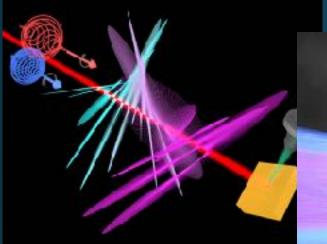
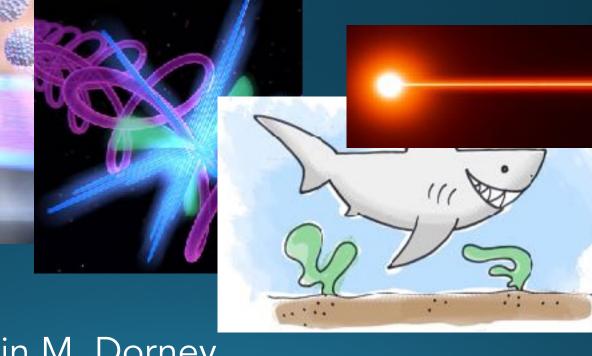
## How to Create and Control Nature's Most Exotic Light Source:

X-ray and Attosecond Light Science at the Molecular, Nano, and Atomic Frontiers





Kevin M. Dorney

Kapteyn-Murnane Group, JILA and University of Colorado Boulder

# KM Group and JILA: Excellent students, collaborators, and **advisors**

#### KM Group Spring 2017



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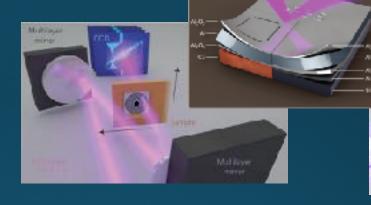


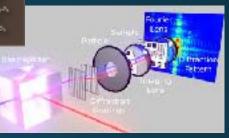


#### Attosecond Extreme Nonlinear Optics

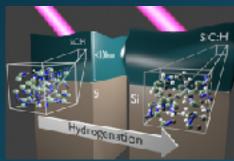


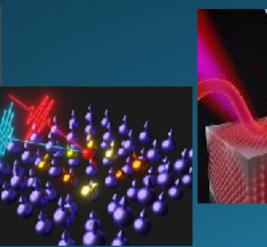
Coherent x-ray Imaging



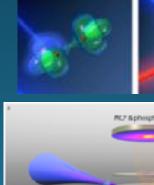


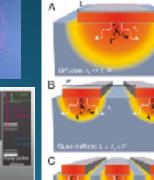
#### **Ultrafast Materials Science**



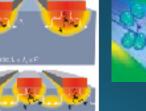


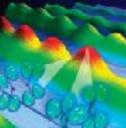














## "Oh, you do scientific research? Neat! What exactly are you working on???"



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### Mastering Fundamentals

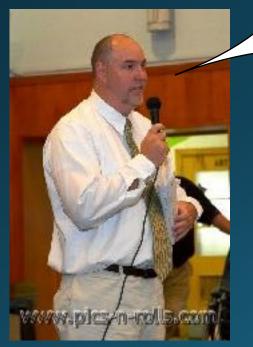
"If ya want to dunk, make 3's, and break knees, ya gotta learn how to dribble, pass, pivot, etc."

Worw.pics-n-rolls.com



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"If ya want to dunk, make 3's, and break knees, ya gotta learn how to dribble, pass, pivot, etc."

- Basic research
- Small-scale systems
- Idealized environments
- Little (initial) real world impact



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## Understanding and Manipulating Nature!

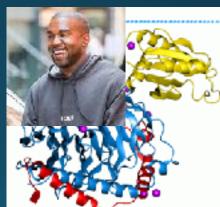
Blue LEDs (2014)



<u>Laser (1964)</u>



<u>G Proteins (2012)</u>



**Green Fluorescent** 

**Protein** (2008)

<u>CCDs Detectors</u> (2009)



4

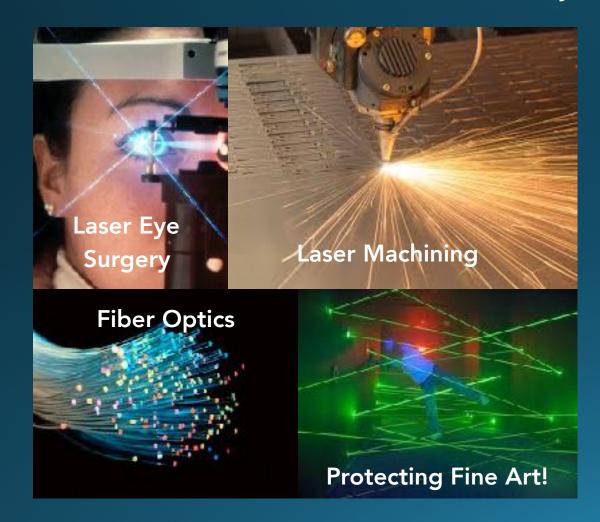




Visible/Invisible Laser Benefits Society



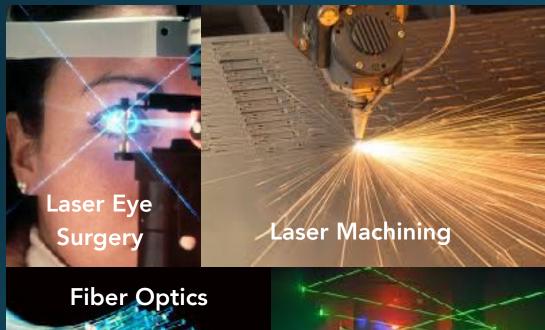
### Visible/Invisible Laser Benefits Society

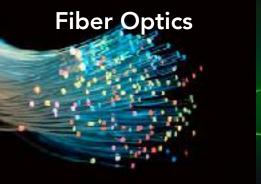




**Protecting Fine Art!** 

### Visible/Invisible Laser Benefits Society



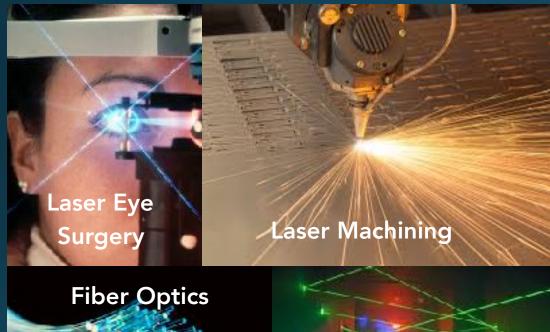




X-Ray Light Benefits Society



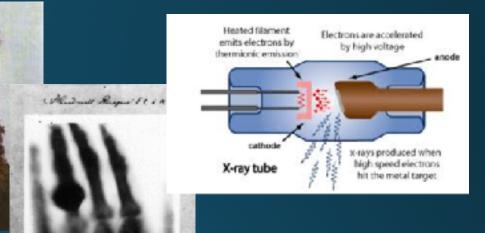
### Visible/Invisible Laser Benefits Society





### X-Ray Light Benefits Society

Terhog B Comment

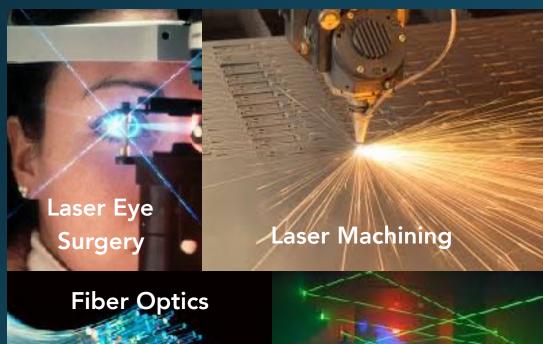


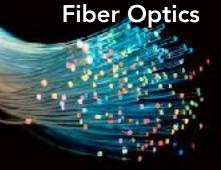


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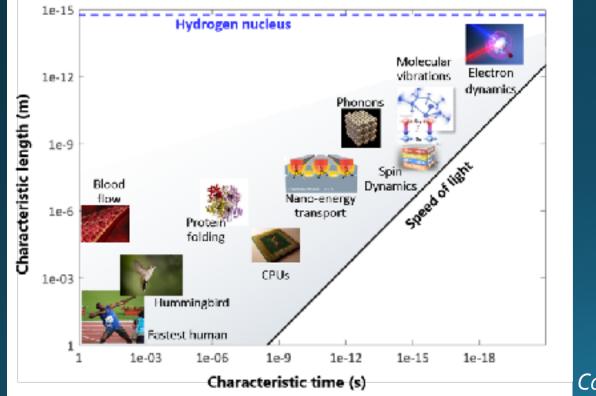
Protecting Fine Art!



- Direct observation of atomic and molecular scale transformations at their natural **time** and **length scales**.

