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Advancing Optics and Photonics Worldwide

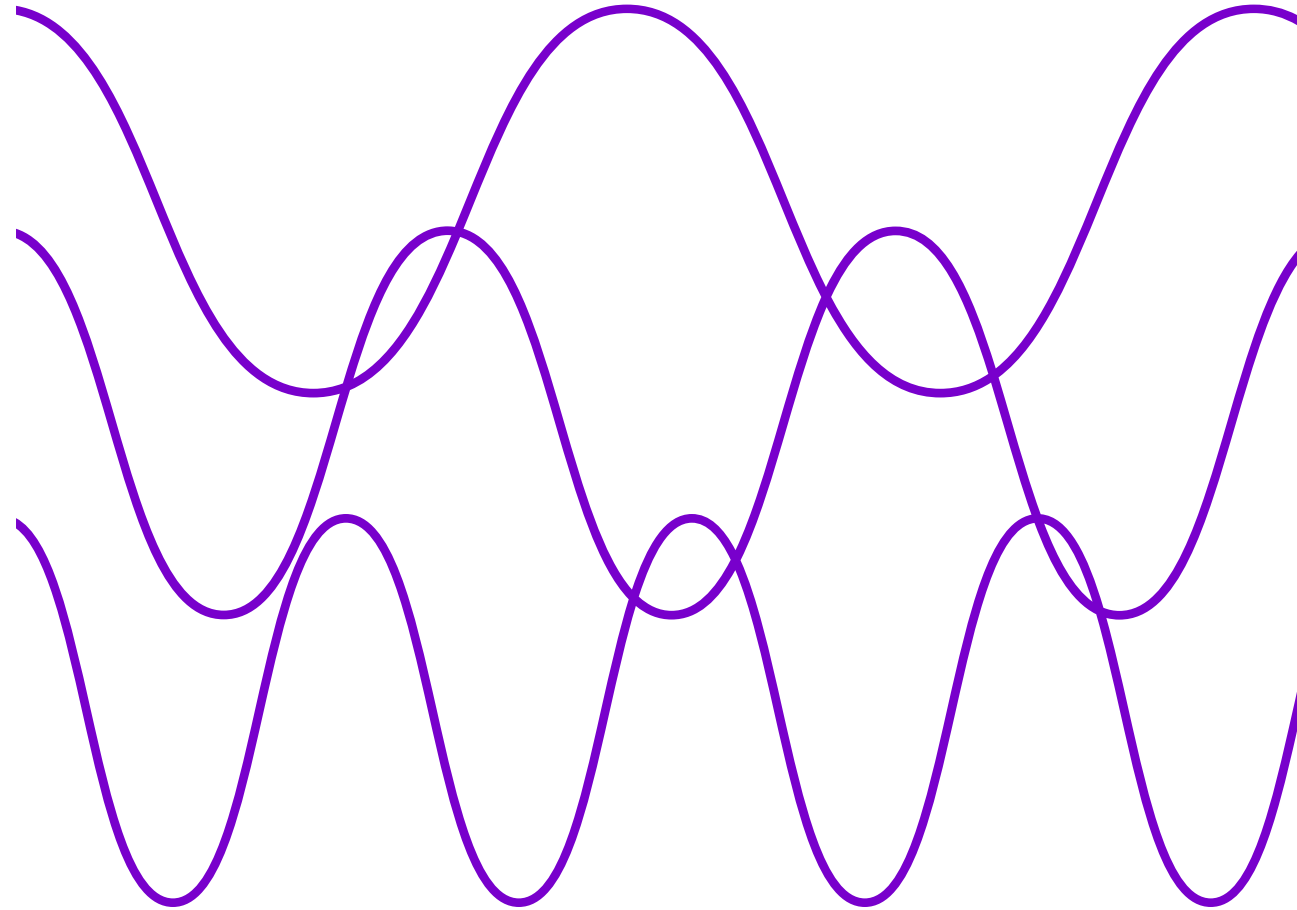
Formerly  
**OSA**

**Nonlinear Optics Technical Group**

# **Time-Variant Systems in Nonlinear Optics: From Frequency Conversion to Beating the Time-Bandwidth Limit**

**Maxim Shcherbakov, University of California, Irvine**

24 September 2021



# Technical Group Executive Committee



**Amol Choudhary**  
*Indian Institute of Technology*



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**Alexander Solntsev**  
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Technical Group**

# About the Nonlinear Optics Technical Group

**Our technical group focuses on the physics of nonlinear optical materials, processes, devices, & applications.**

**Our mission is to connect the 4000+ members of our community through technical events, webinars, networking events, and social media.**

## **Our past activities have included:**

- Webinar on High-order Dispersion Solitons and Topological Photonics in Silicon
- Transitioning into a Career in Optics Panel Discussion at FiO 2019
- Emerging Trends in Nonlinear Optics - A Review of CLEO: 2019
- Emerging Biomedical Applications of Nonlinear Optics

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# Connect with our Technical Group

**Join our online community to stay up to date on our group's activities. You also can share your ideas for technical group events or let us know if you're interested in presenting your research.**

## **Ways to connect with us:**

- Our website at [www.osa.org/ol](http://www.osa.org/ol)
- On LinkedIn at [www.linkedin.com/groups/8302249](http://www.linkedin.com/groups/8302249)
- On Facebook at [www.facebook.com/opticanonlineoptics](http://www.facebook.com/opticanonlineoptics)
- Email us at [TGactivities@osa.org](mailto:TGactivities@osa.org)

# Today's Speaker



## Maxim Shcherbakov

*University of California, Irvine*

- Assistant professor with the Department of Electrical Engineering and Computer Science at the University of California, Irvine
- Received his Ph.D. in Physics from Lomonosov Moscow State University, Russia
- Joined Cornell University as a postdoctoral associate in 2016
- Main interests are artificial optical materials and their nonlinear and quantum optics applications, deep-subwavelength lithography, and augmented and mixed reality devices

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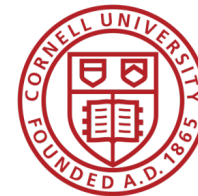
# Time-Variant Systems in Nonlinear Optics: From Frequency Conversion to Beating the Time-Bandwidth Limit



Maxim Shcherbakov

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

Sponsors and collaborators:

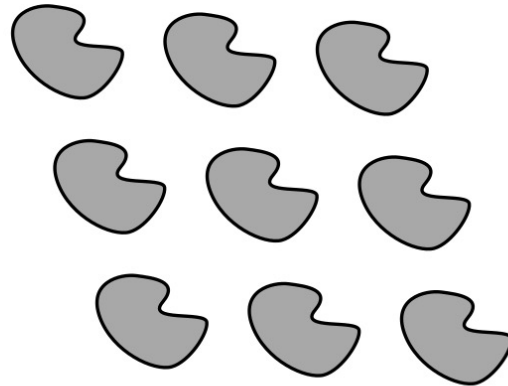


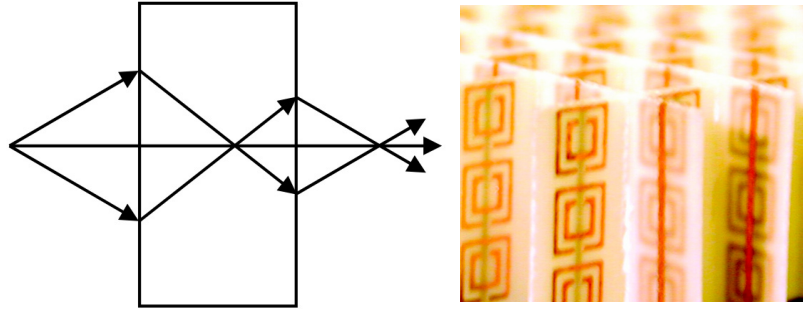
With groups of  
Gennady Shvets (Cornell)  
Igal Brenner (Sandia)  
Hayk Harutyunyan (Emory)  
Enam Chowdhury (Ohio State)

Nonlinear Optics Technical Group Webinar  
September 24, 2021



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

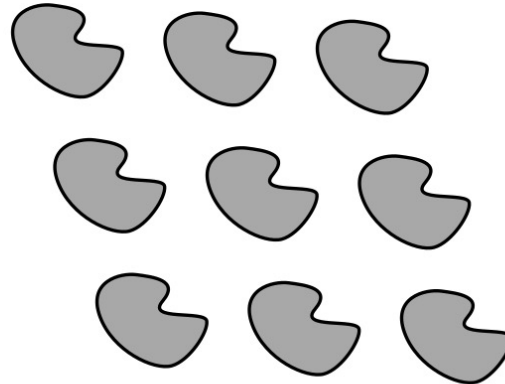




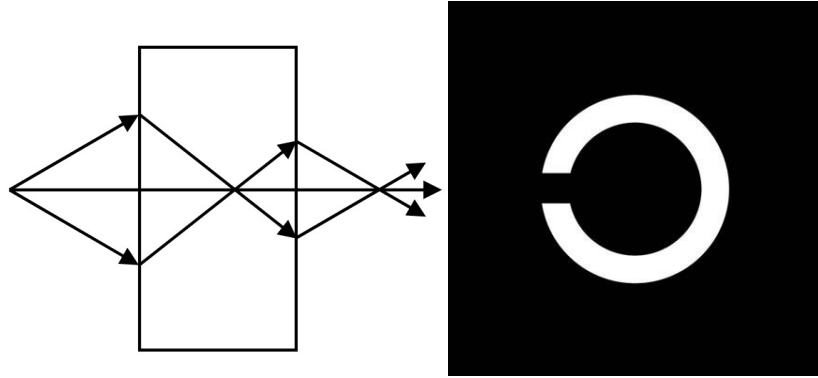
## Negative refraction and LHM

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

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



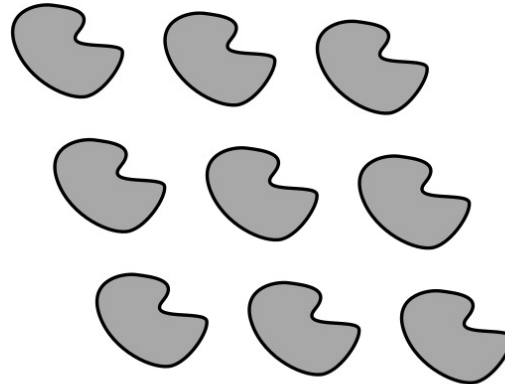


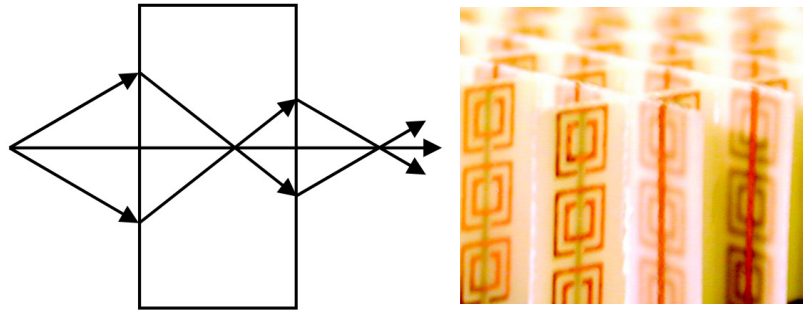


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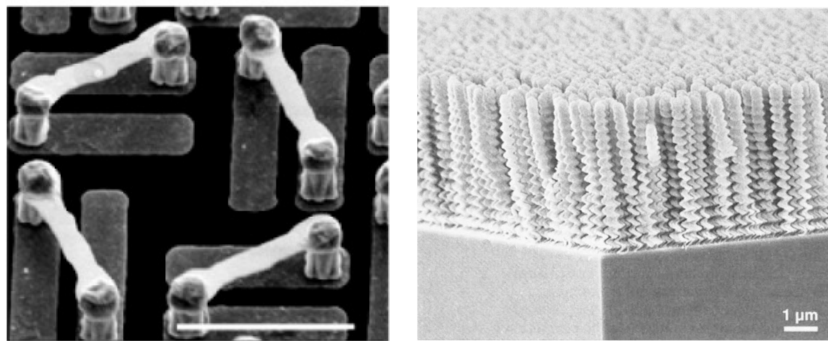
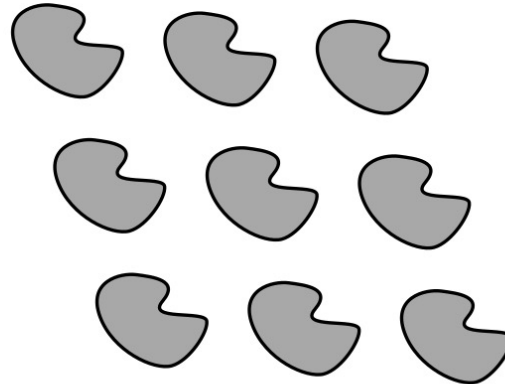




## Negative refraction and LHM

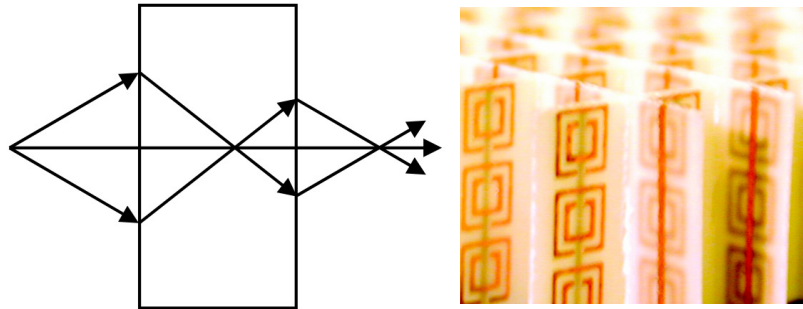
Veselago, Pendry, Shelby, Smith, Schultz, Lezec, Shalaev,  
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$$\mathbf{P}(\mathbf{r}) = \epsilon_0 \chi(\mathbf{r}) \mathbf{E}(\mathbf{r})$$



## Polarization and chirality

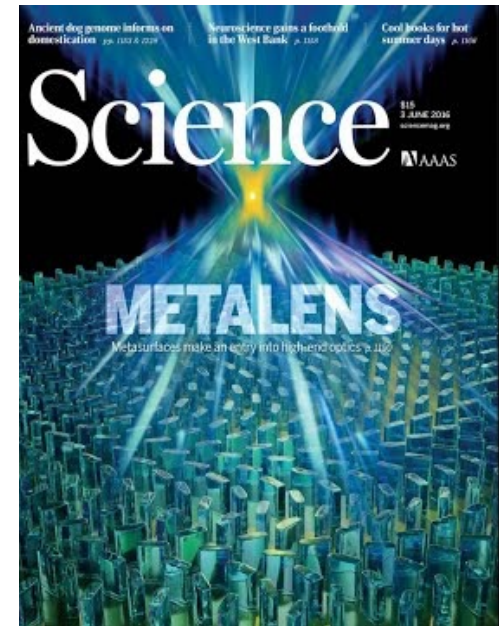
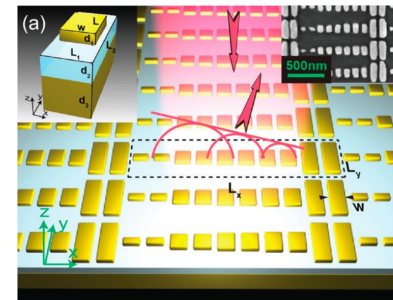
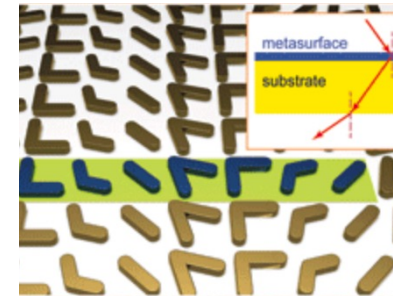
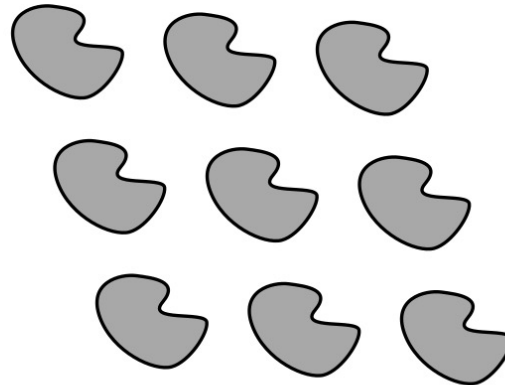
Pendry, Lakhtakia, X Zhang, Zheludev, He, Wegener, Pertsch,  
Soukoulis, Ozbay, HT Chen, S Zhang



## Negative refraction and LHM

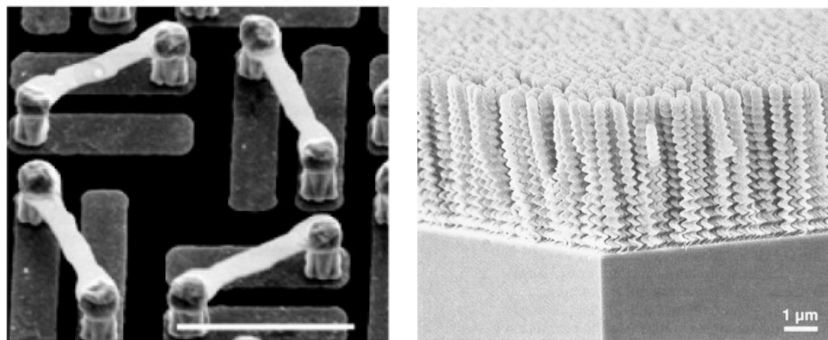
Veselago, Pendry, Shelby, Smith, Schultz, Lezec, Shalaev, X Zhang, Soukoulis, Sihvola, Tretyakov, Fan

