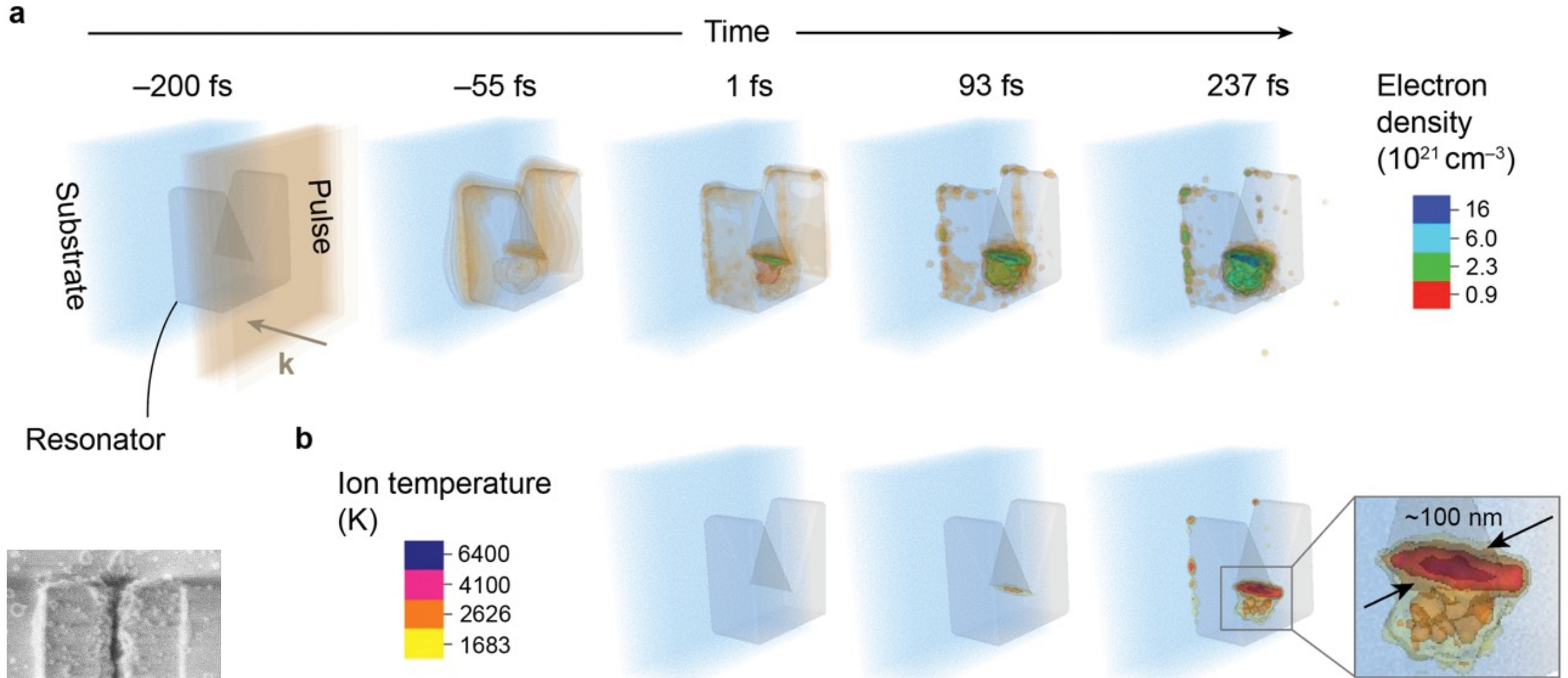
Max: 0.000  
Min: 0.000







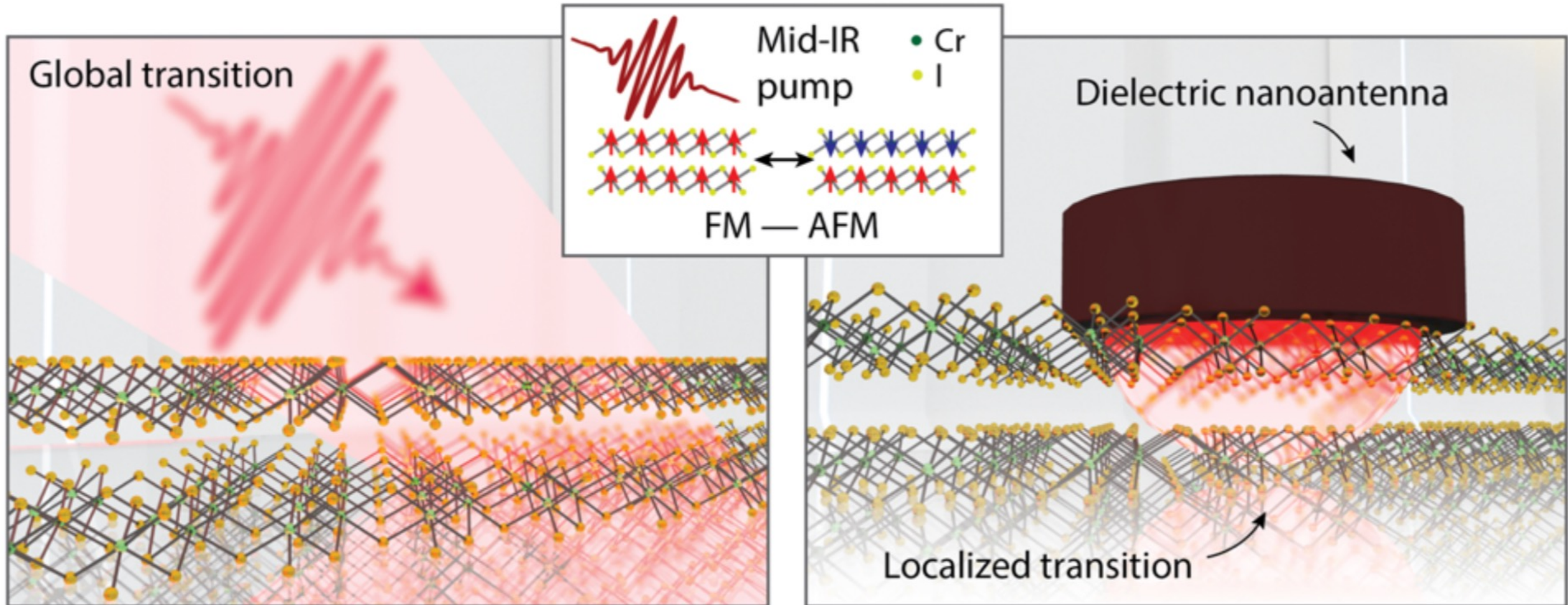
# Particle-in-cell simulations



Trench parameters: Length  $\sim 1.5 \mu\text{m}$ , Width at bottom  $\sim 50 \text{ nm}$ , Depth = 600 nm