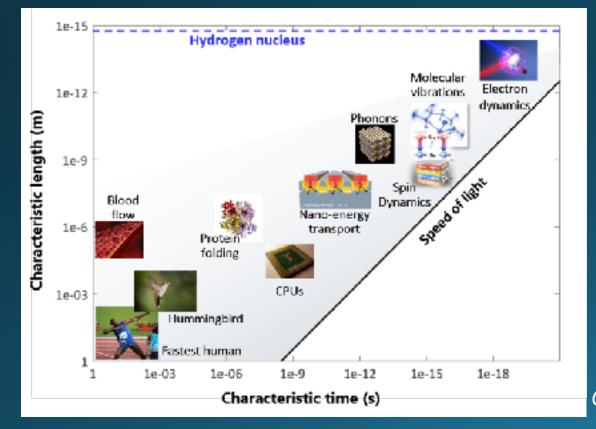


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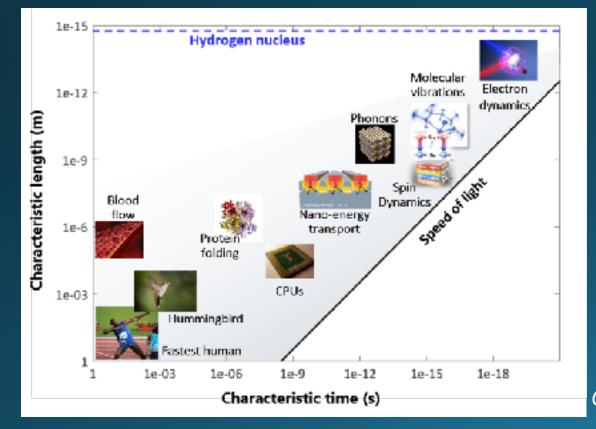
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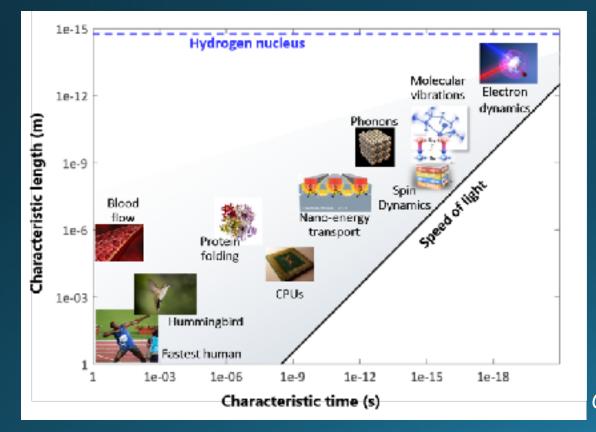
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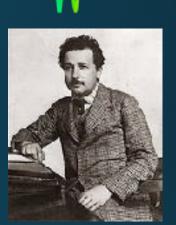


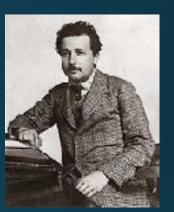
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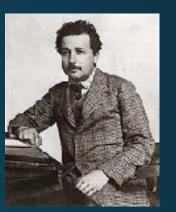


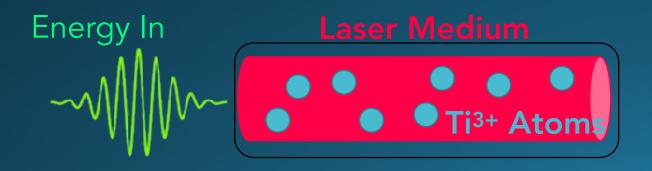




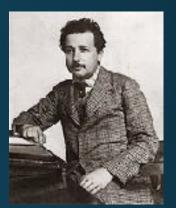






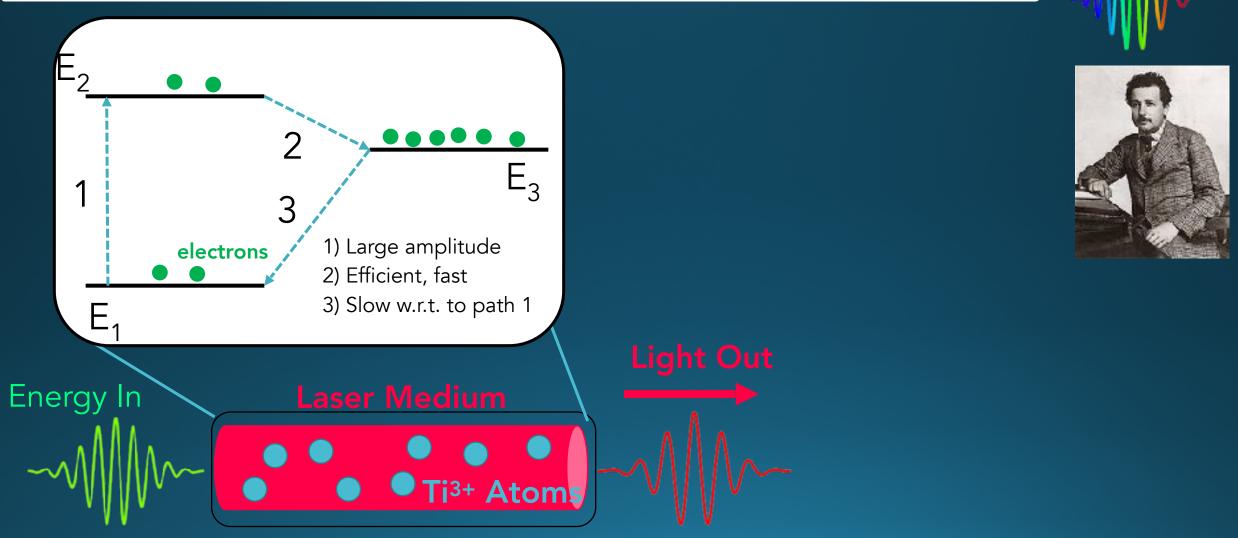


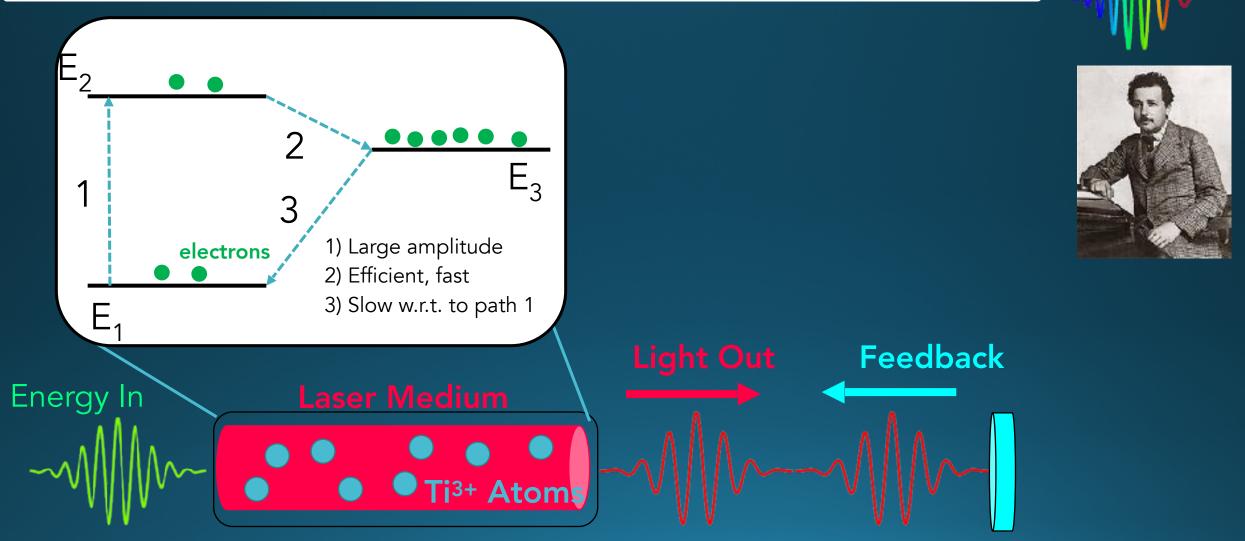
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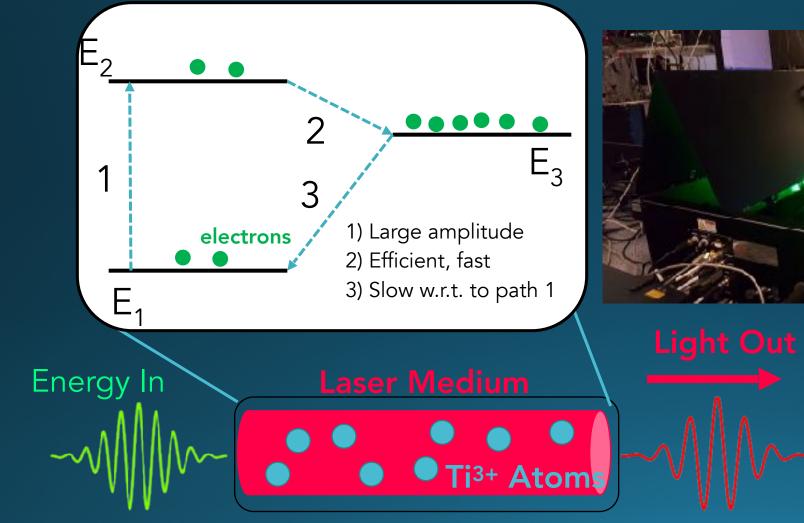