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



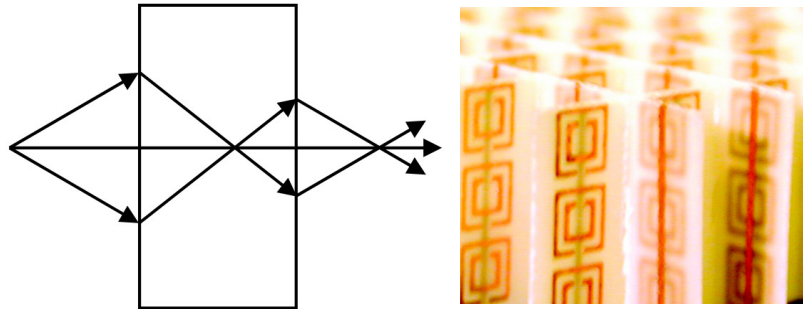
## Metasurface-based devices

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



## Polarization and chirality

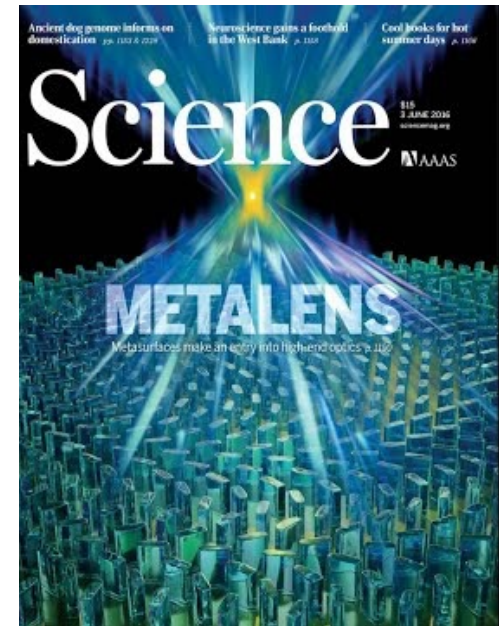
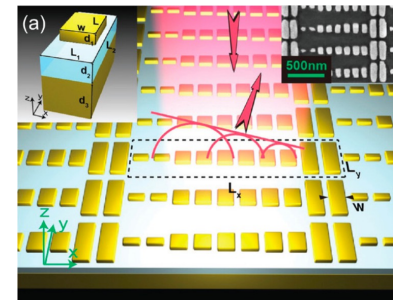
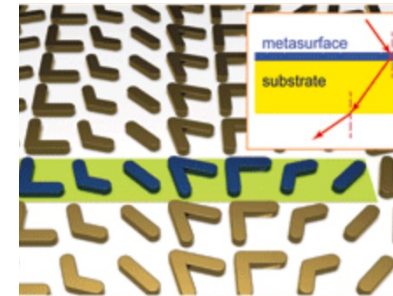
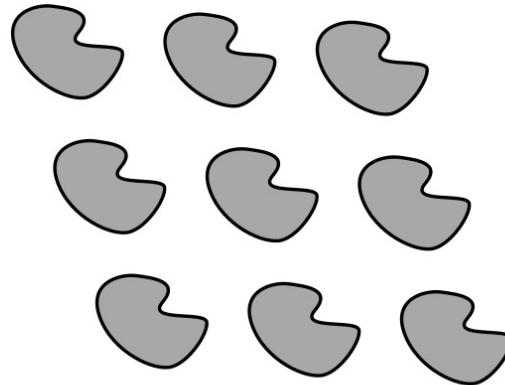
Pendry, Lakhtakia, X Zhang, Zheludev, He, Wegener, Pertsch, Soukoulis, Ozbay, HT Chen, S Zhang



## Negative refraction and LHM

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

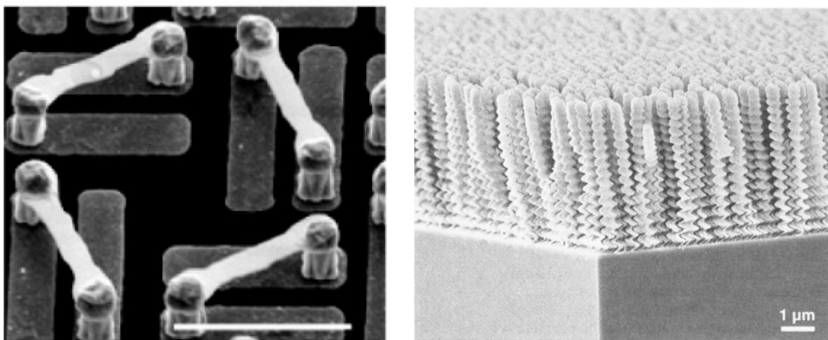
$$\mathbf{P}(\mathbf{r}) = \epsilon_0 \chi(\mathbf{r}) \mathbf{E}(\mathbf{r})$$



## Metasurface-based devices

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

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



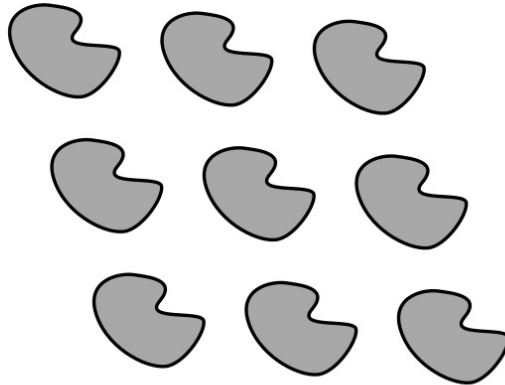
## Polarization and chirality

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



# Metamaterials: tailored nonlinear and spatio-temporal response

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

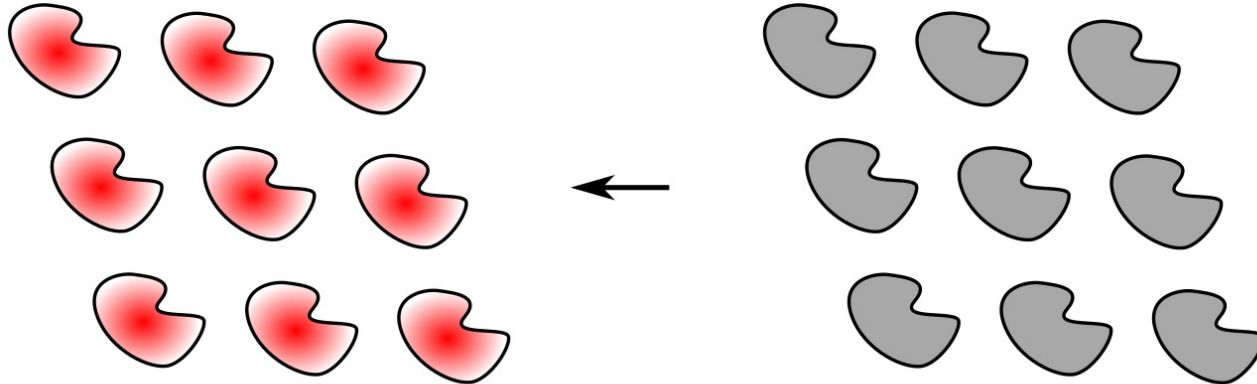




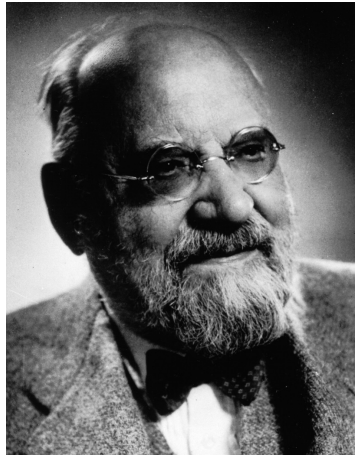
# Metamaterials: tailored nonlinear and spatio-temporal response

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

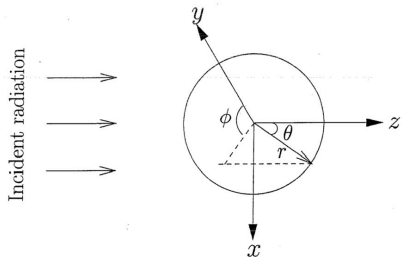
$$\mathbf{P}(\mathbf{r}) = \varepsilon_0 \chi(\mathbf{r}) \mathbf{E}(\mathbf{r})$$



Nonlinear metamaterials



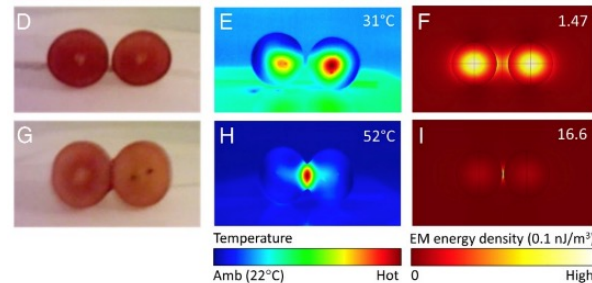
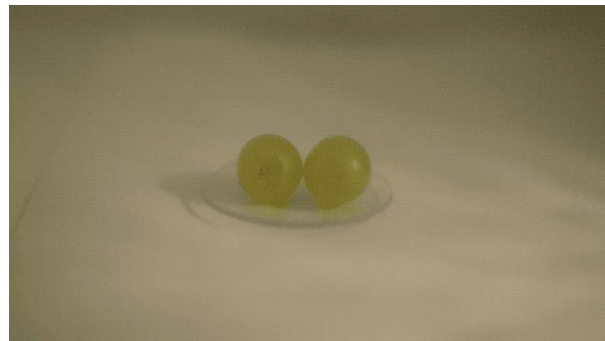
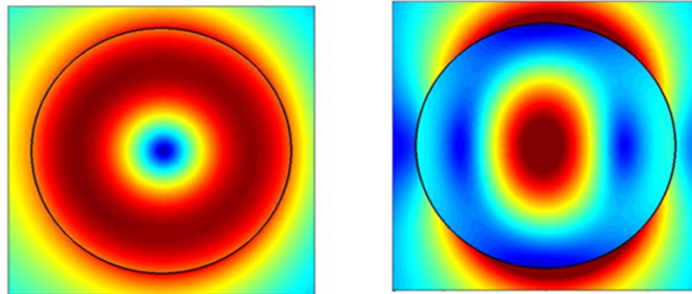
Gustav Mie  
(1869 – 1957)



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

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

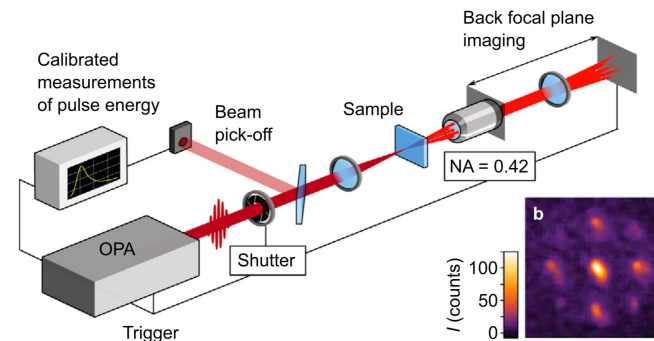
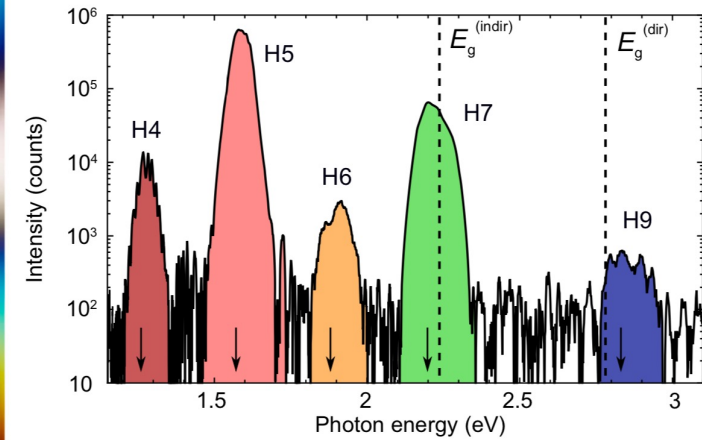
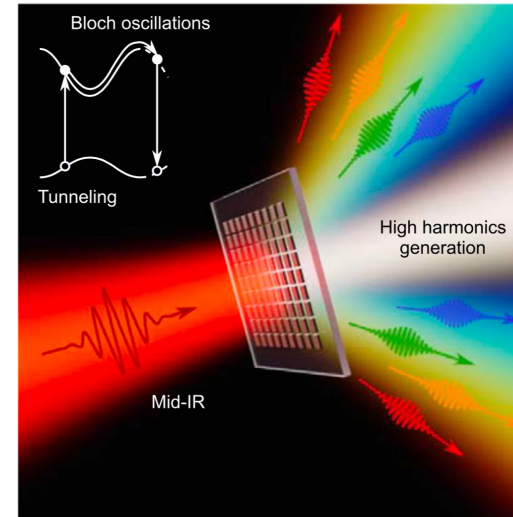
(1) Magnetic dipolar (2) Electric dipolar



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

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

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



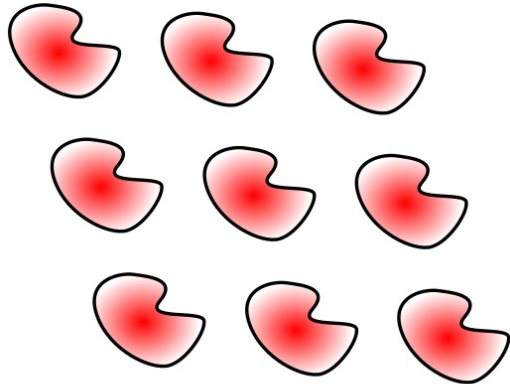
Record-breaking  
conversion from an  
ultrathin material

Shcherbakov et al.,  
*Nat. Commun.* **10**, 1345 (2019)



# Metamaterials: tailored nonlinear and spatio-temporal response

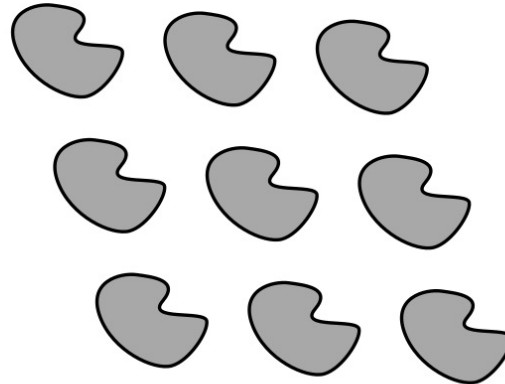
$$\mathbf{P}(\mathbf{r}) = \varepsilon_0 \sum \chi^{(n)}(\mathbf{r}) \mathbf{E}^n(\mathbf{r})$$



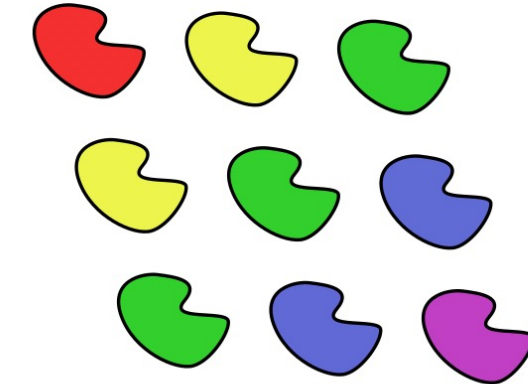
Nonlinear metamaterials

- 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)

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



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



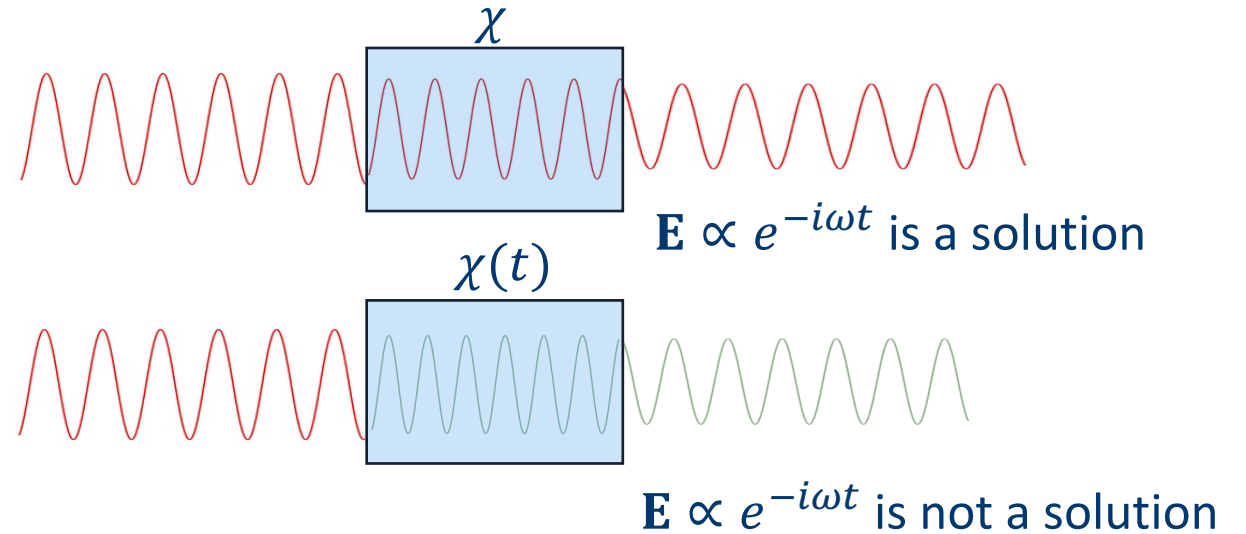
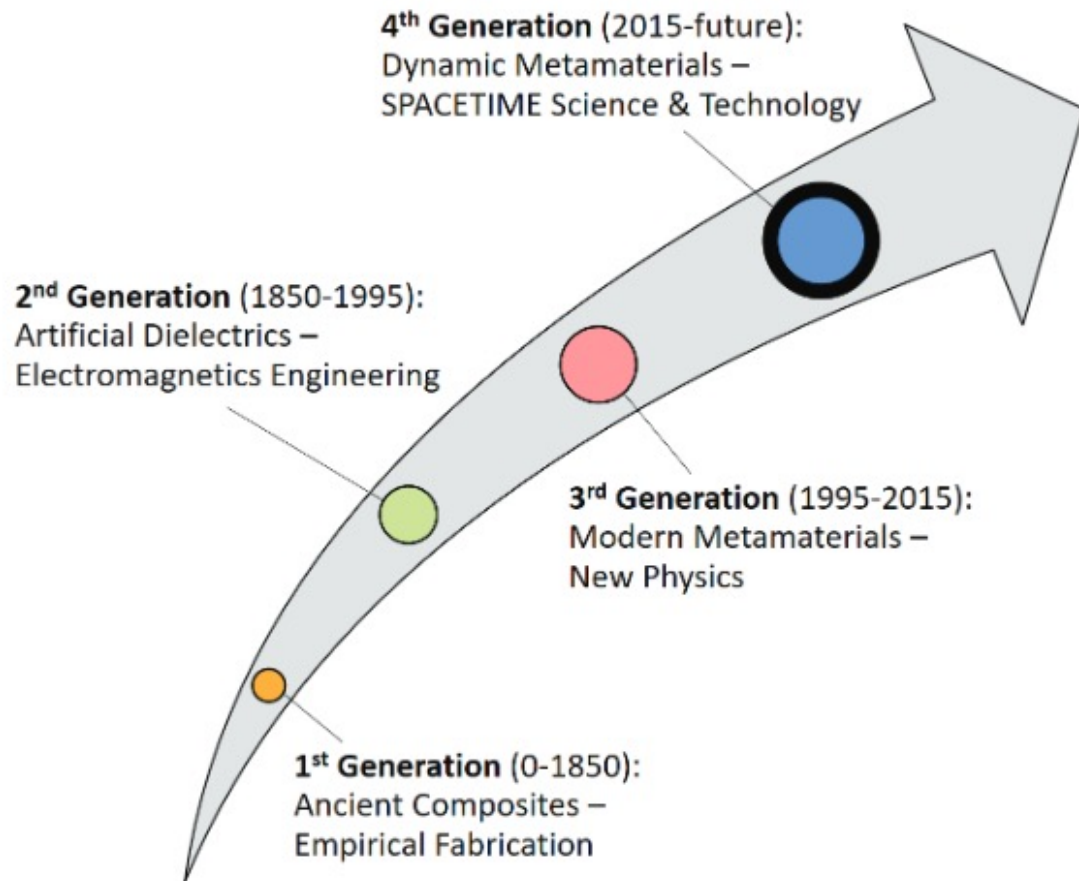
Time-variant metamaterials