# Strong fields + 2D materials

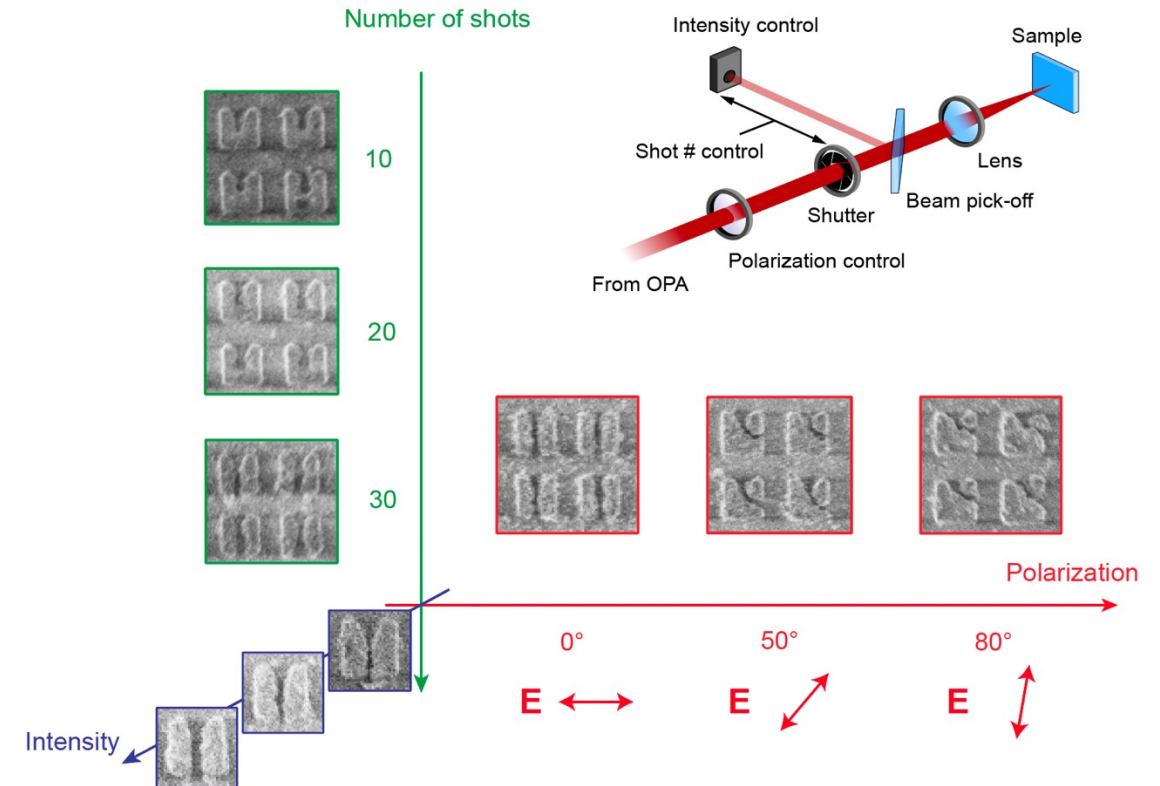
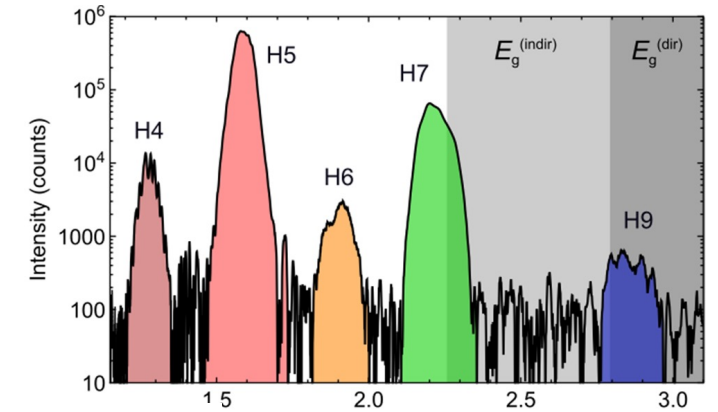
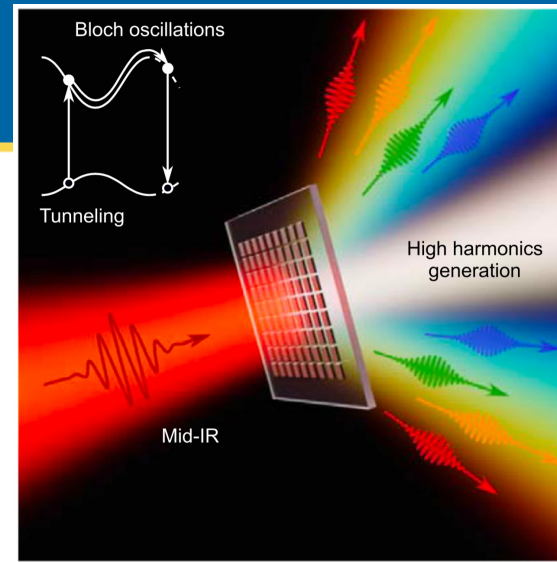


Great outlook to control elementary excitations in low-dimensional materials



# Outline

- Strong fields:  
high harmonics generation
- Very strong fields:  
laser damage and nanomachining



*Nature Communications* **12**, 4185 (2021)

*APL Materials* **9**, 060701 (2021)

*Advanced Optical Materials* **9**, 2100240 (2021)



# Acknowledgements

Shcherbakov Lab, Summer 2022



## Sponsors

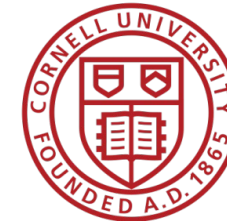


Young Faculty Award



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INSTITUTE OF TECHNOLOGY

## Collaborators



Thank you to collaborators:

Shvets group (Cornell), Brener group (Sandia), Chowdhury group (Ohio State)  
Vuckovic group (Stanford), Kuznetsov group (A\*Star Singapore)