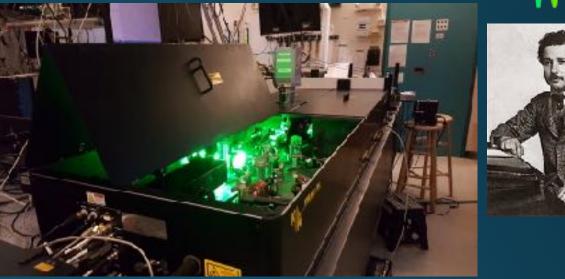


7









Light Out Feedback

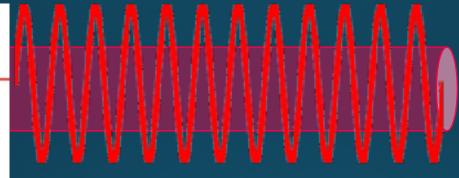






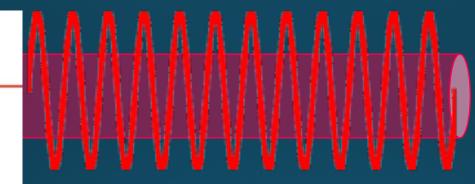










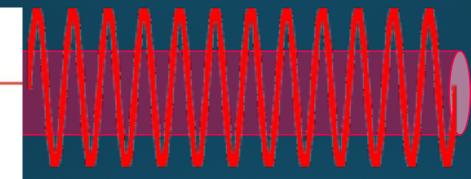


## <u>CW Laser Light</u>

Precise frequency/wavelength High Average Power Very Long Term Stability Easily Engineered/Designed







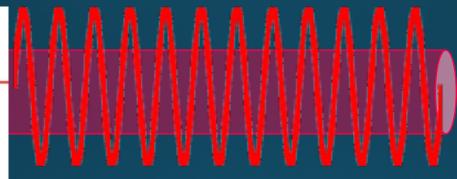
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• Ultrafast lasers emit light in **extremely short, high intensity pulses.** 







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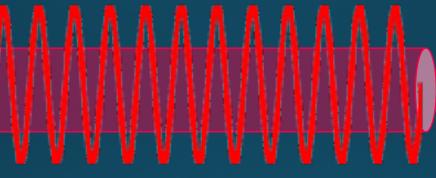
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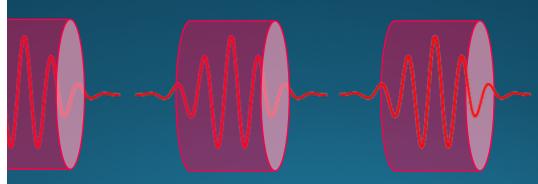




Precise frequency/wavelength High Average Power Very Long Term Stability Easily Engineered/Designed

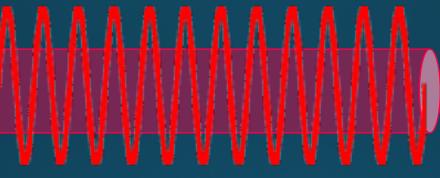
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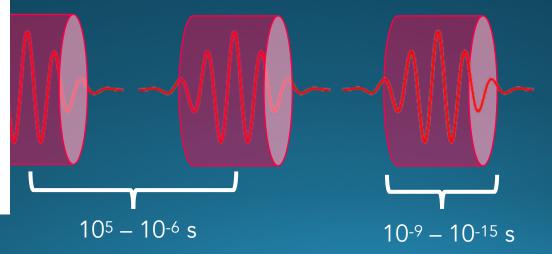


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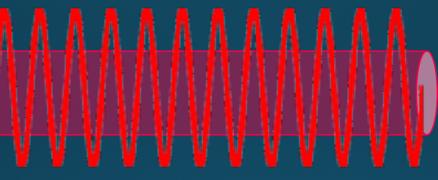
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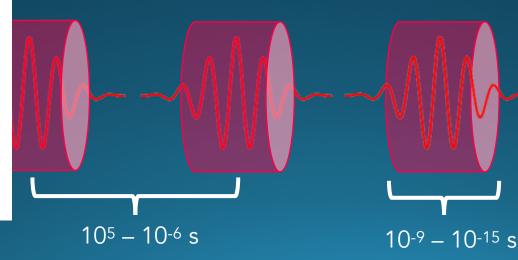


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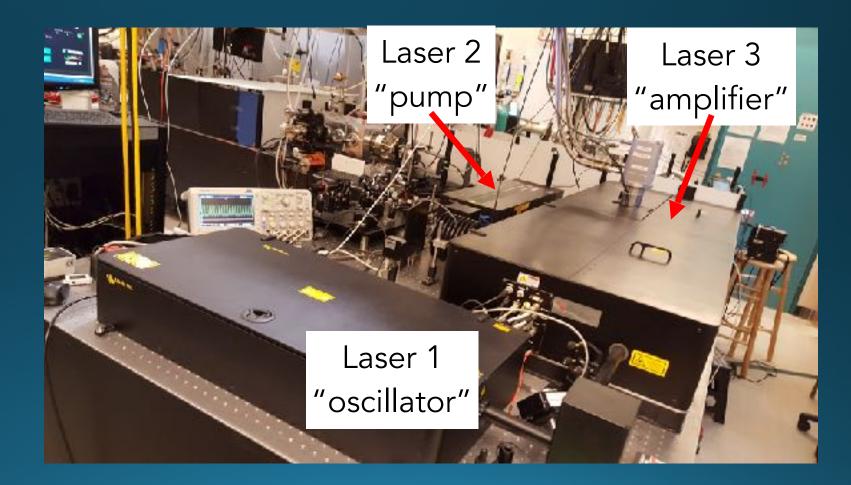


## UF Laser Light

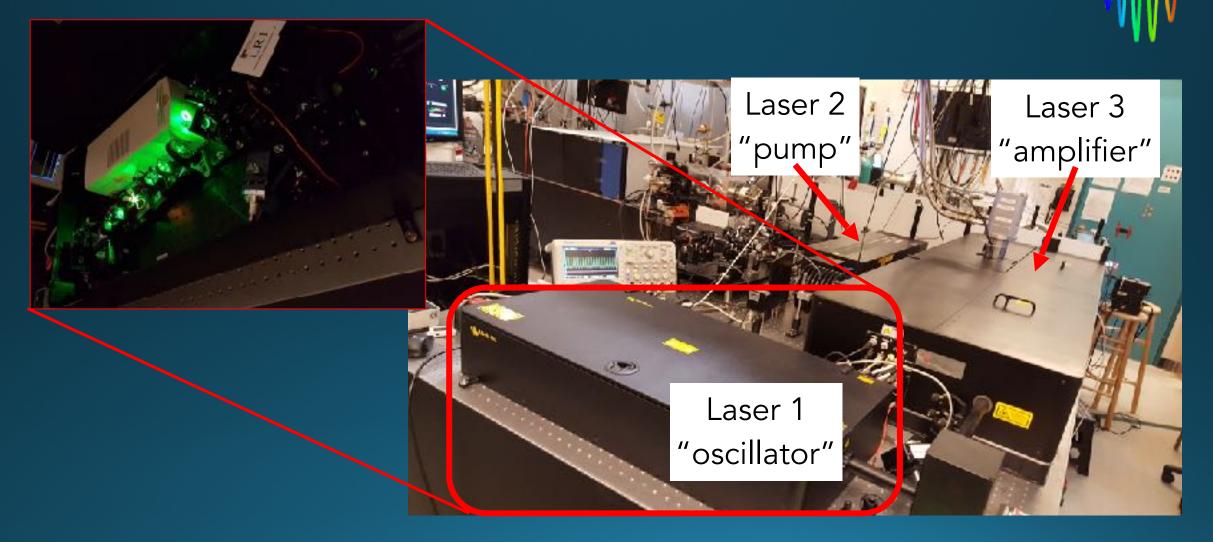
Many Frequencies/Wavelengths Very High Intensity Flashes "on" for 10<sup>-9</sup> – 10<sup>-15</sup> s More Complicated to Design











# Ultrafast Laser Amplifiers in the KM Group: Really Big Freakin' Lasers!



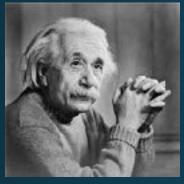
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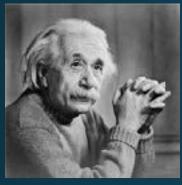
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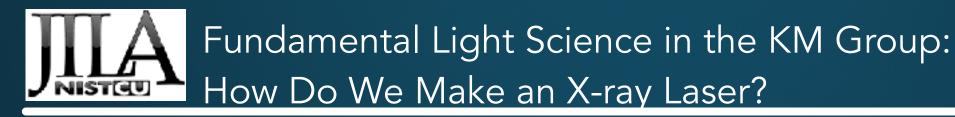


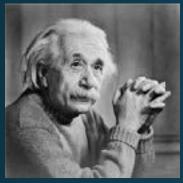




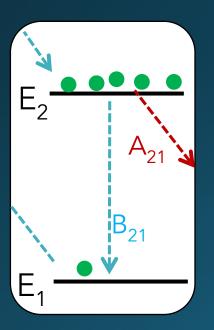


- Unfortunately, it is extremely difficult
  - X-rays are very high in energy
  - Other competing processes in laser material