- 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



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

Caloz and Deck-Leger, *IEEE Trans Ant Propag* **68**, 1569 (2020)

Focus of this talk:  
Aperiodic modulation in resonators



## Time-variant semiconductor metasurfaces. Outline

- Frequency conversion
- Breaking the time-bandwidth limit
- Discussion: Time-variant  $\epsilon$  nonlinear?
- Conclusion

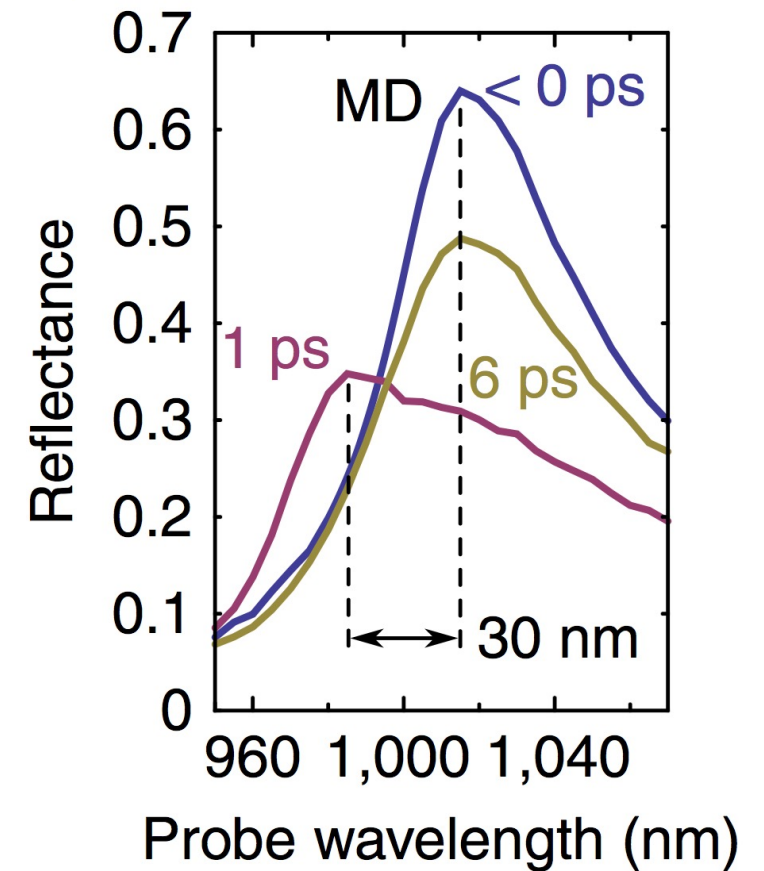
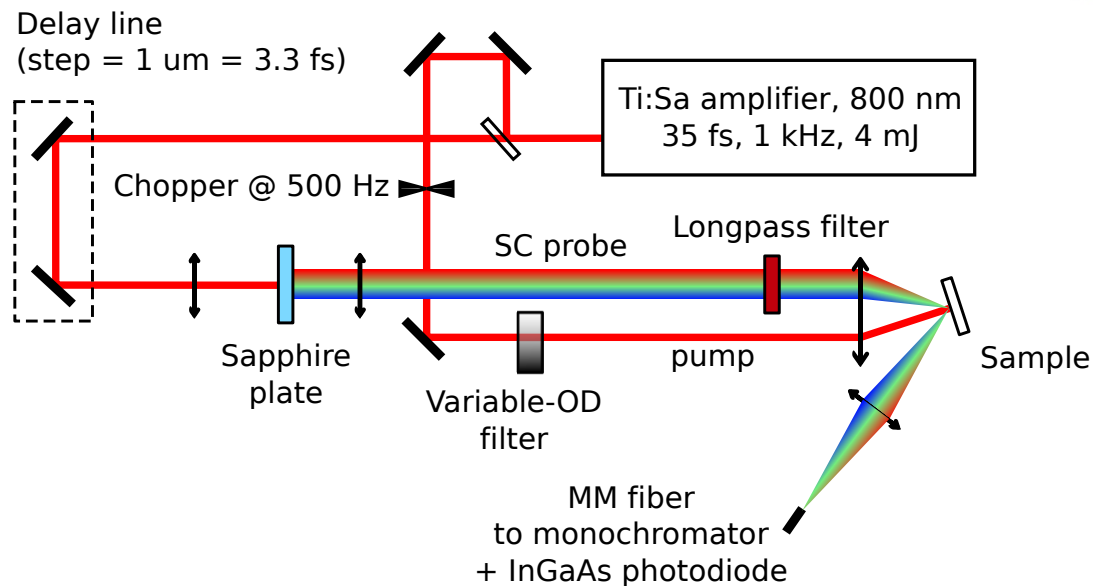
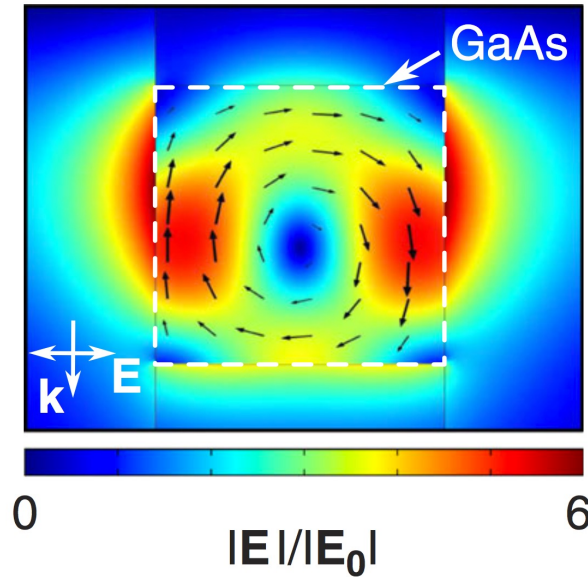
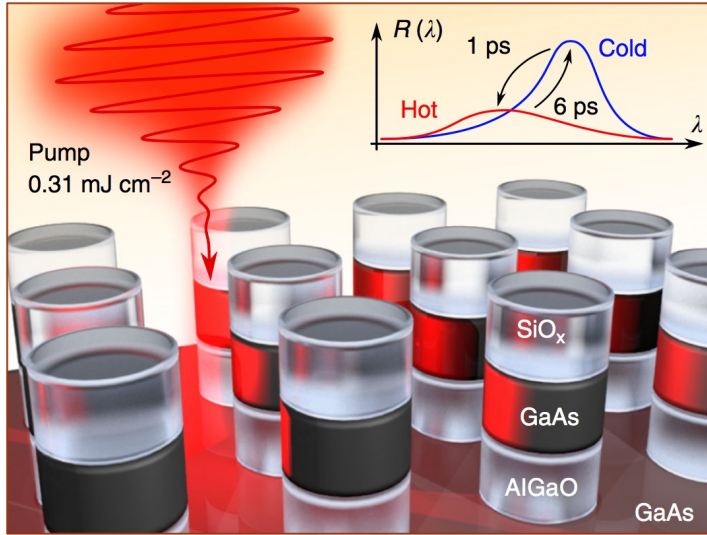


## Time-variant semiconductor metasurfaces. Outline

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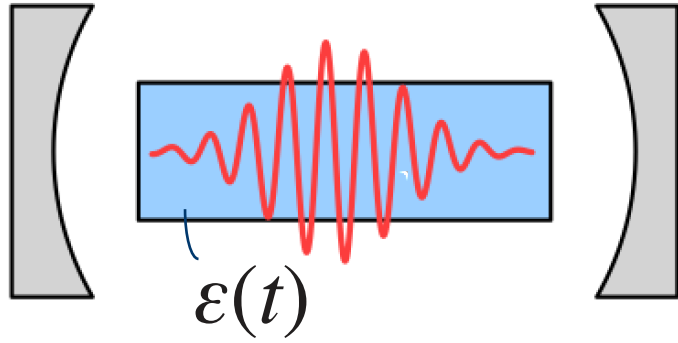


# Time-variant metasurfaces – how to?



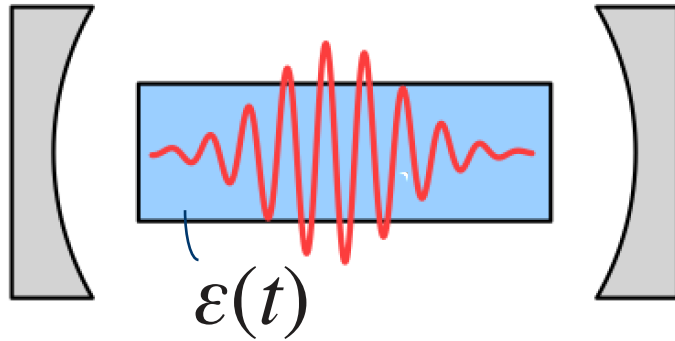


# Light in time-modulated cavities





# Light in time-modulated cavities



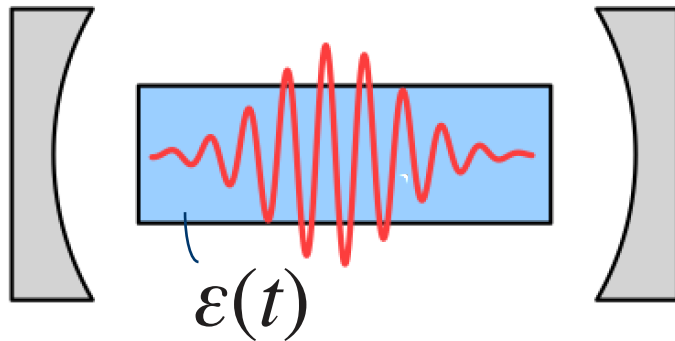
$$\ddot{a} + \frac{\dot{\epsilon} + \sigma}{\epsilon} \dot{a} + \frac{k^2}{\epsilon \mu_0} a = 0$$

Time-dependent  
damping

Time-dependent  
frequency



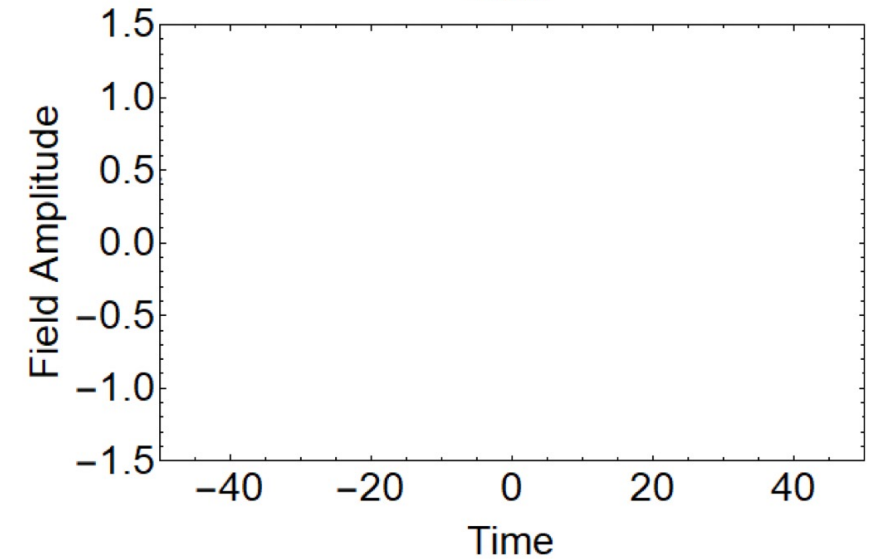
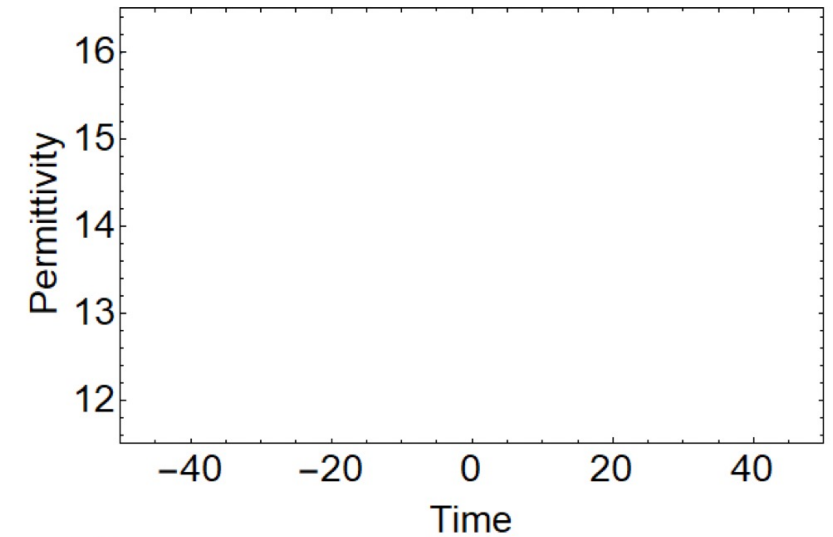
# Light in time-modulated cavities



$$\ddot{a} + \frac{\dot{\epsilon} + \sigma}{\epsilon} \dot{a} + \frac{k^2}{\epsilon \mu_0} a = 0$$

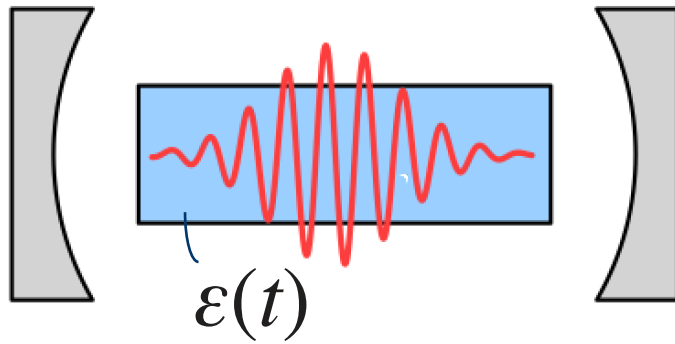
Time-dependent  
damping

Time-dependent  
frequency





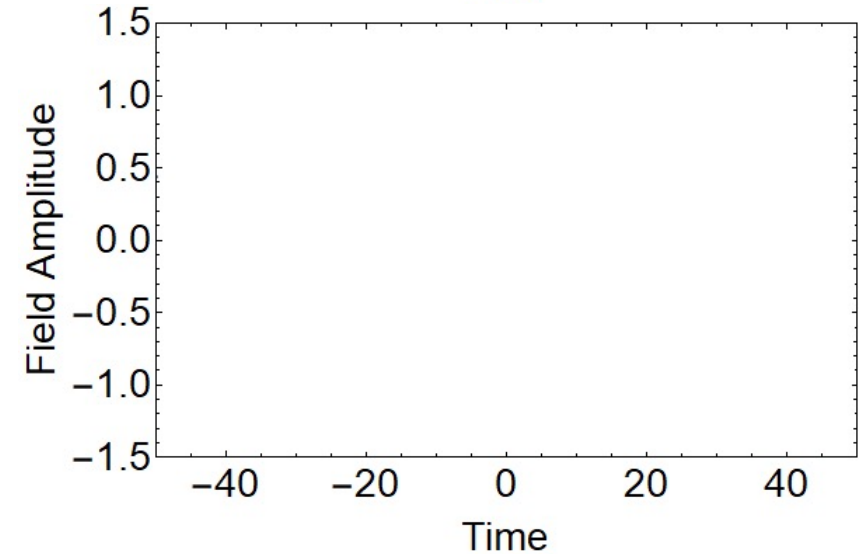
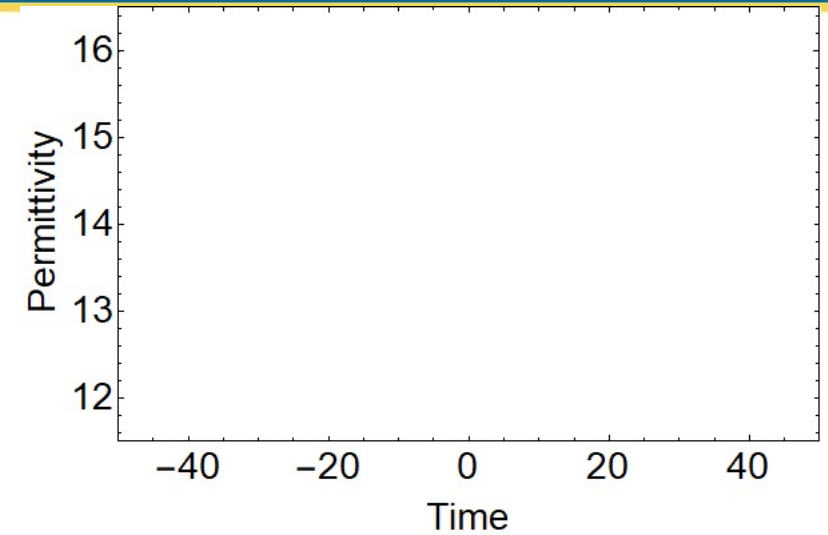
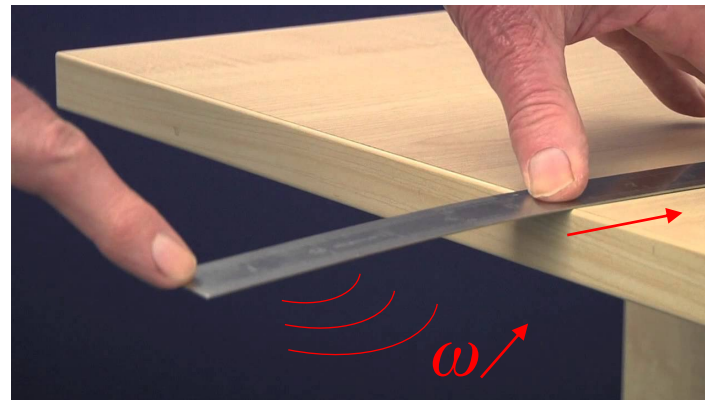
# Light in time-modulated cavities



$$\ddot{a} + \frac{\dot{\epsilon} + \sigma}{\epsilon} \dot{a} + \frac{k^2}{\epsilon \mu_0} a = 0$$

Time-dependent  
damping

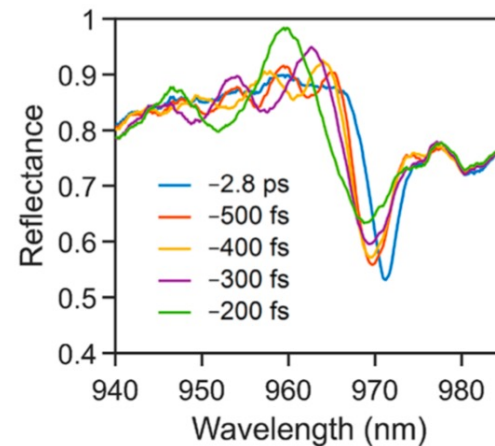
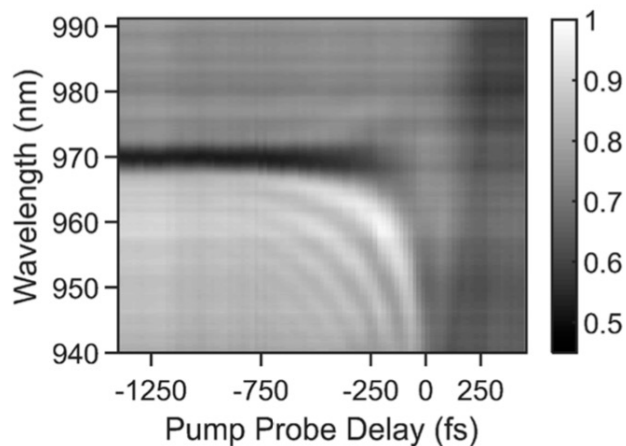
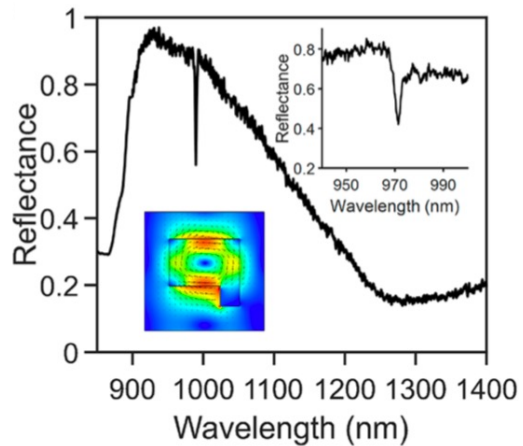
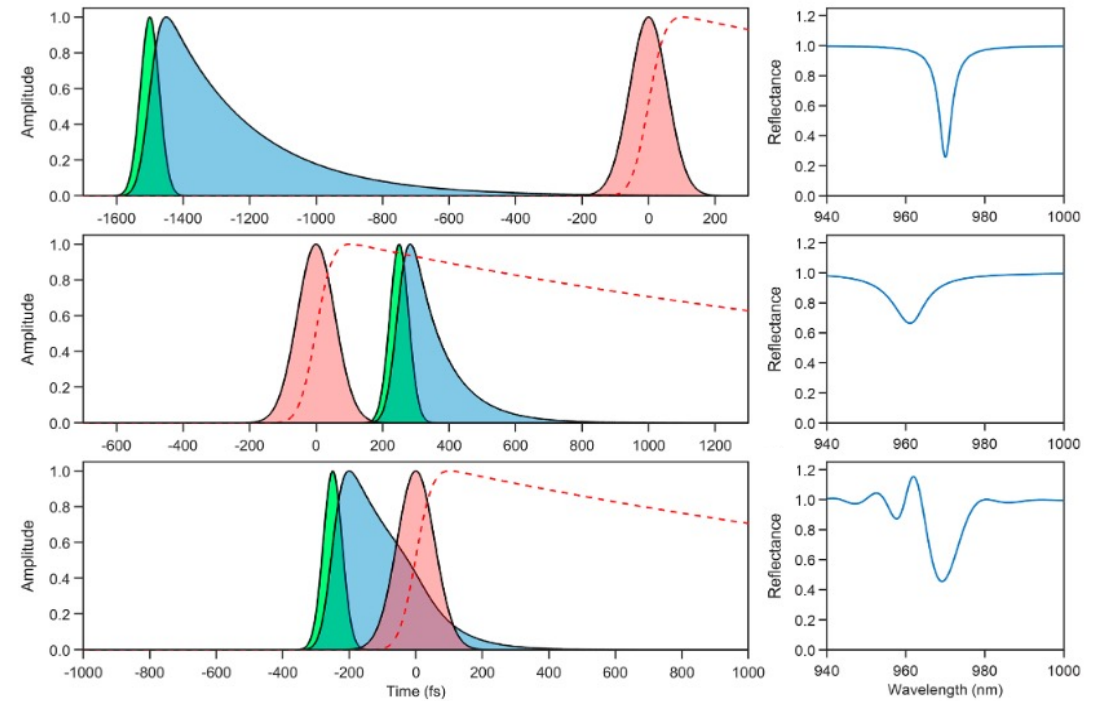
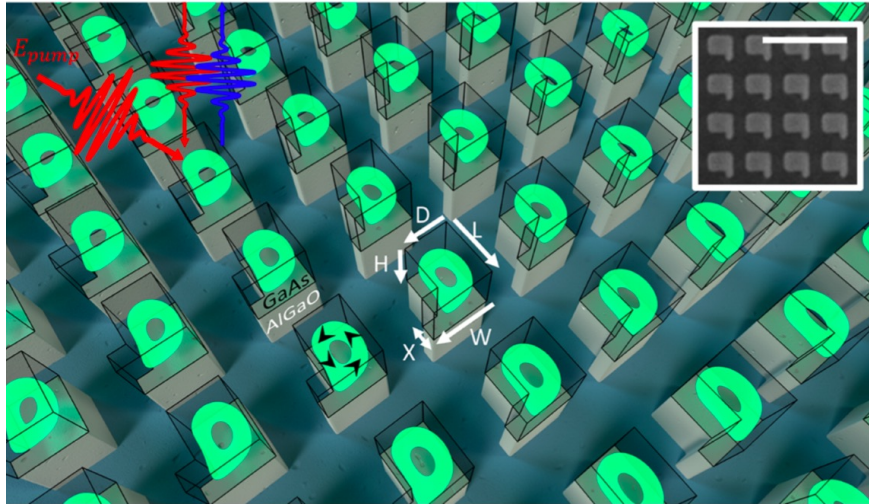
Time-dependent  
frequency







# Tuning the color of light



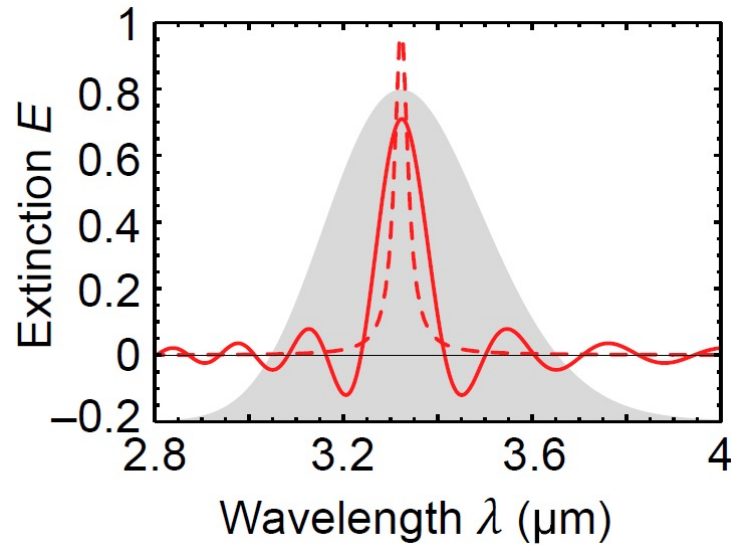
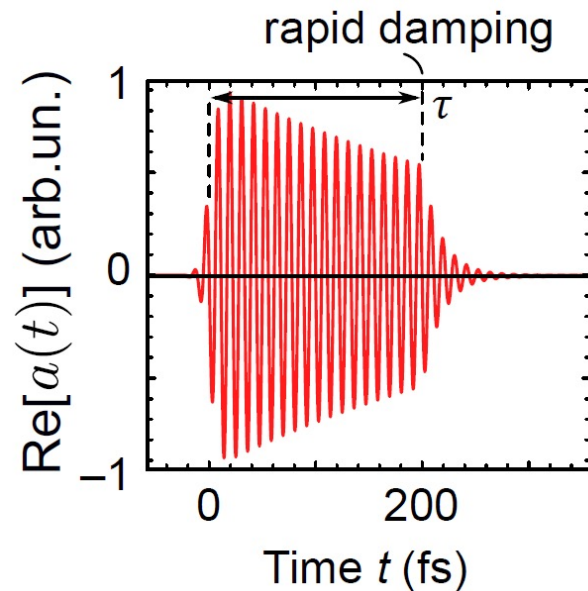
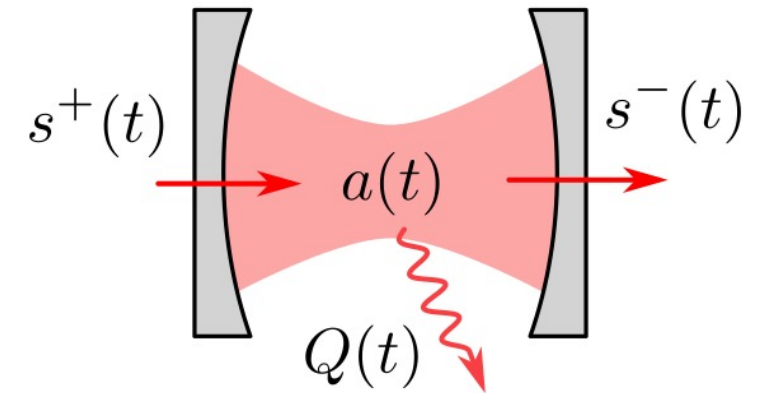
Early demonstrations:  
Lipson, Muskens, Agrawal,  
Tanabe/Kuramochi, others

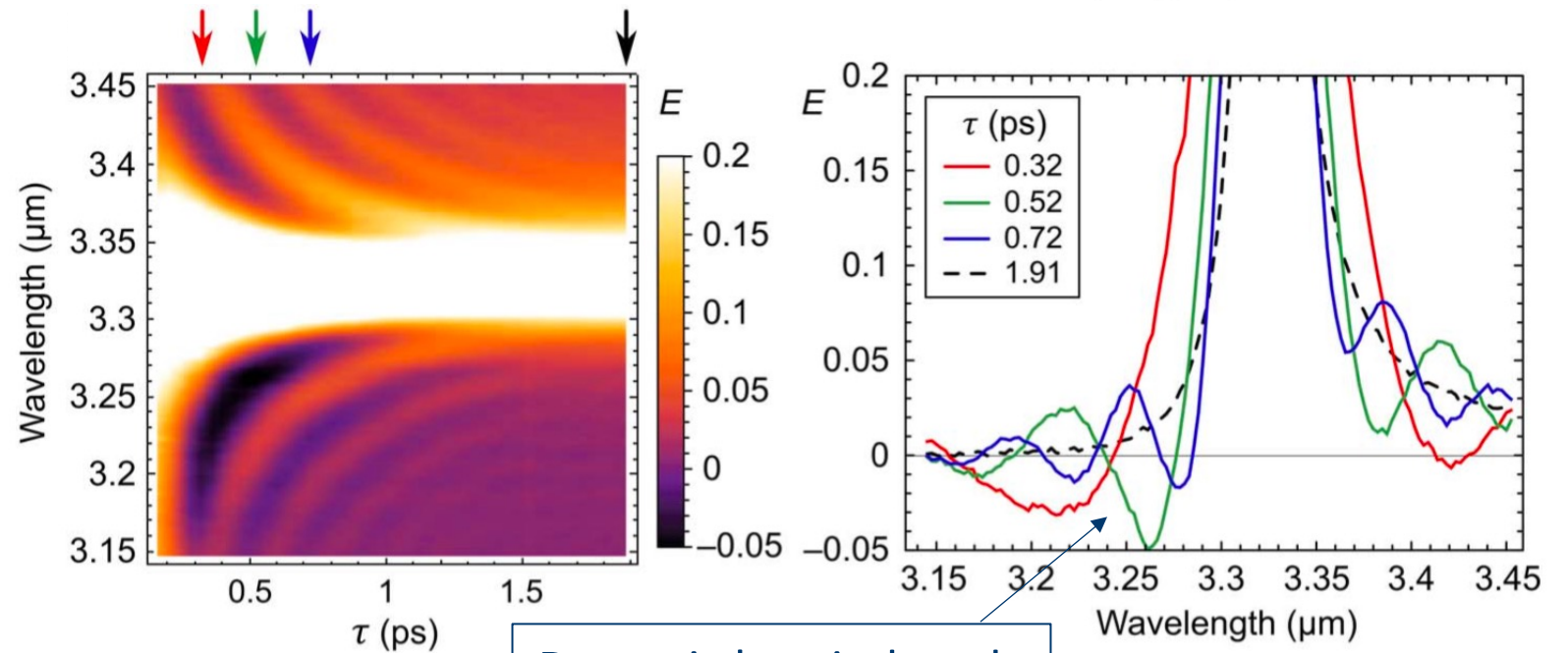
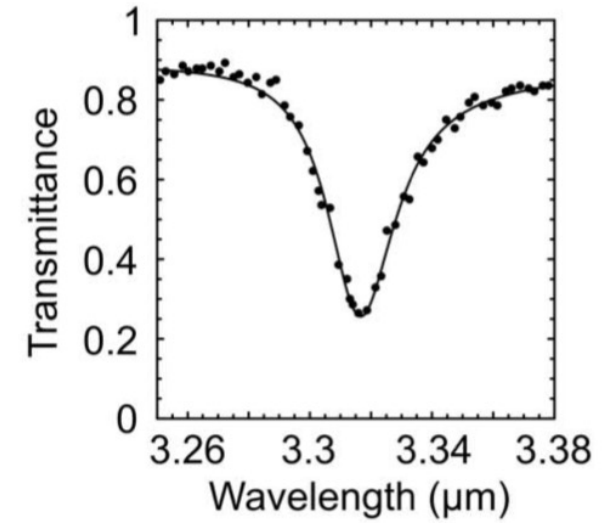
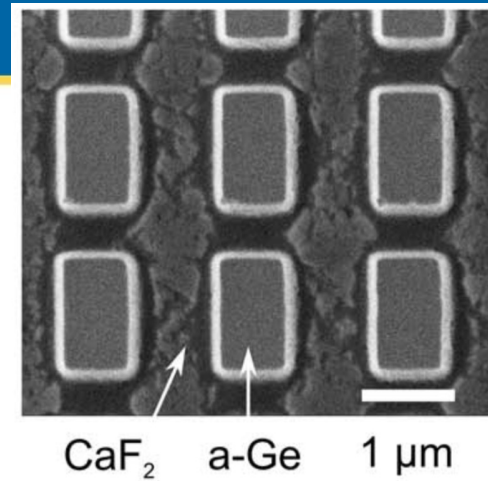
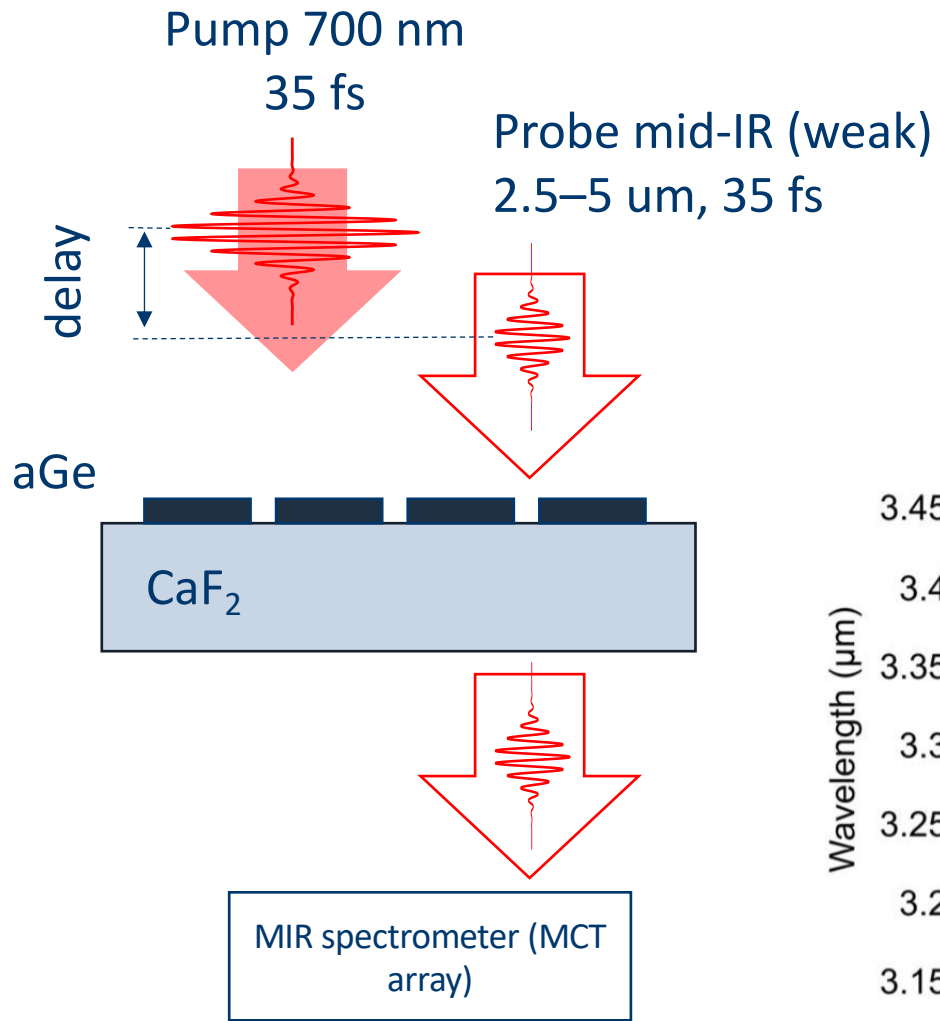


# Frequency conversion by dynamic losses

$$\dot{a}(t) + i\omega_0 a(t) + [\gamma_r + \gamma_{nr}(t)]a(t) = \sqrt{\gamma_r} s^+(t),$$

$$s^-(t) = s^+(t) - \sqrt{\gamma_r} a(t)$$





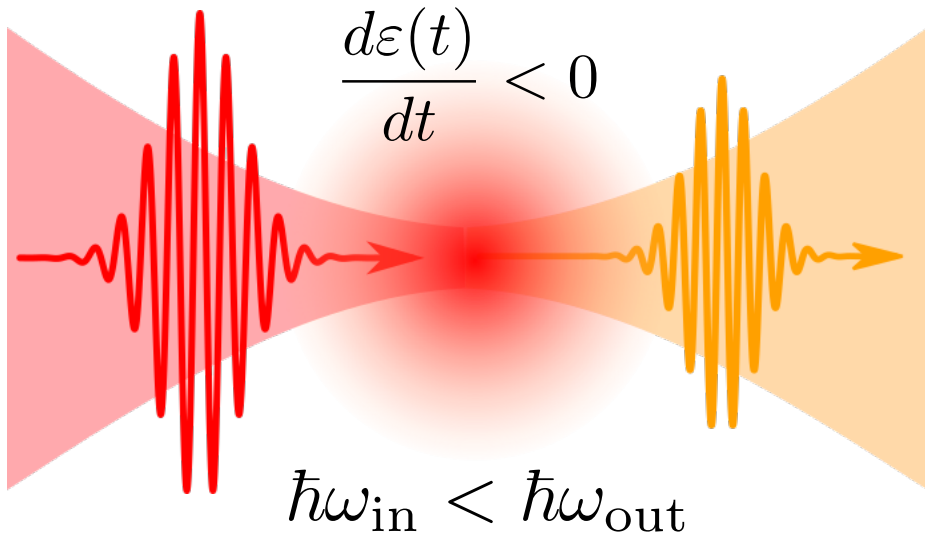
Dynamic loss-induced negative extinction

Shcherbakov et al., *Optica* **6**, 1441 (2019)

See also: Bruck et al., *Nat. Photonics* **8**, 54 (2014) – microresonators

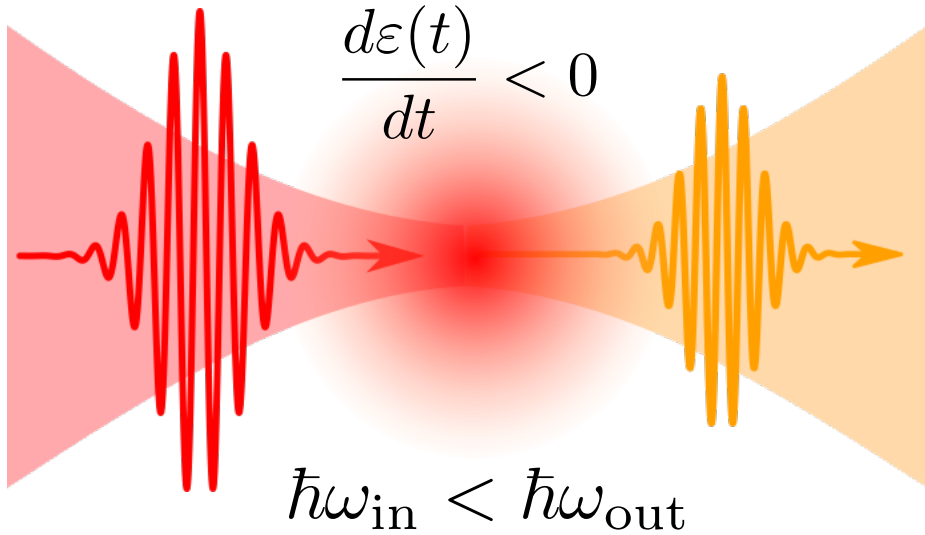


# Photon acceleration by dynamic plasma





# Photon acceleration by dynamic plasma



Theory:

[1] L. B. Felsen and G. M. Whitman, IEEE Trans. Antennas Propag. AP-18, 242 (1970).