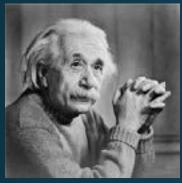




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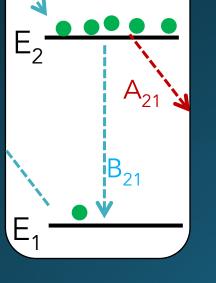




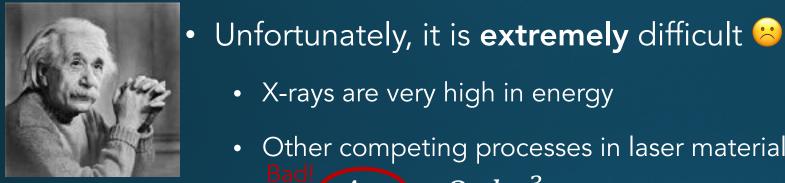


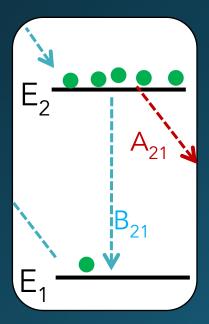
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$$\frac{A_{21}}{B_{21}} = \frac{8\pi h v^3}{c^3} \propto v^3 \ (\lambda^{-3})$$









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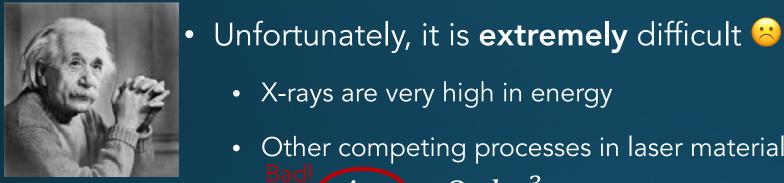
Good!

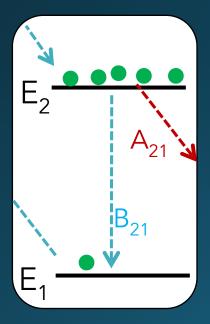
 $B_{21}$ 

•



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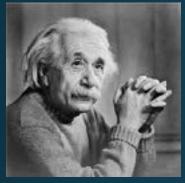


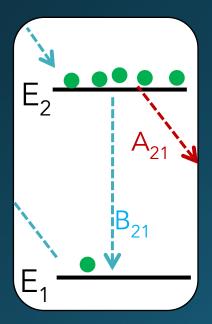
Power 
$$\propto \left(\frac{1}{\sigma_g}\right) \left(\frac{1}{\tau}\right) (hv) \propto v^5$$



Fundamental Light Science in the KM Group: How Do We Make an X-ray Laser?

• So... We already have lasers and x-rays... Is it really that hard to combine them?





- Unfortunately, it is **extremely** difficult 😕
  - X-rays are very high in energy

Goo

• Other competing processes in laser material

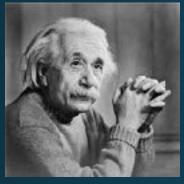
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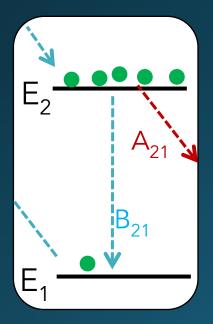
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Fundamental Light Science in the KM Group: How Do We Make an X-ray Laser?





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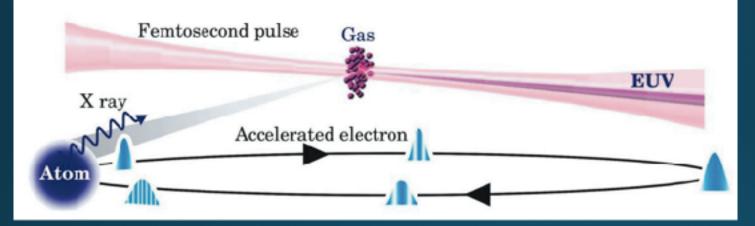


Kuchiev, JETP, 45. 404 (1987)

Classical: Corkum. PRL 1993

QM: Kulander, Schafer, Krause. SILAP 1992



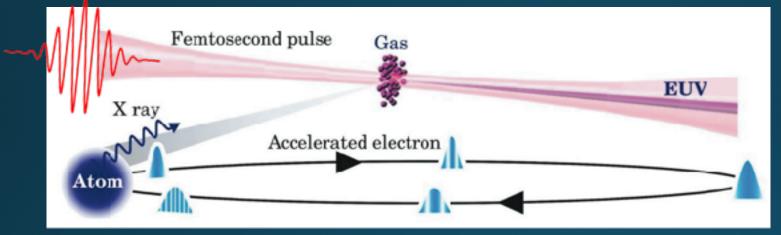


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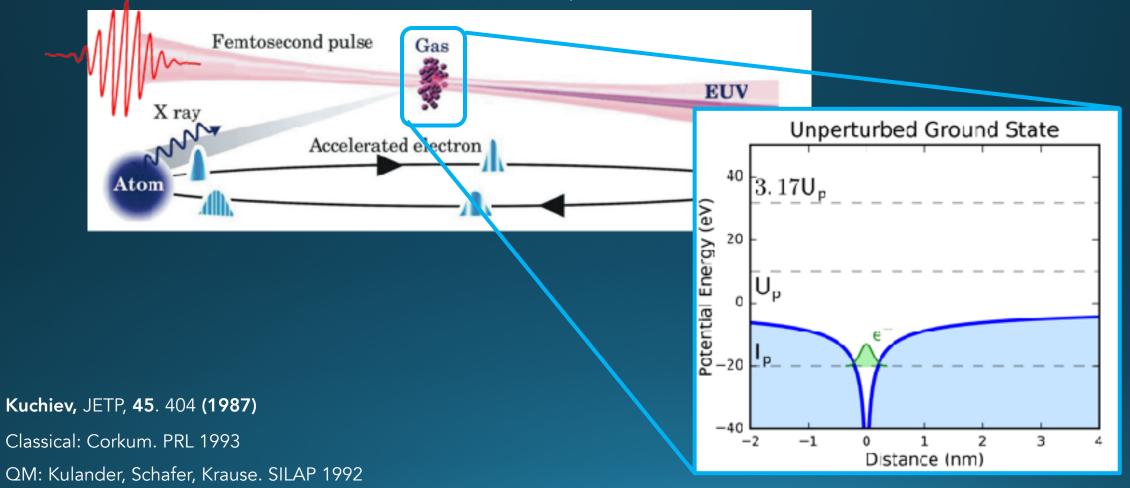


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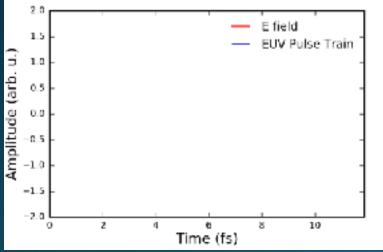
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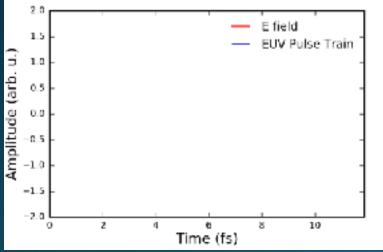
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- <u>Microscopically</u>
  - Field-driven electrons recombine over a distribution of times (e.g., energies), generating harmonics at some multiple of the laser frequency.
  - Emitted light is coherent, high energy, and sub-femtosecond (10-15 s)!







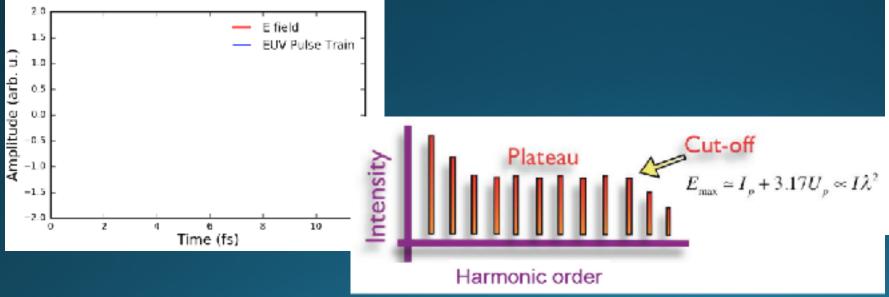
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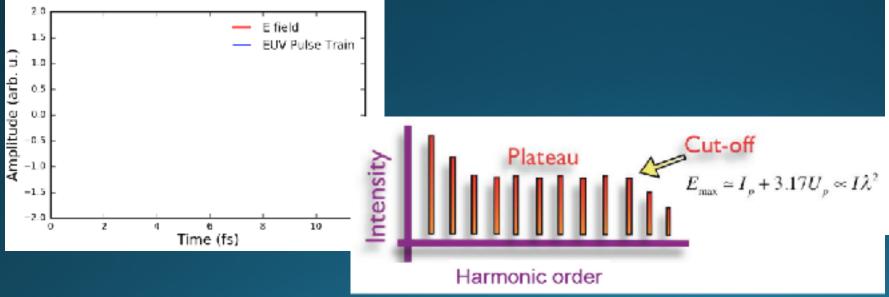


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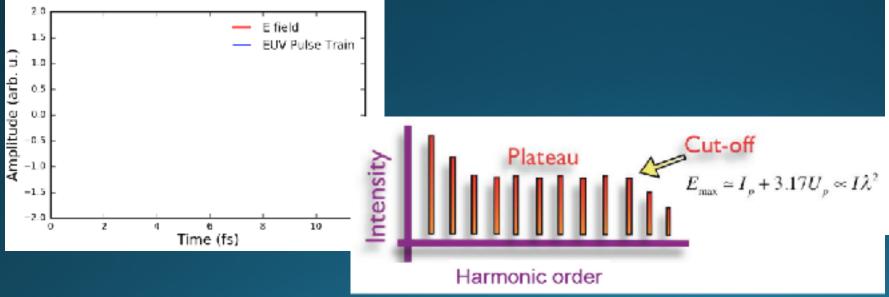


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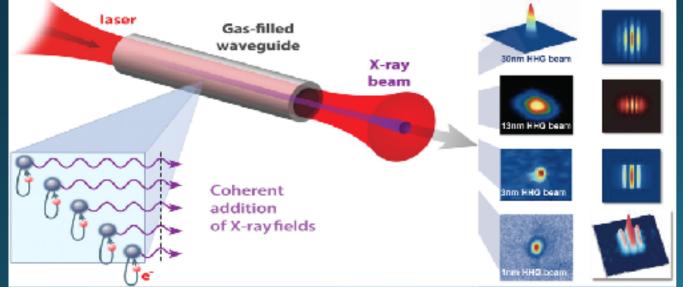




- However, practical applications demand a high brightness (photon flux).
- <u>Macroscopically</u>
  - Add contribution of many atoms coherently, bright x-rays!
  - But, the upconversion process is violent and chaotic...
  - Careful control of experiment allows for bright x-ray beams to be generated!



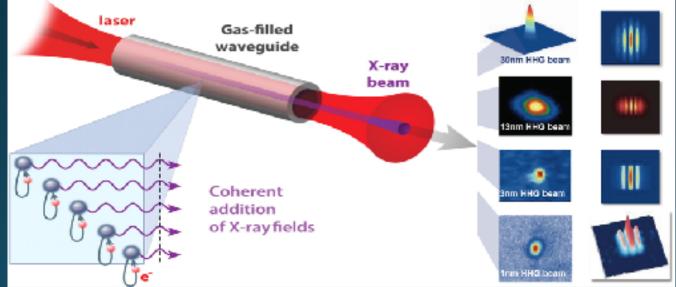
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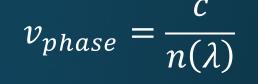




# Using HHG to Generate Bright X-rays: Spectrotemporal Properties and Structure

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#### <u>Careful control of experiment allows for bright x-ray beams to be generated!</u> ٠

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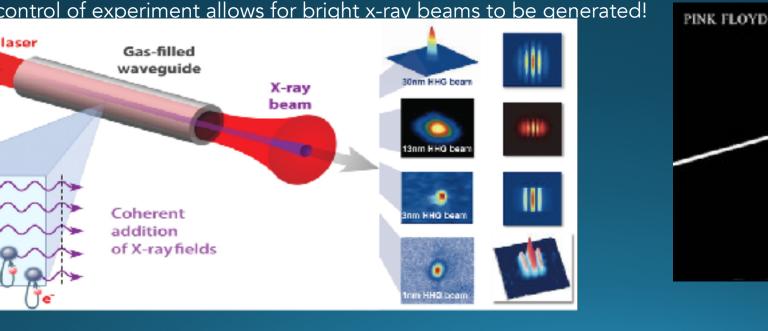
Spectrotemporal Properties and Structure

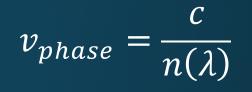
laser Gas-filled waveguide 30nm HHG beam X-ray beam 3nm HHQ bear Coherent Im HHO bear addition of X-ray fields

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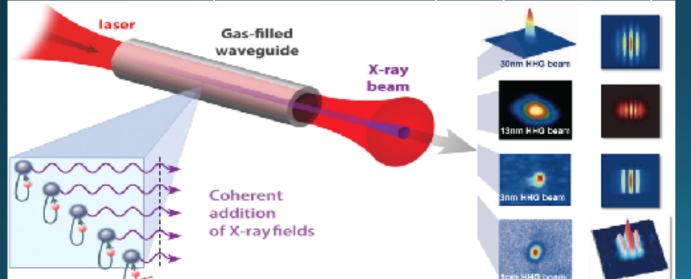


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Using HHG to Generate Bright X-rays:

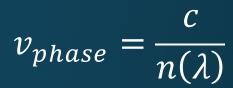
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- Add contribution of many atoms coherently, bright x-rays!
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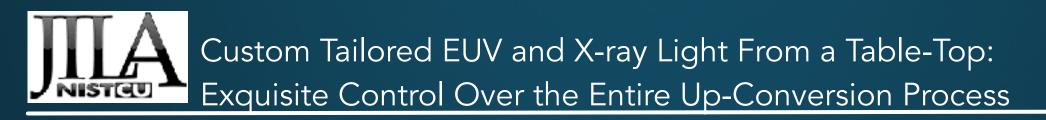




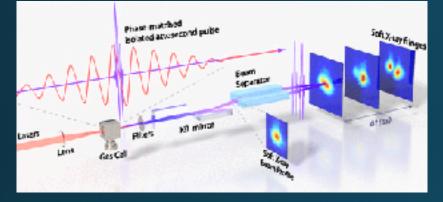




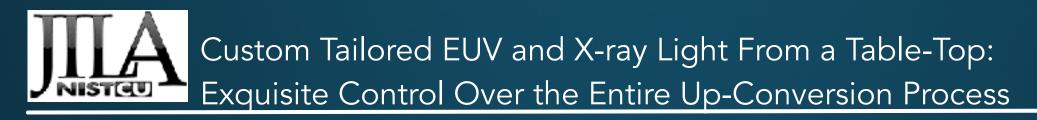
Chen, PNAS, 111, 2014 Popmintchev, Science, 6086, 2012 Popmintchev, Science, 6265, 2015 Fan, PNAS, 2015



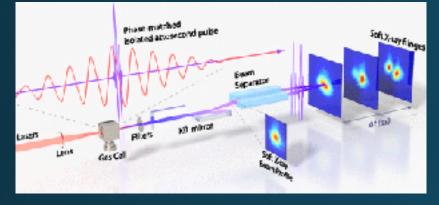
## Phase-matched Isolated Attosecond Pulses



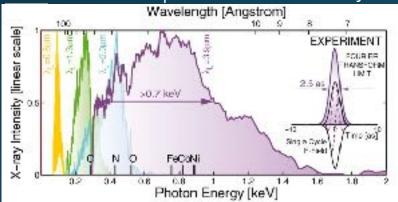
Chen, PNAS, 111, 2014 Popmintchev, Science, 6086, 2012 Popmintchev, Science, 6265, 2015 Fan, PNAS, 2015



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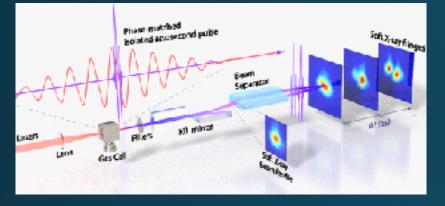
### Coherent, Zeptosecond, keV X-rays



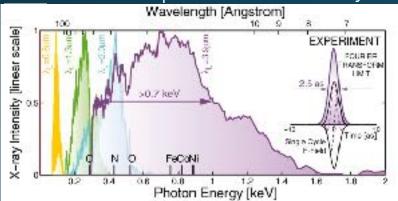
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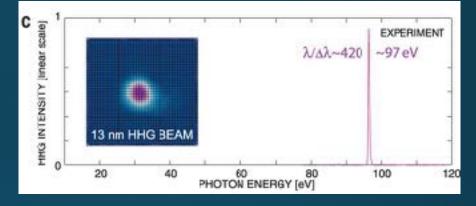






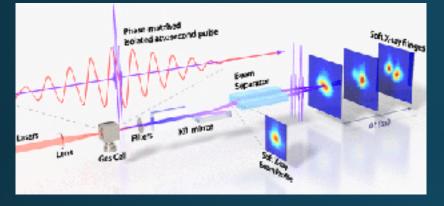
Chen, PNAS, 111, 2014 Popmintchev, Science, 6086, 2012 Popmintchev, Science, 6265, 2015 Fan, PNAS, 2015