Experiments:

[2] S. Kuo, Phys. Rev. Lett. **65**, 1000 (1990).

[3] C. Joshi, C. Clayton, K. Marsh, D. Hopkins, A. Sessler, and D. Whittum, IEEE Trans. Plasma Science **18**, 814 (1990).

[4] E. Yablonovitch, Phys. Rev. Lett. **31**, 877 (1973).

[5] V. Mironov, A. Sergeev, E. Vanin, G. Brodin, and J. Lundberg, Phys. Rev. A **46**, 6178 (1992).

[6] B. M. Penetrante, J. N. Bardsley, W. M. Wood, C. W. Siders, and M. C. Downer, J. Opt. Soc. Am. B **9**, 2032 (1992).

[7] W. Wood, C. Siders, and M. Downer, Phys. Rev. Lett. **67**, 3523 (1991).

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29 MAY 1989

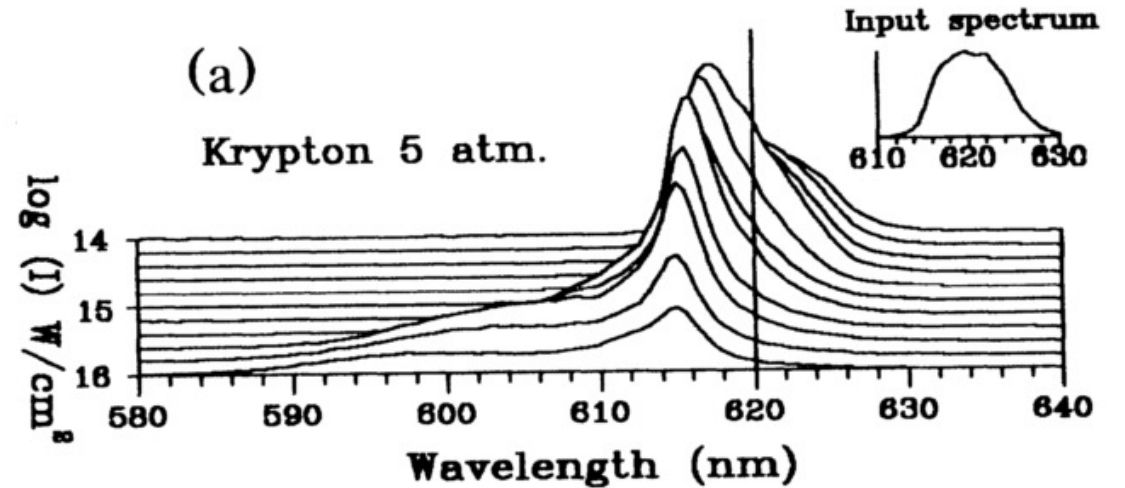
## Photon Accelerator

S. C. Wilks,<sup>(1)</sup> J. M. Dawson,<sup>(1)</sup> W. B. Mori,<sup>(1)</sup> T. Katsouleas,<sup>(1)</sup> and M. E. Jones<sup>(2)</sup>

<sup>(1)</sup>Department of Physics, University of California, Los Angeles, California 90024

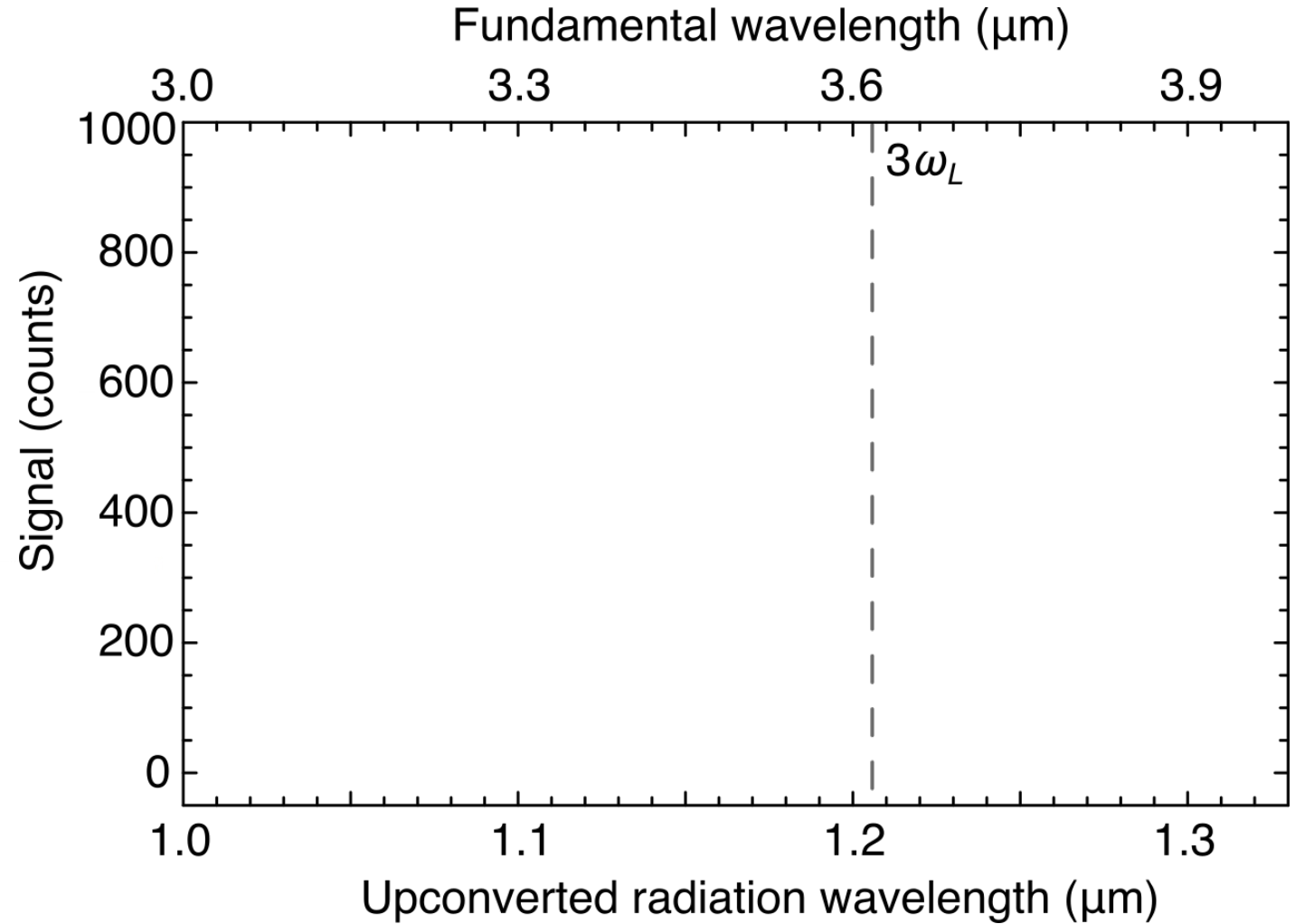
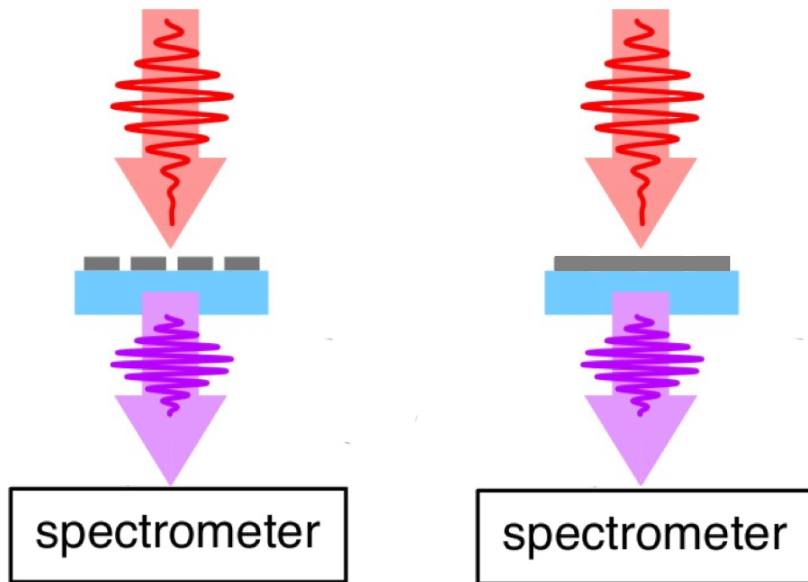
<sup>(2)</sup>Los Alamos National Laboratory, Los Alamos, New Mexico 87545

(Received 22 February 1989)



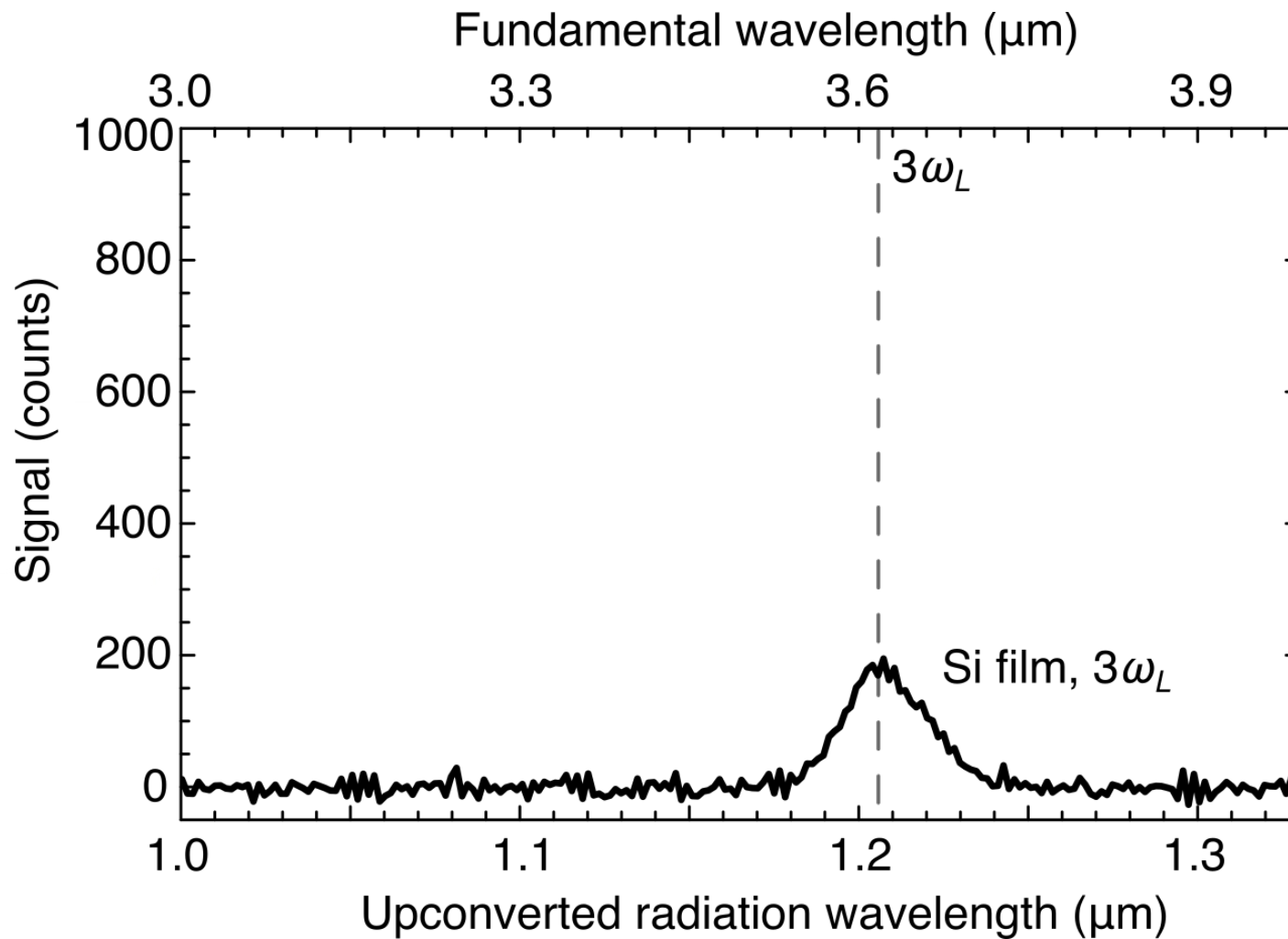
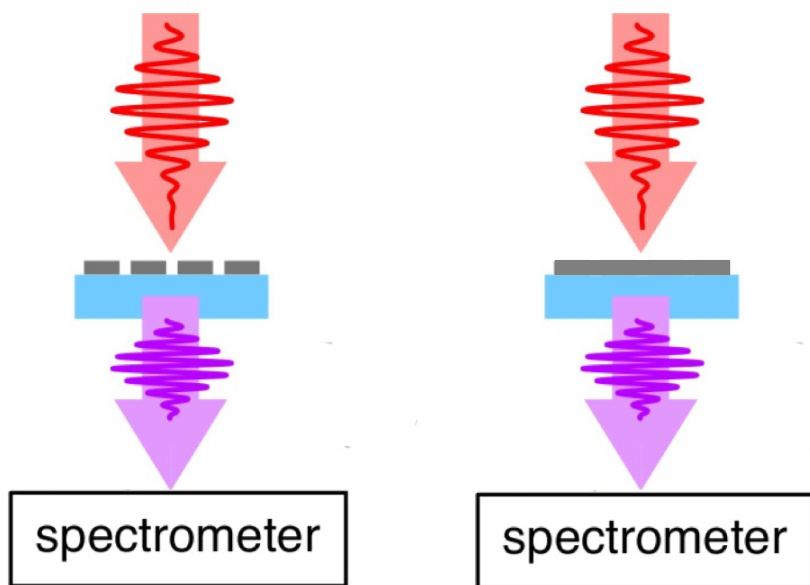


# Photon acceleration probed by THG



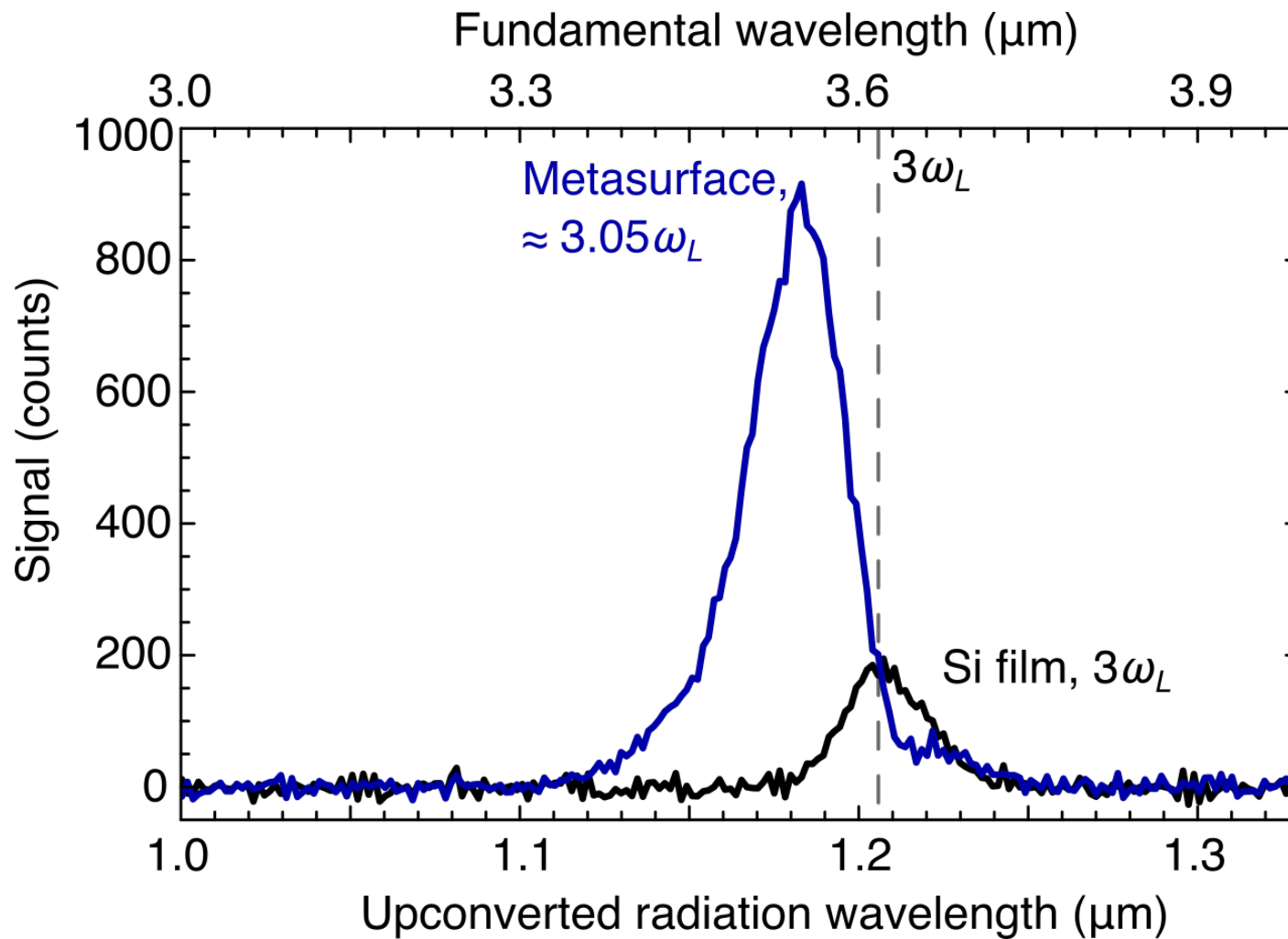
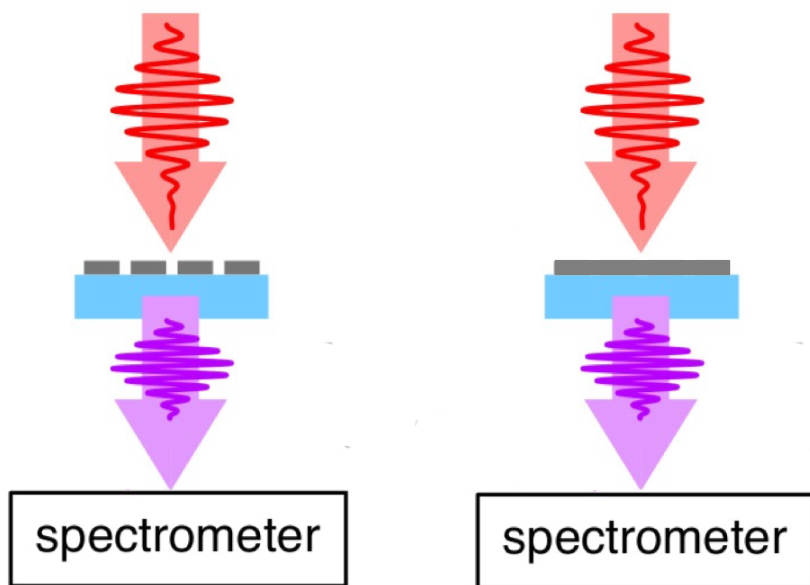


# Photon acceleration probed by THG





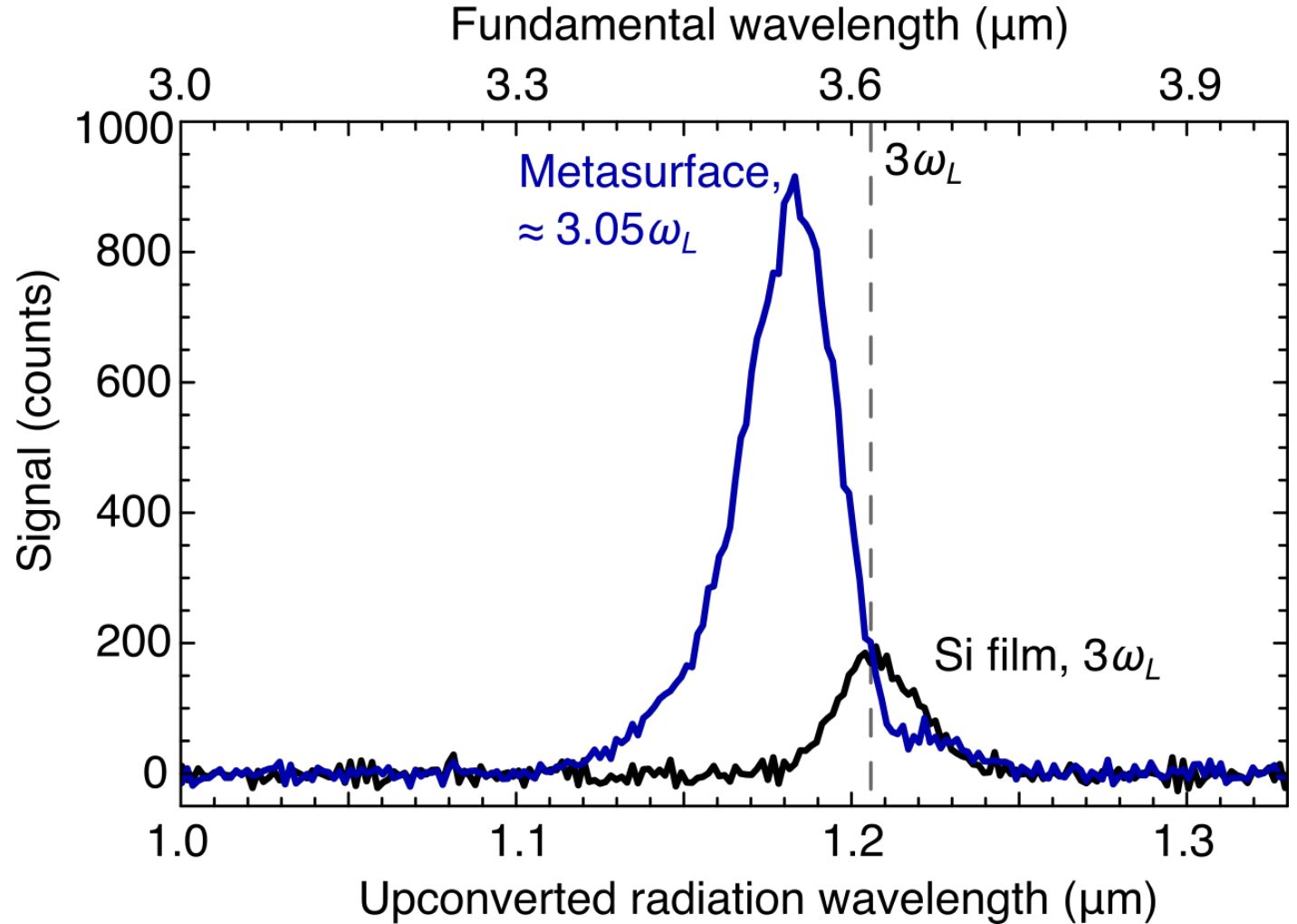
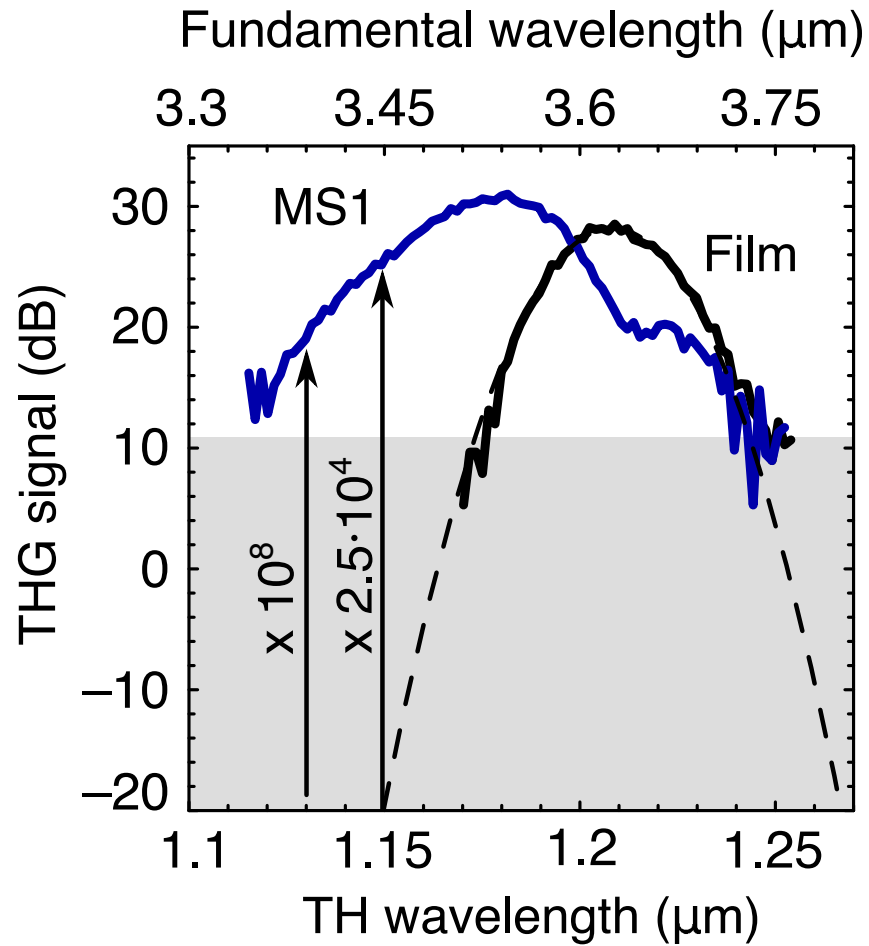
# Photon acceleration probed by THG





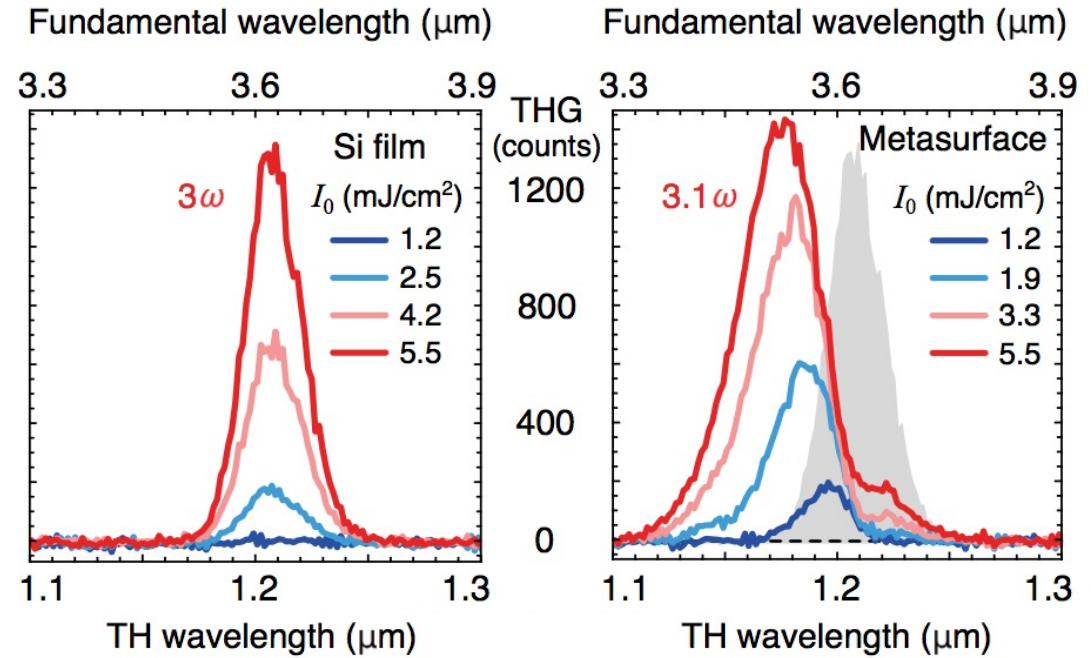
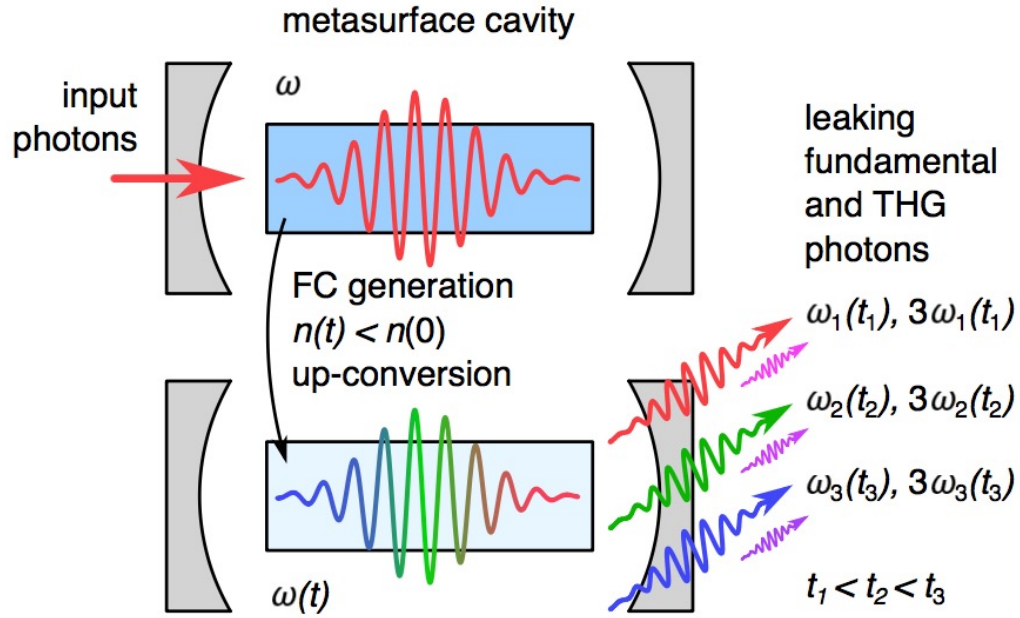


# Photon acceleration probed by THG

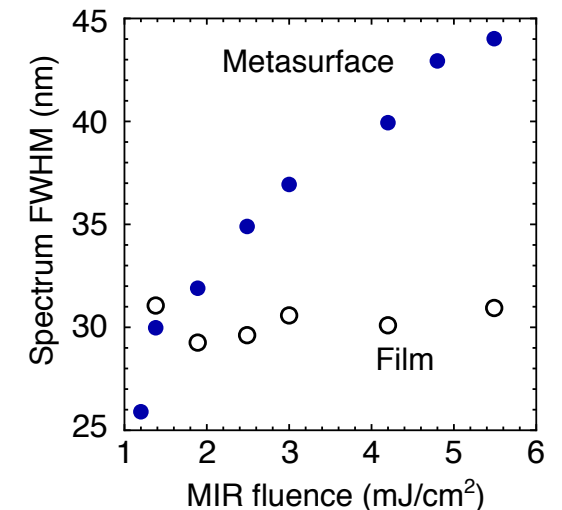
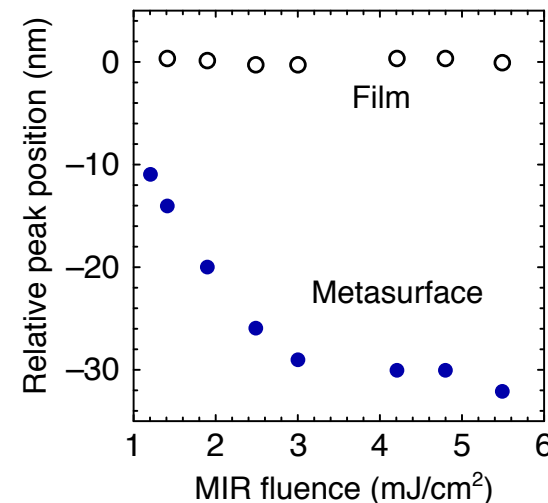




# Photon acceleration probed by THG



Photon acceleration  
at  $10^{-4} \times$  intensity  
and  $10^{-3} \times$  thickness  
of that in gases



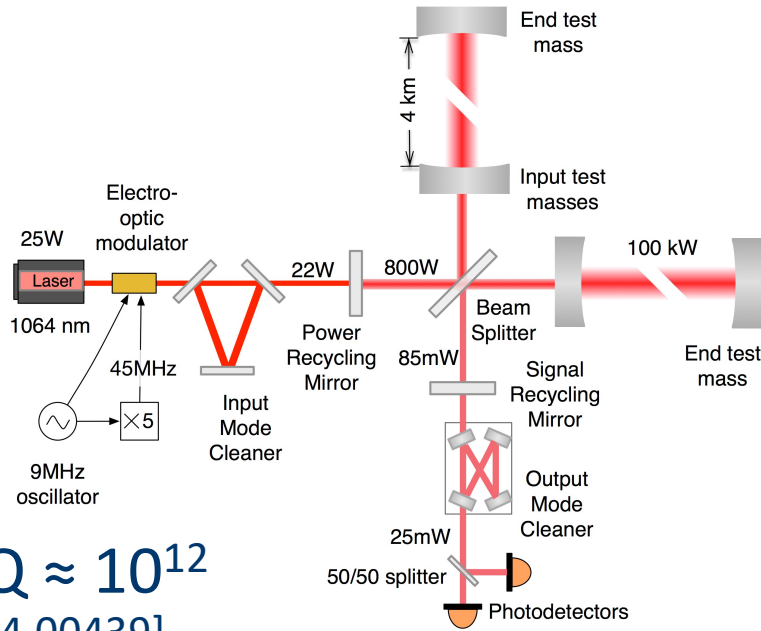


## Time-variant semiconductor metasurfaces. Outline

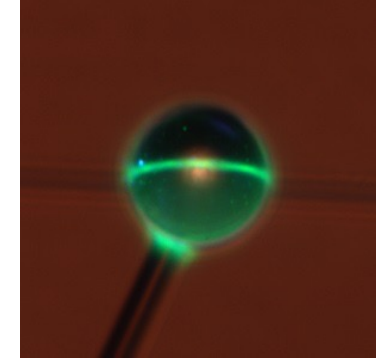
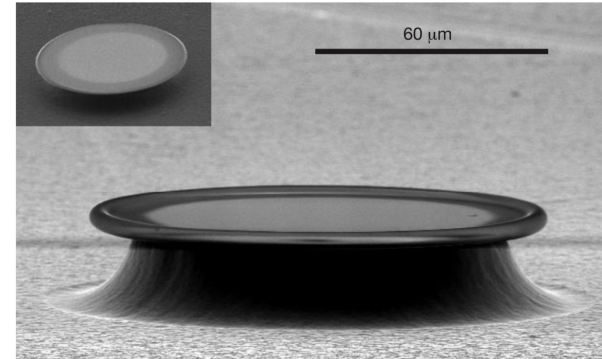
- Frequency conversion
- **Breaking the time-bandwidth limit**
- Discussion: Time-variant  $\epsilon$  nonlinear?
- Conclusion



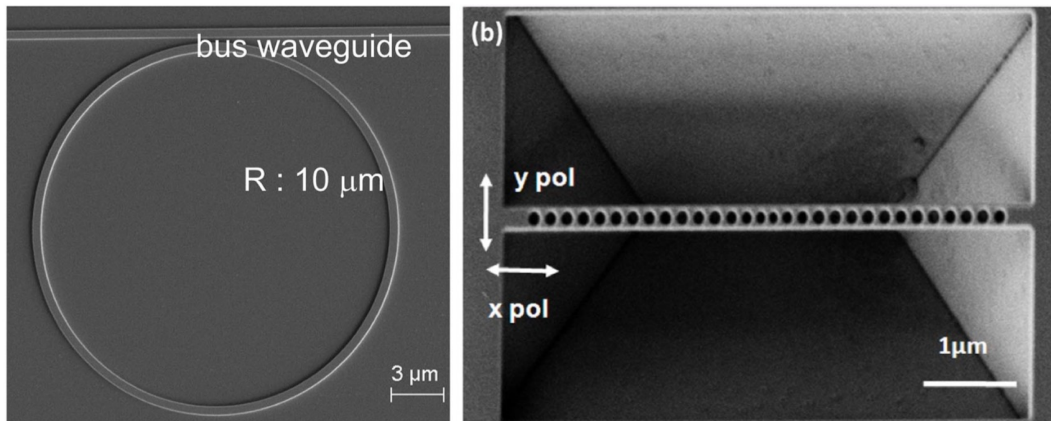
# Resonators and the time-bandwidth limit



LIGO:  $Q \approx 10^{12}$   
[arXiv:1604.00439]



WGM resonators:  $Q \approx 10^{(8-10)}$   
[Vahala, Kippenberg, Gorodetskiy, Oraevsky,...]