### Bright, Isolated Harmonics

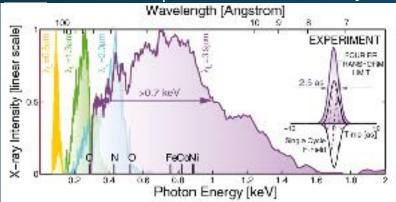




#### Phase-matched Isolated Attosecond Pulses

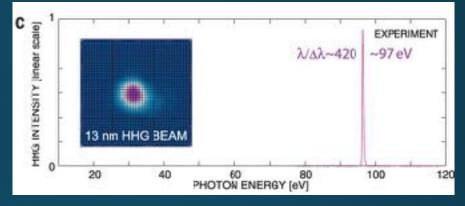


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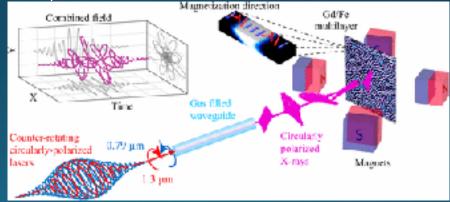


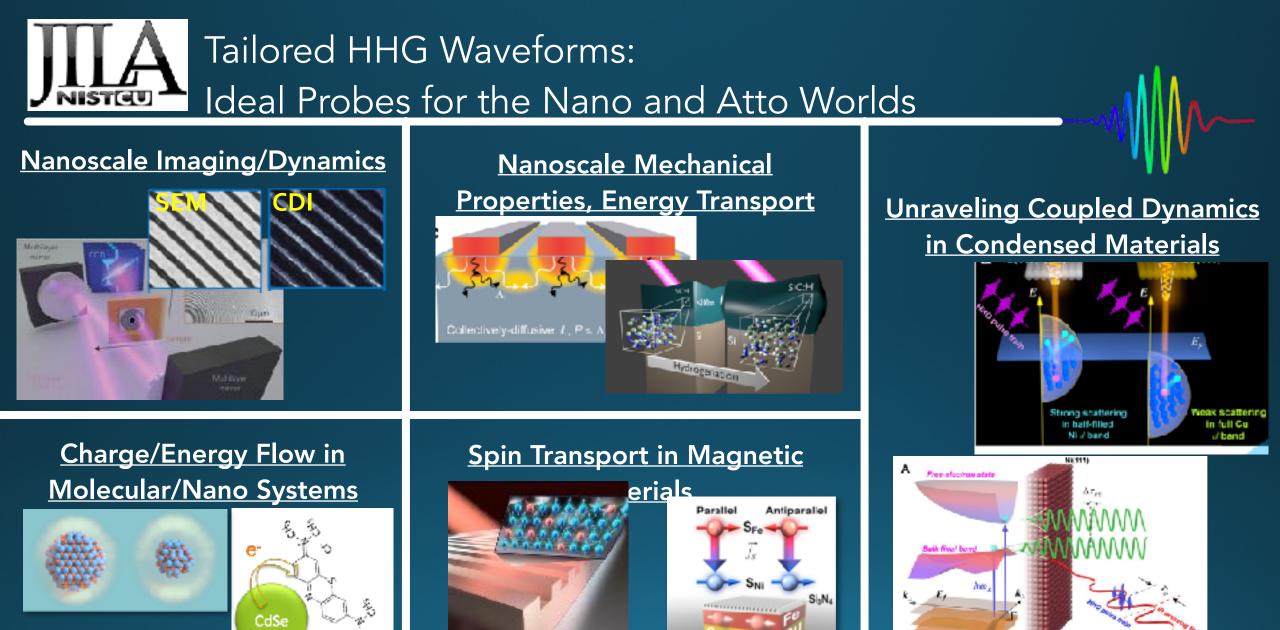
Chen, PNAS, 111, 2014 Popmintchev, Science, 6086, 2012 Popmintchev, Science, 6265, 2015 Fan, PNAS, 2015

#### Bright, Isolated Harmonics



#### Elliptical and Circularly Polarized Harmonics





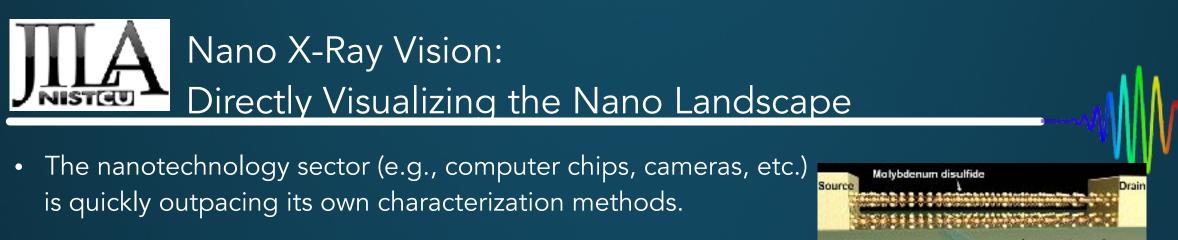
16

Askial forset

SIO.



- The nanotechnology sector (e.g., computer chips, cameras, etc.) is quickly outpacing its own characterization methods.
  - Why? Diffraction limited resolution of imaging techniques!
  - Shorter wavelengths = smaller features!



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- Shorter wavelengths = smaller features!

Malybdenum di	isulfide 1 Drain
Zirconium dioxide	1 nanometer carbon nanotube (gate)
Silicon	and the second



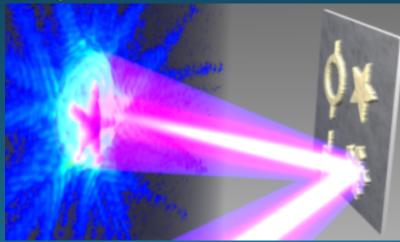




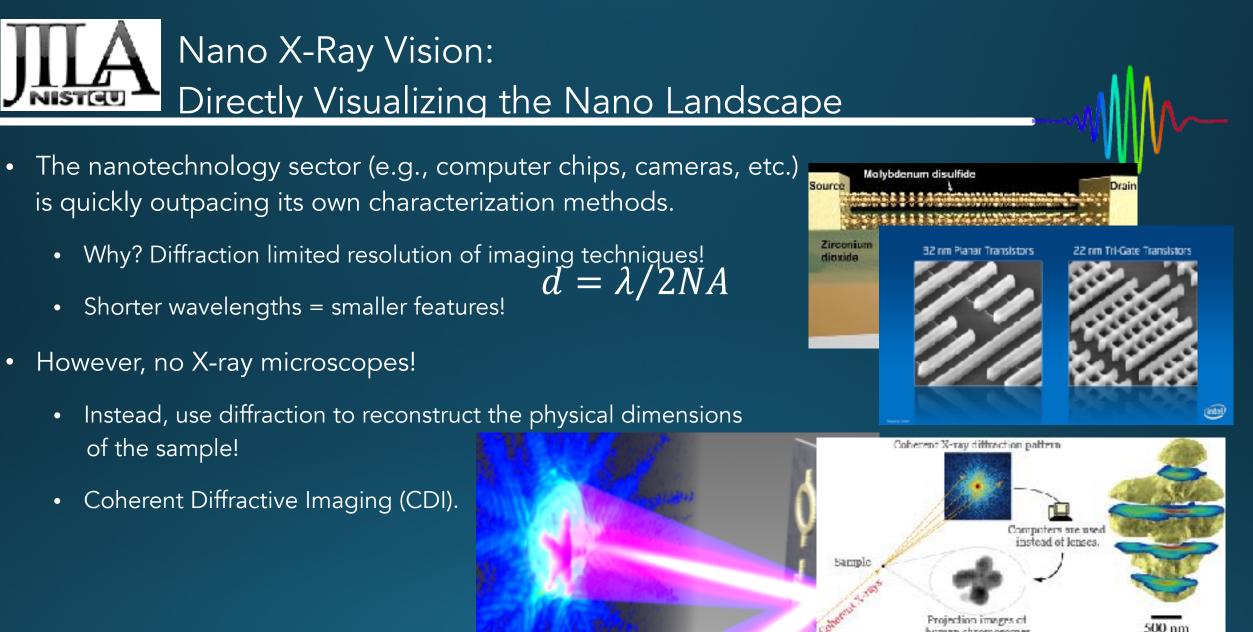
- However, no X-ray microscopes!
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  - Coherent Diffractive Imaging (CDI).