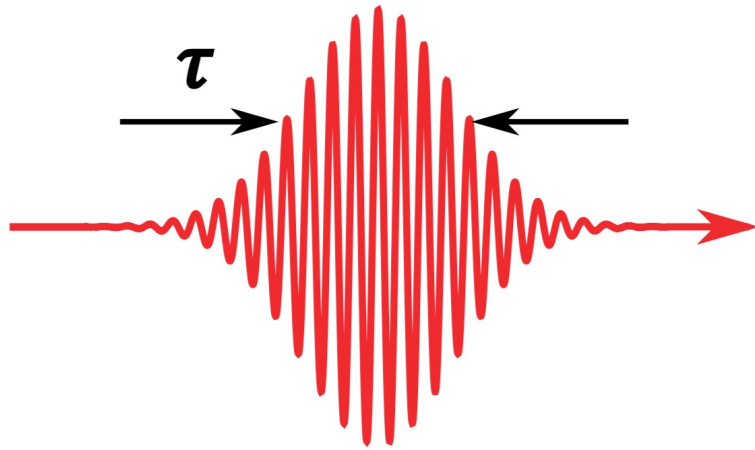


On-chip cavities:  $Q \approx 10^{(4-7)}$   
[Lipson, Gaeta, Loncar, Crozier, Painter...]

The time-bandwidth limit:  
$$\Delta t \times \Delta f \geq 1$$
  
(Kupfmuller principle)



# Problem: broad pulses, narrow resonances



Assume a standard Ti:Sapphire pulse:

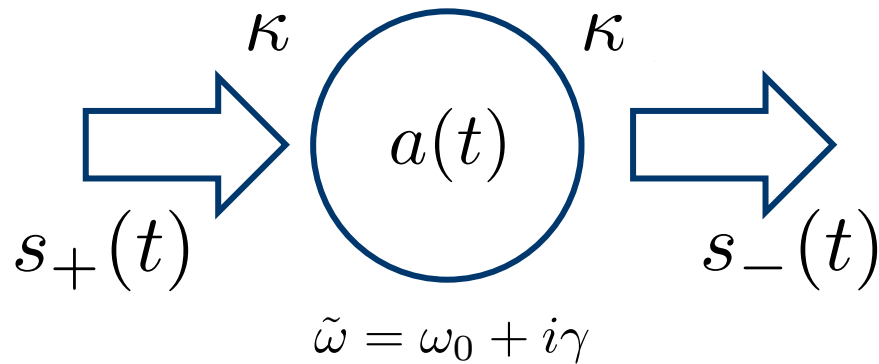
$$\tau = 85 \text{ fs}, \lambda_c = 800 \text{ nm}$$

FWHM bandwidth = **10 nm**

Resonators with  $Q \gtrsim 100$   
do not take advantage  
of the full pulse bandwidth



# Chirped pulse + time-varying cavity

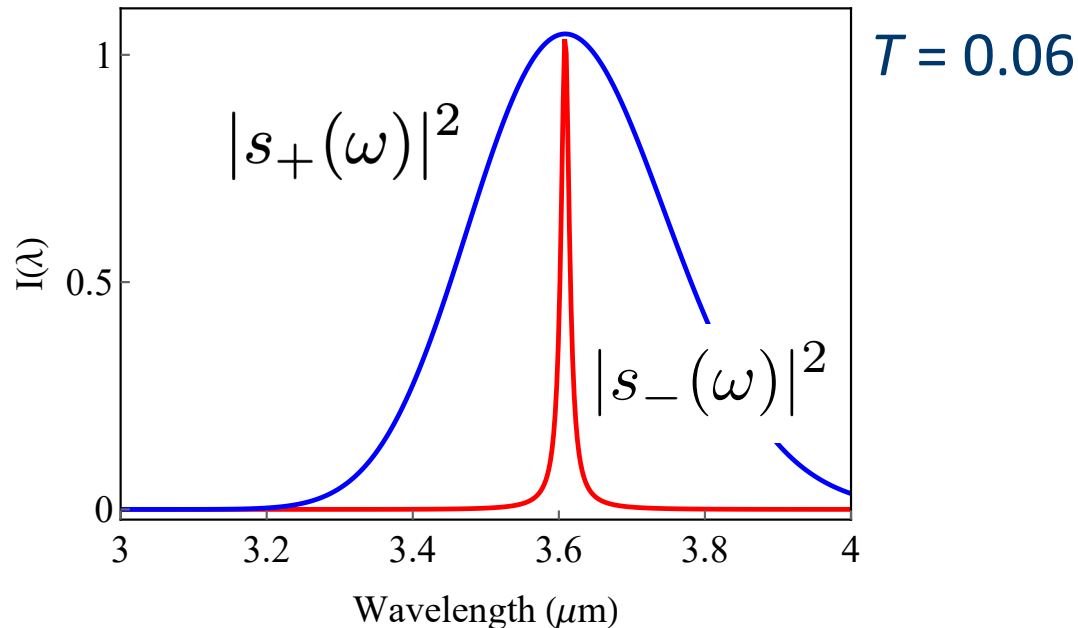


$$\dot{a}(t) + i\tilde{\omega}(t)a(t) = \kappa s_+(t)$$

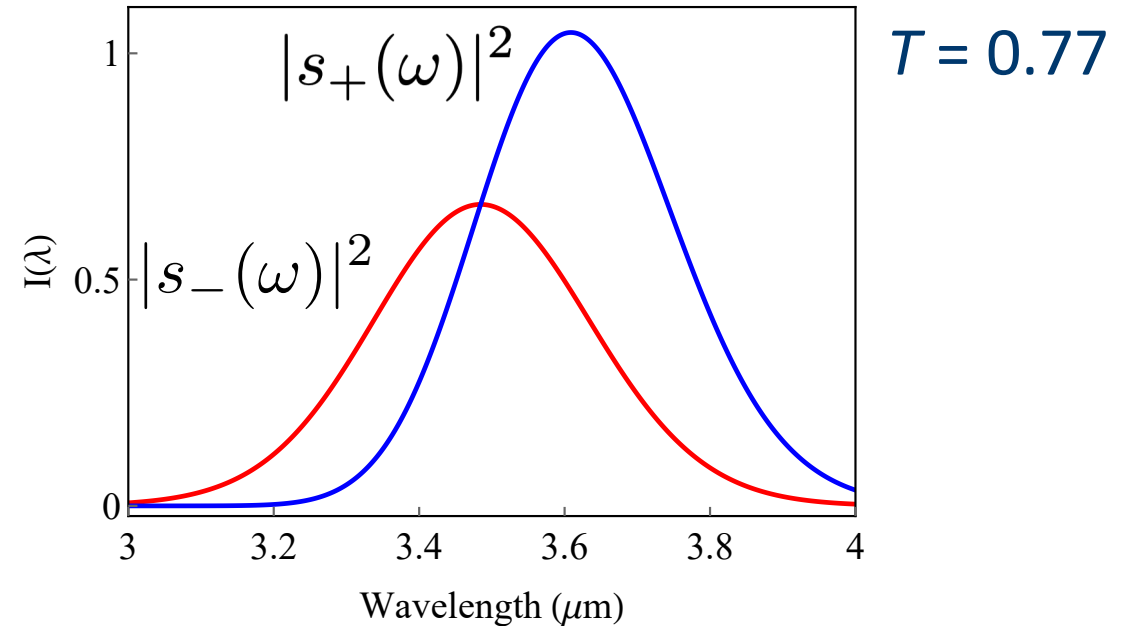
$$s_+(t) = s_0 e^{-i\omega\left(1 + \frac{t\delta}{2}\right)t - \frac{t^2}{\tau^2}}$$

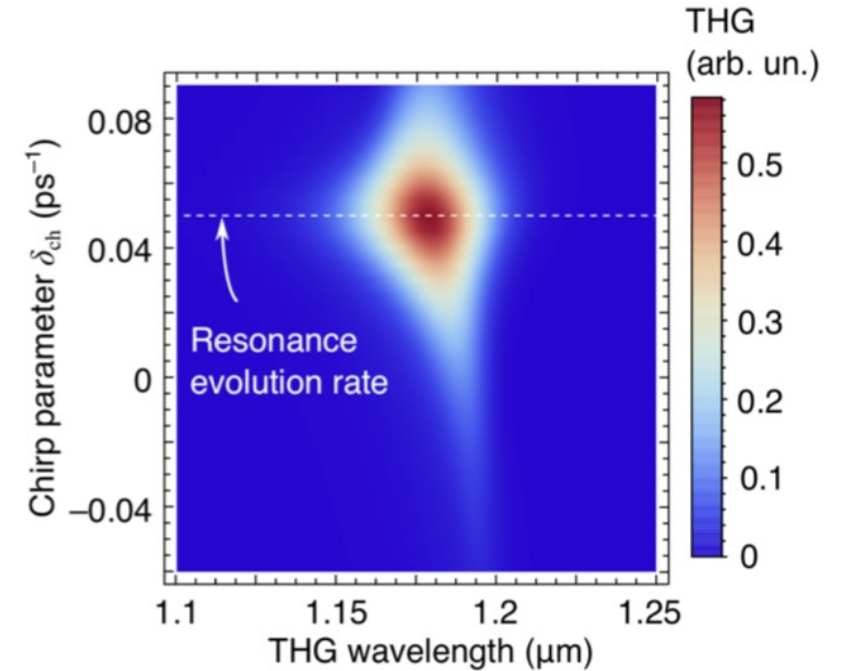
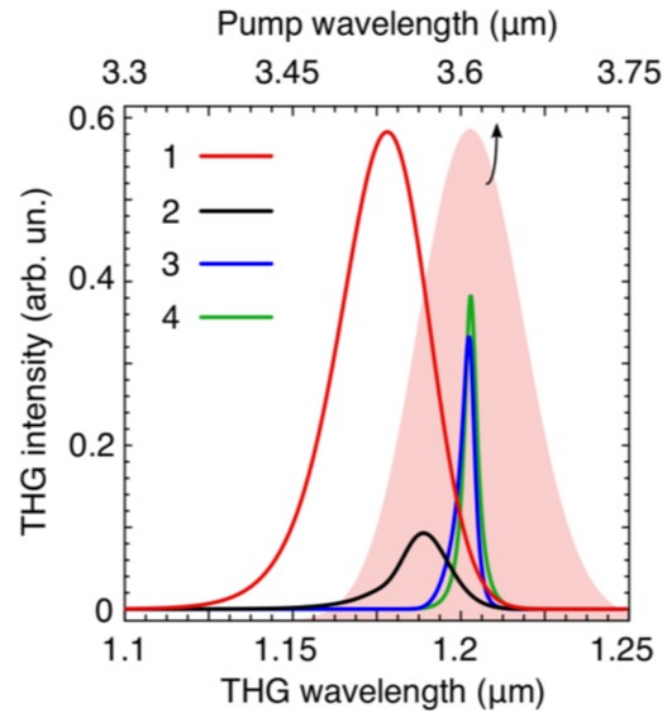
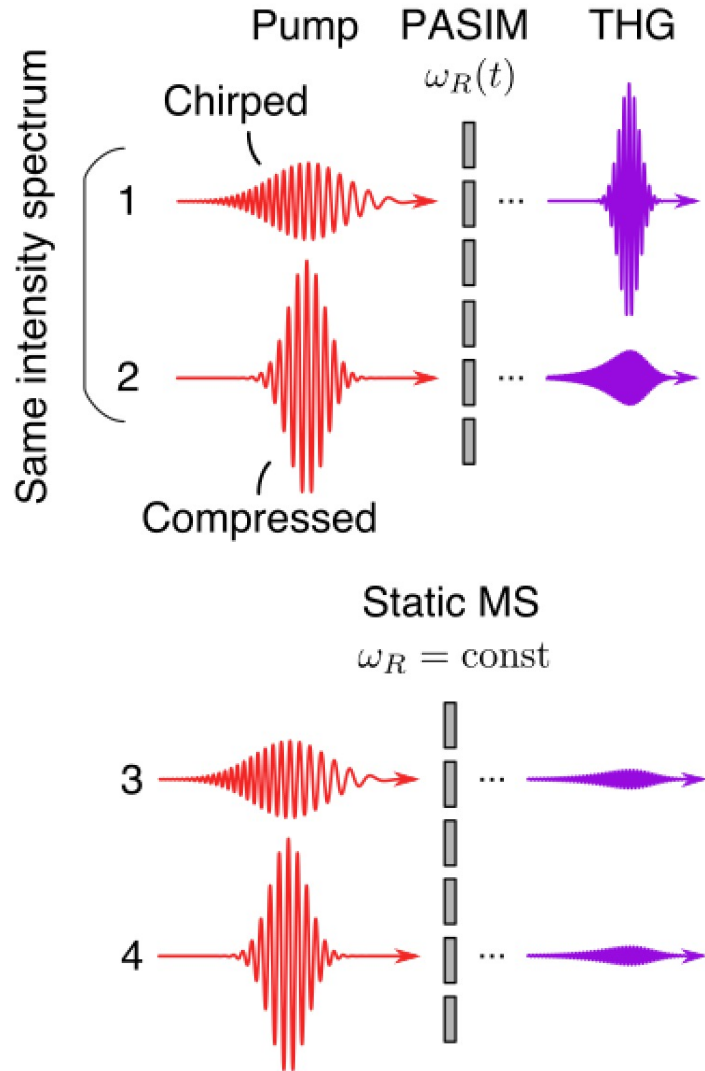
$$s_-(t) = \kappa a(t)$$

Case 1:  $\omega_0$  fixed



Case 2:  $\omega_0 = \omega_0(0)(1 + \alpha t)$ ,  $\alpha = \delta$





Broadband nonlinear response  
from a narrowband (= resonant) cavity

Theory: *Nat. Commun.* **10**, 1345 (2019)

Numerical verification: *PRA* **100**, 063847 (2019)



## Time-variant semiconductor metasurfaces. Outline

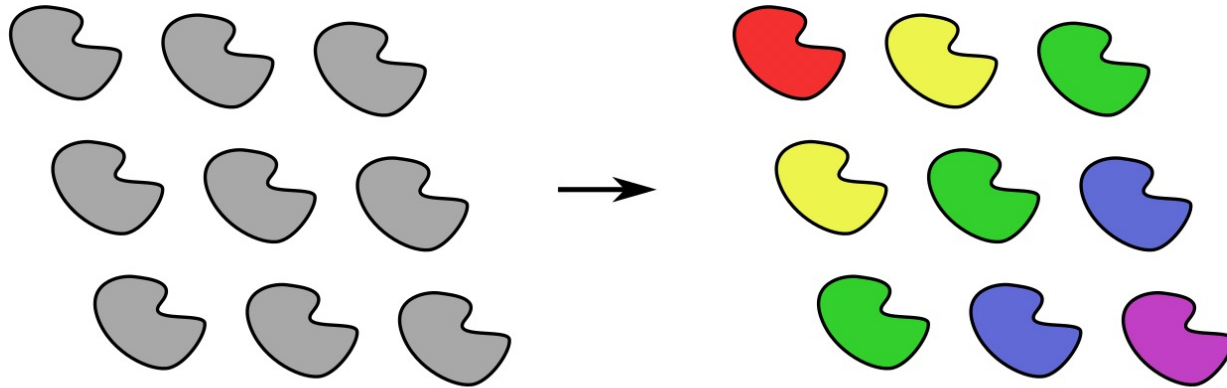
- Frequency conversion
- Breaking the time-bandwidth limit
- **Discussion: Time-variant  $\epsilon$  nonlinear?**
- Conclusion





# Time-variant: also, linear?

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



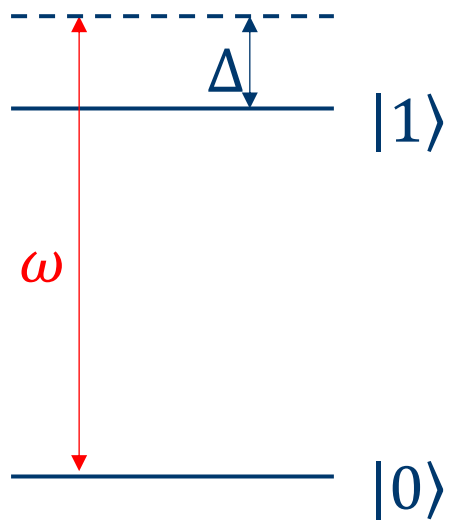
Opinion 1. *Obviously so*: polarization is linear is  $E$ !

Jayathurathnage et al., arxiv:2011.00262v3 (2020) – Sergey Tretyakov’s Group; Lee et al., “Linear frequency conversion via sudden merging of meta-atoms in time-variant metasurfaces,” Nature Photonics 12, 765 (2018) – Bumki Min’s Group; predominantly from RF community

Opinion 2. *Obviously not*:  $\chi(\mathbf{r}, t, t')$  is driven by an external source, which is mixing with  $E$  and generating new frequencies! See, e.g., Raman/Brillouin sidebands.

See QM-description of optical nonlinearities, including Raman, FC-induced blueshift etc.