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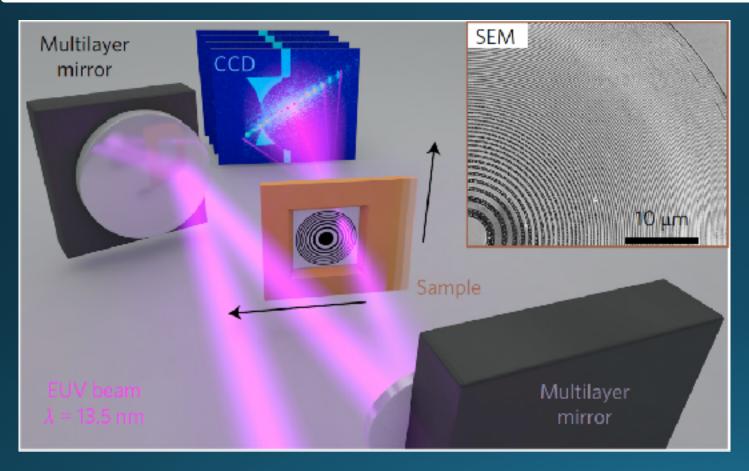


32 rm Planar Transistors	22 nm Tri-Gate Transistors
	954. 15
	001000
	Profile - Road

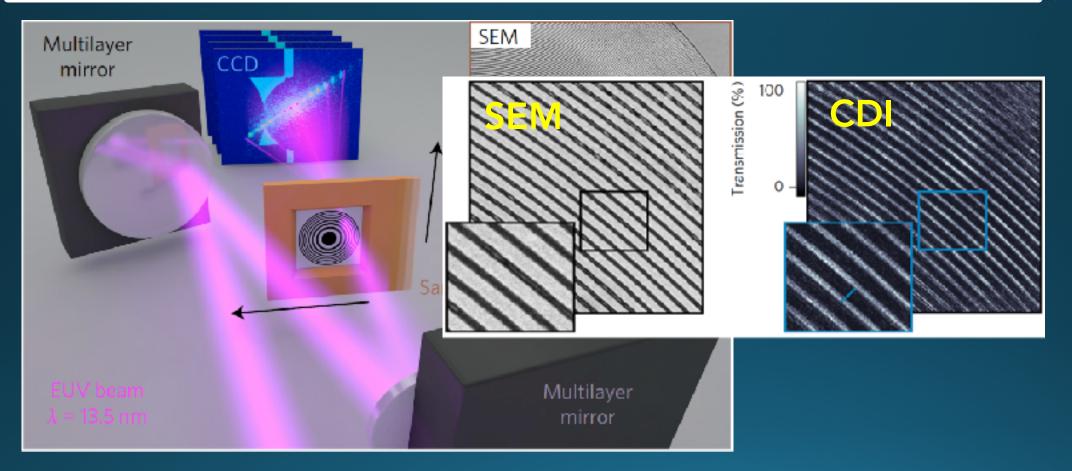


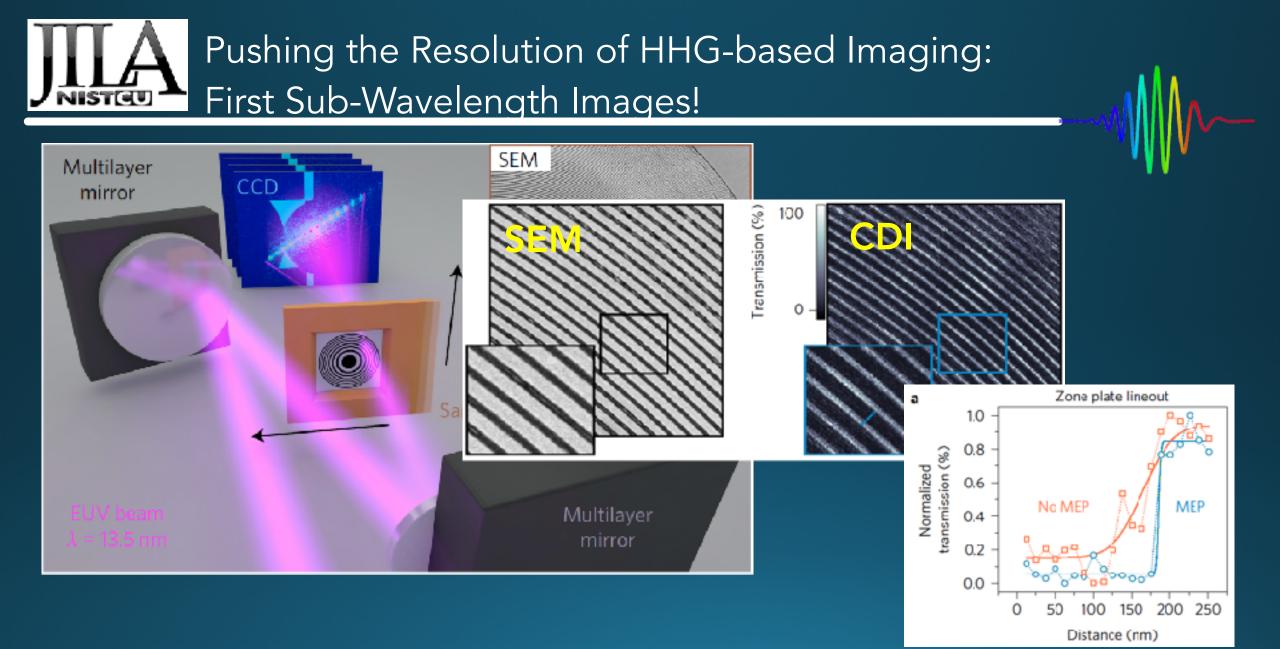
human chromosome

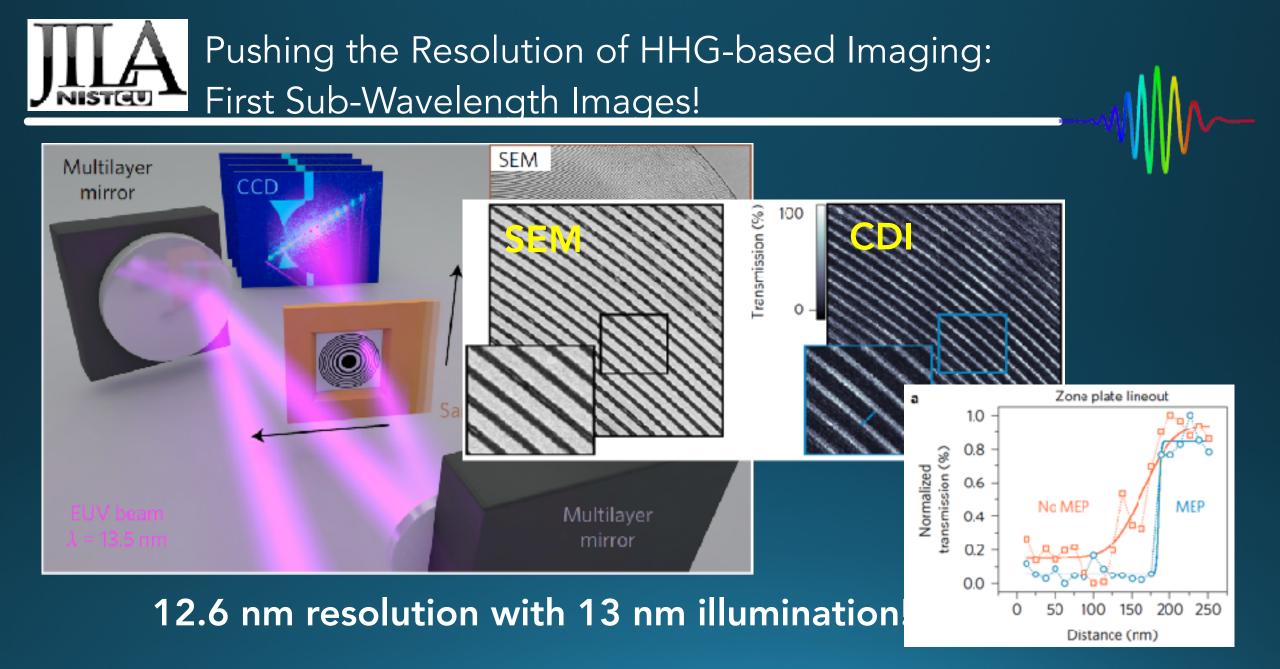
## Pushing the Resolution of HHG-based Imaging: First Sub-Wavelength Images!



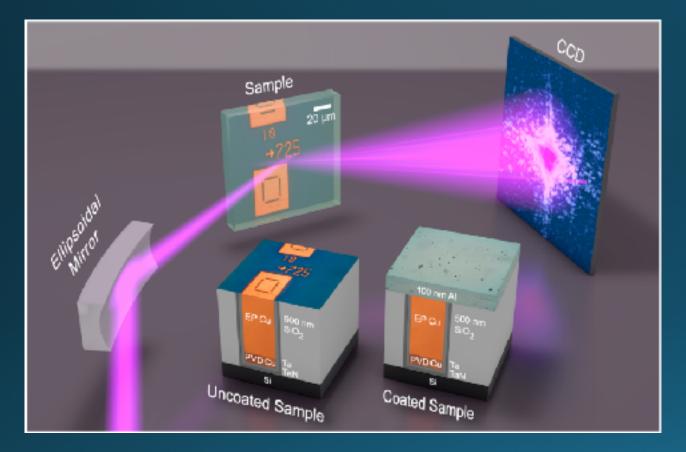
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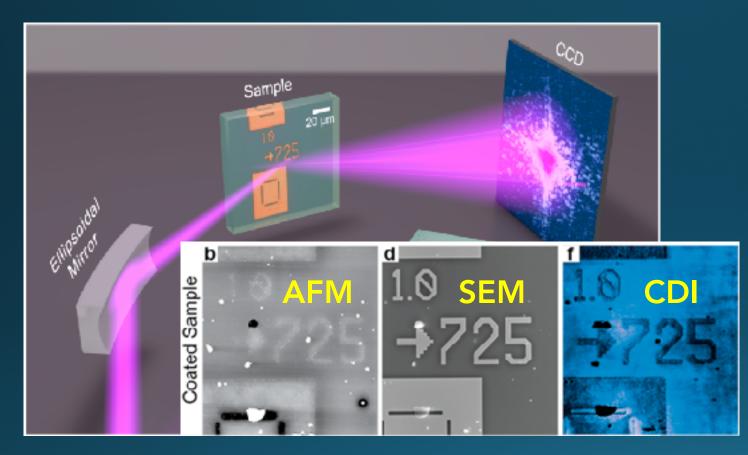