FCD + NLSE: Zhou et al., *Light Sci. App.* 6, e17008 (2017);

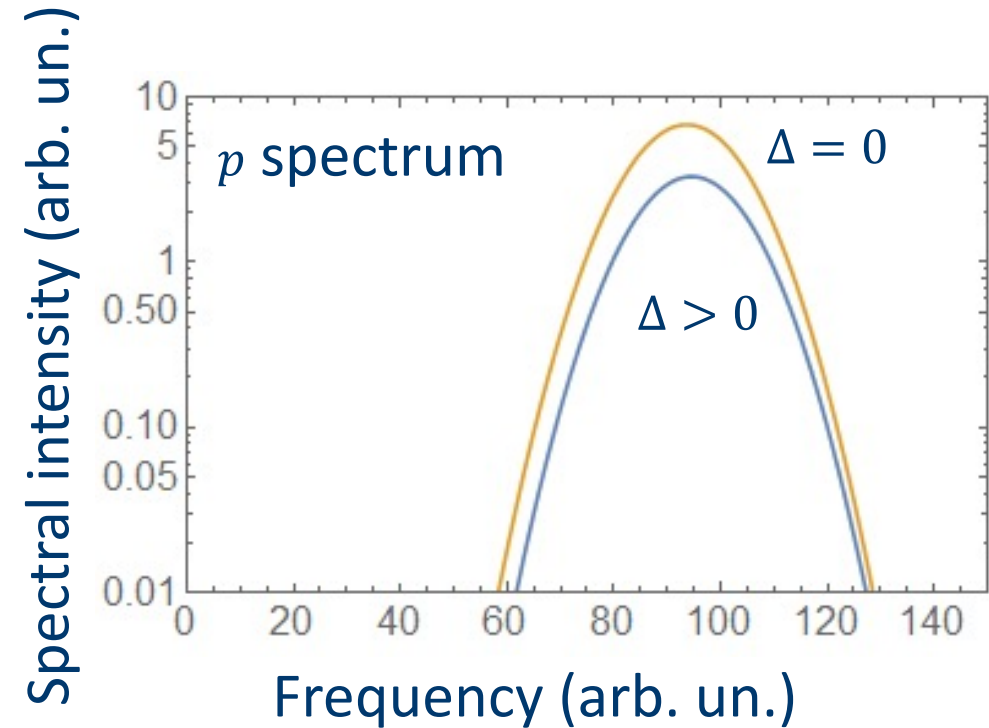


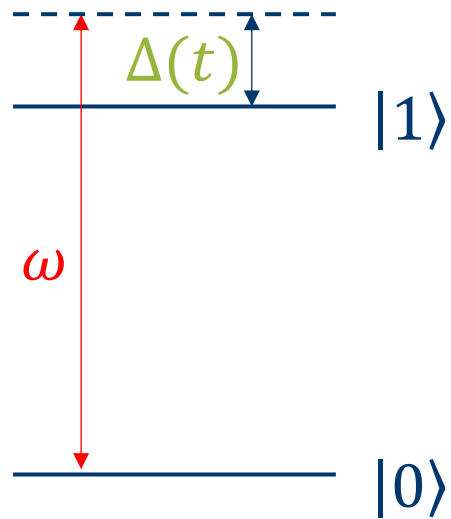
$$\dot{p} = \left( i\Delta - \frac{1}{T_2} \right) p - \frac{i}{\hbar} |\mu|^2 E w$$

$$\dot{w} = -\frac{w + 1}{T_1} - \frac{2i}{\hbar} (pE^* - p^*E)$$

Pulsed excitation:

$$E = E_0 e^{-i\omega t - t^2/\tau^2}$$





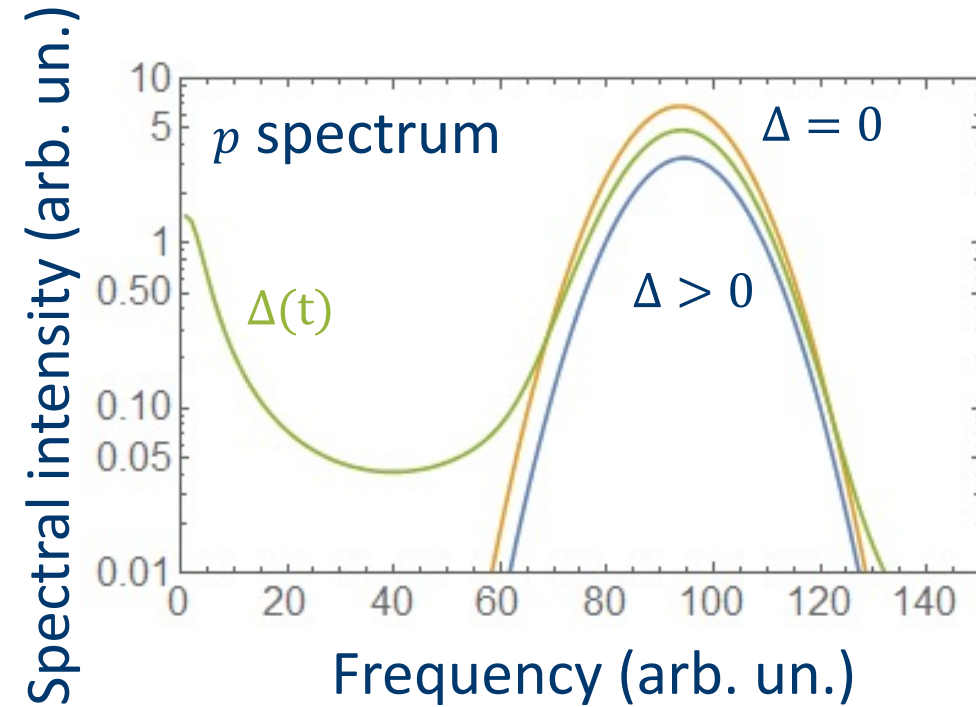
E.g., AC Stark shift

$$\dot{p} = \left( i\Delta(t) - \frac{1}{T_2} \right) p - \frac{i}{\hbar} |\mu|^2 E w$$

$$\dot{w} = -\frac{w + 1}{T_1} - \frac{2i}{\hbar} (pE^* - p^*E)$$

Pulsed excitation:

$$E = E_0 e^{-i\omega t - t^2/\tau^2}$$



Nonlinear? Yes.

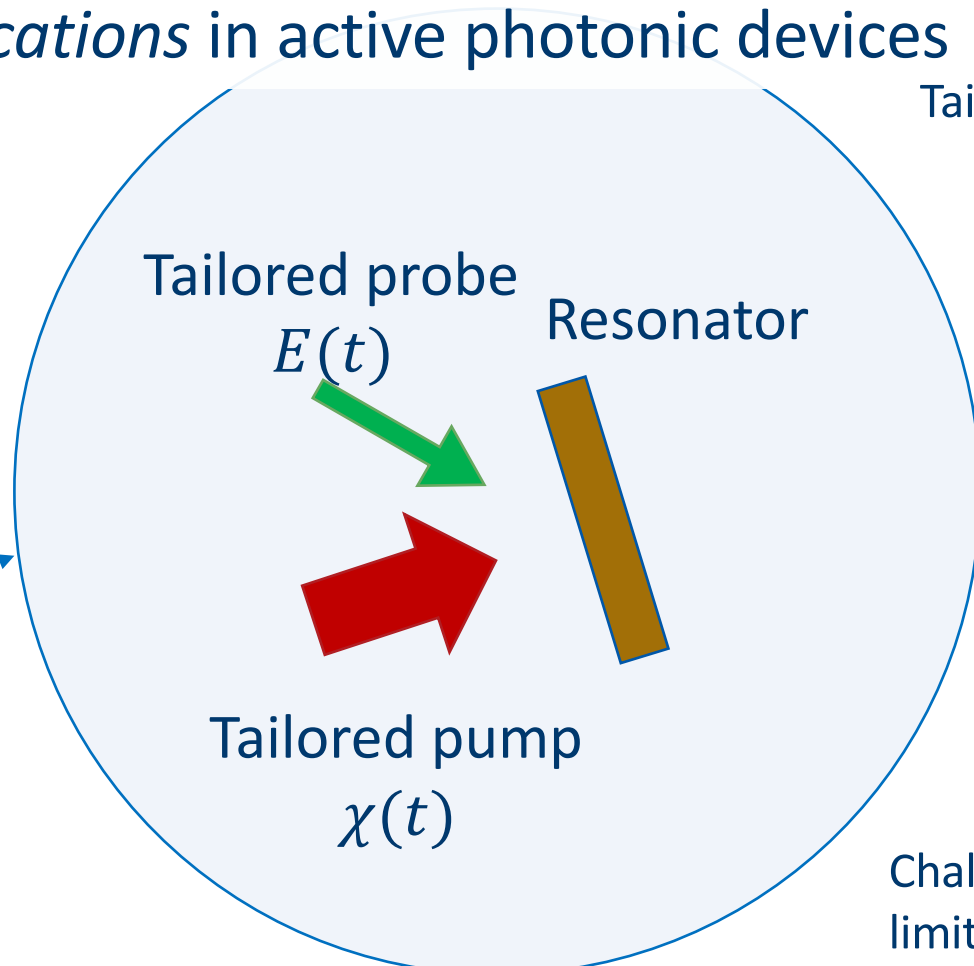
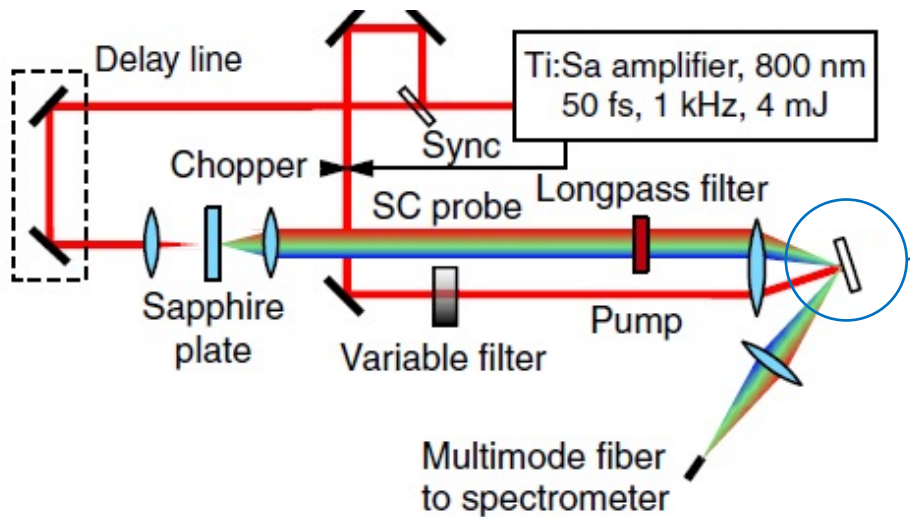
Perturbative? No.

No effective parameters —  
Full QM description needed!



## Utility

Embracing the temporal degree of freedom in metamaterials to advance the *fundamental understanding* of light-matter interactions and *applications* in active photonic devices



Tailoring single-photon spectrum

Frequency conversion\*

Space-time crystals

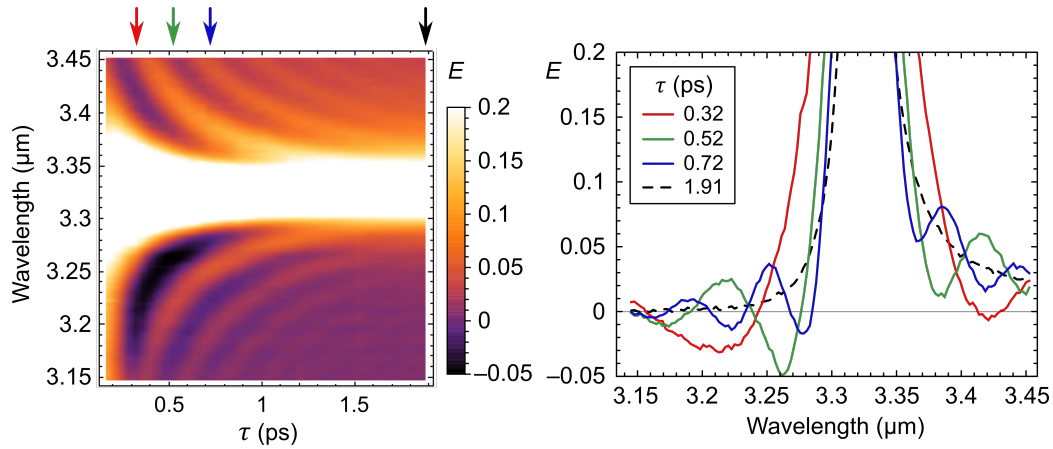
Waveform engineering\*

Non-adiabatic processes\*

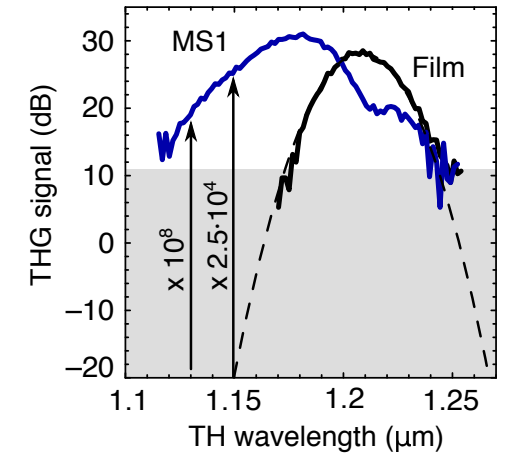
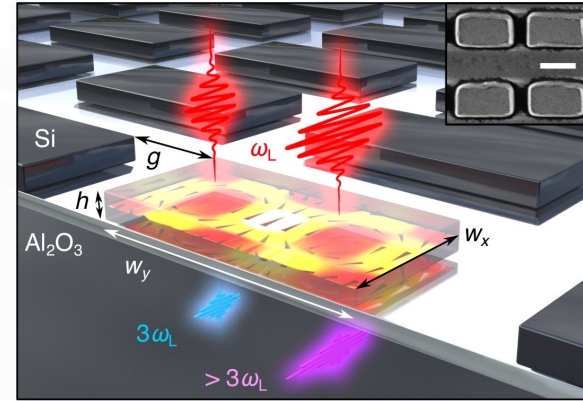
Challenging the fundamental limits of electromagnetism\*



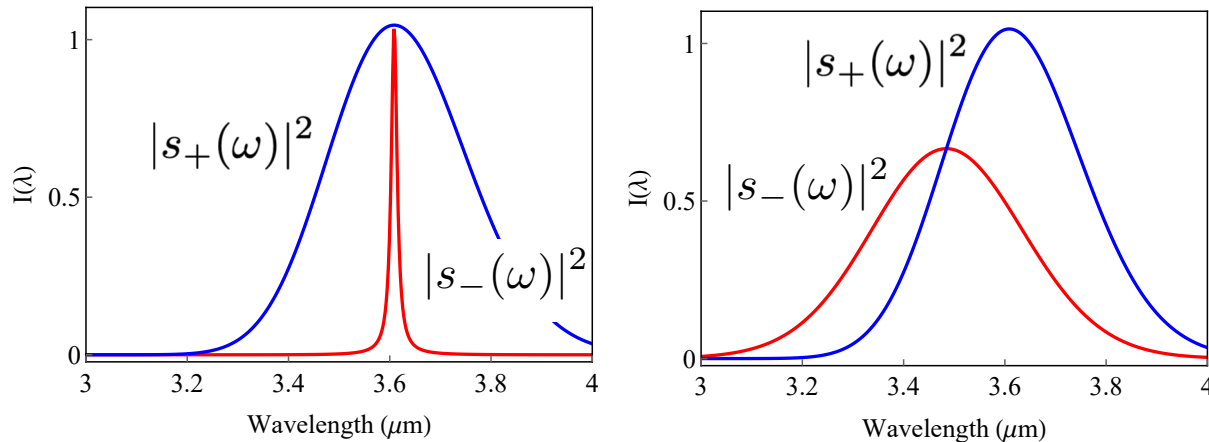
## Frequency conversion



## Photon acceleration (self-conversion)



## Broadband resonant light-matter interactions



*Nat. Commun.* **8**, 17 (2017)

arXiv:2008.03619 (2020)

*Nat. Commun.* **10**, 1345 (2019)

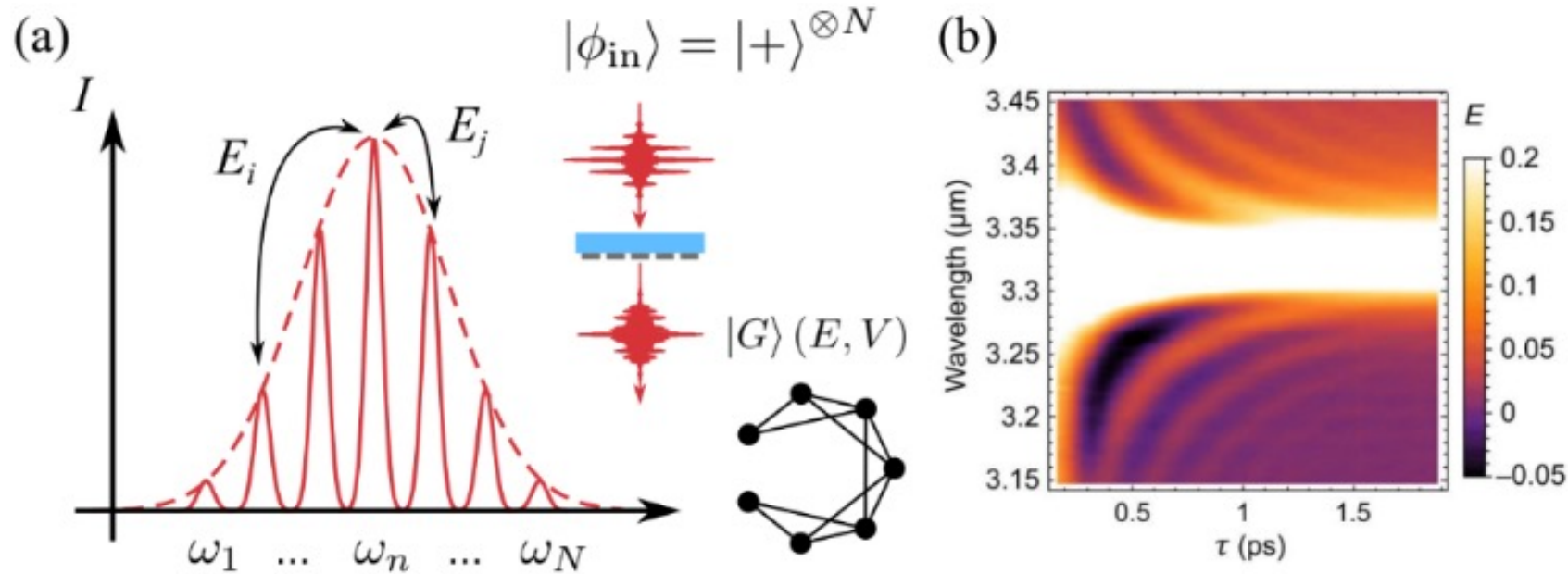
arXiv:2012.06604 (2020)

*Phys. Rev. A* **100**, 063847 (2019)

*Optica* **6**, 1441 (2019)

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

My group @ UC Irvine EECS is hiring  
 Email: [maxim.shcherbakov@uci.edu](mailto:maxim.shcherbakov@uci.edu)  
 Website: [shcherbakov.eng.uci.edu](http://shcherbakov.eng.uci.edu)



**Fig. 5. Quantum networks based on graph states in time-variant resonators.** (a) The input spectrally shaped laser pulse serves as a set of vertices  $V$  for the future graph state. The vertices are entangled in a time-variant metasurface to form a graph state with edges  $E$ . The prepared state  $|G\rangle$  can serve a basis for one-way quantum computing. (b) Preliminary results: classical-optical analog of frequency entanglement with mid-infrared photons [Shcherbakov et al., *Optica* 2019].