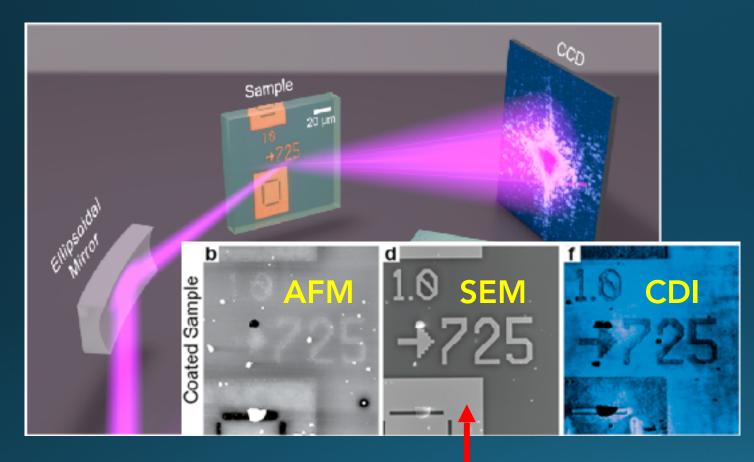






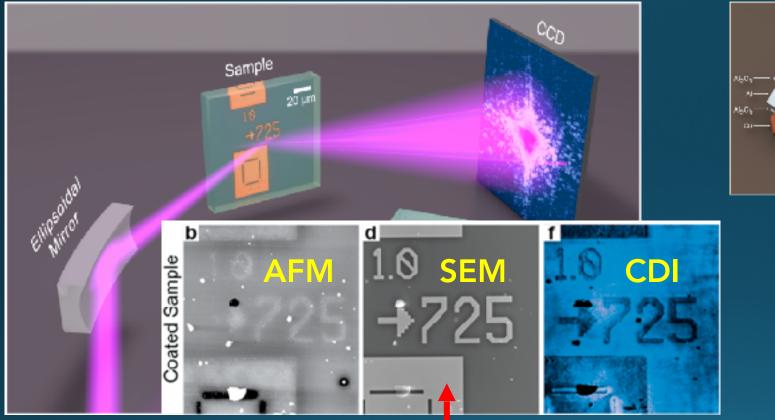


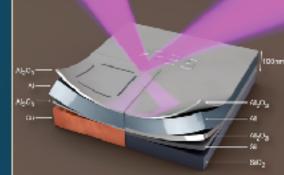




## **Destrcutive**

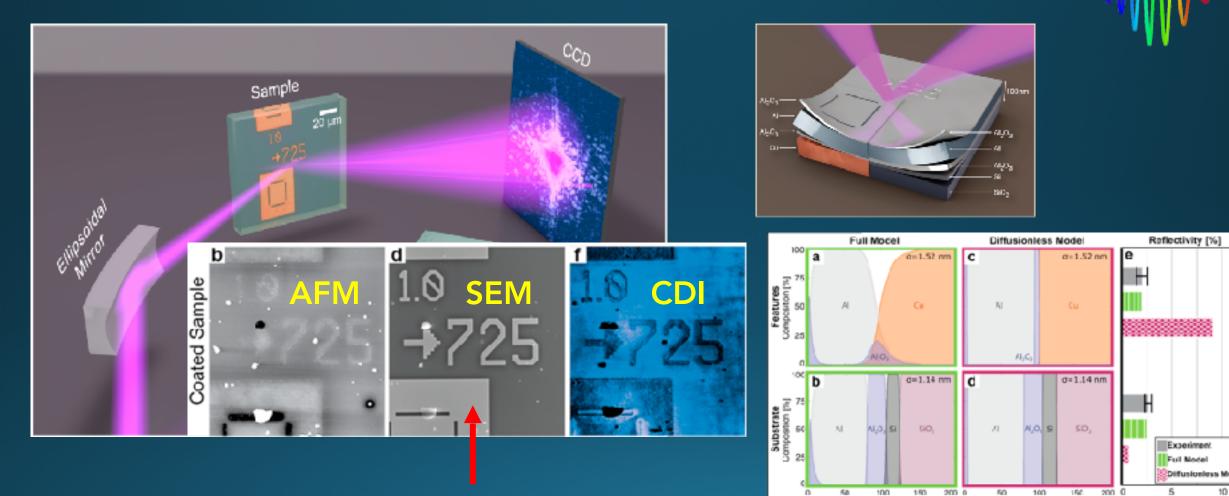






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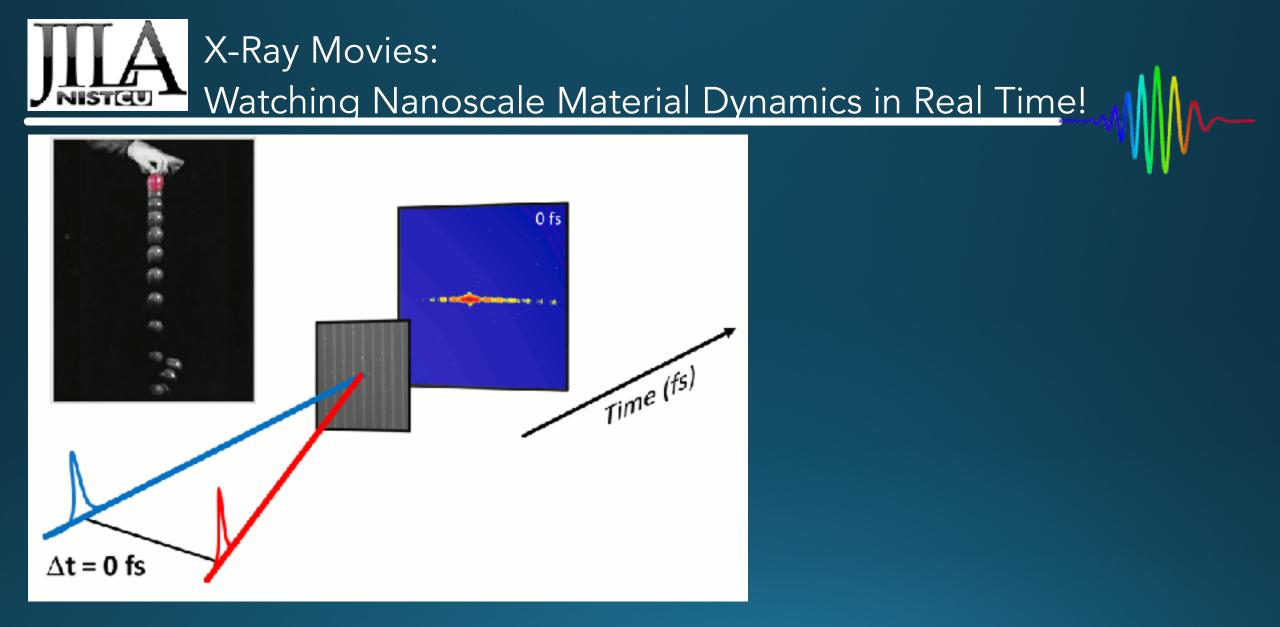


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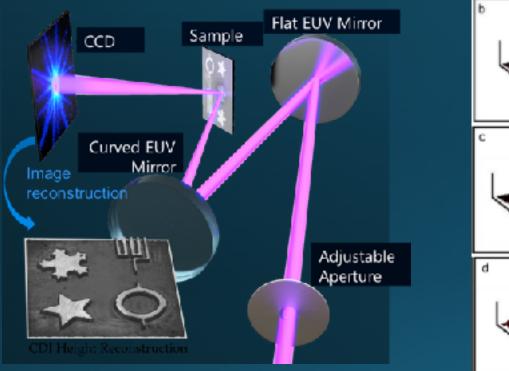
Death [nm]

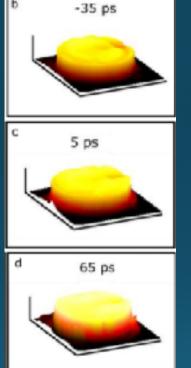
Depth [nm]

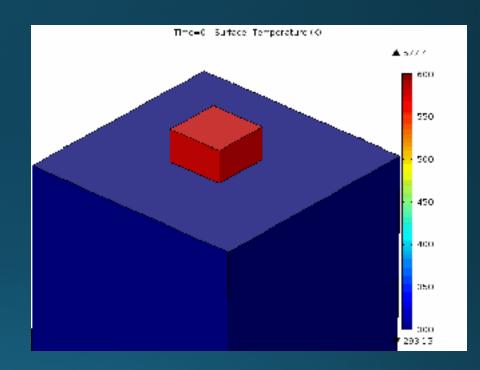






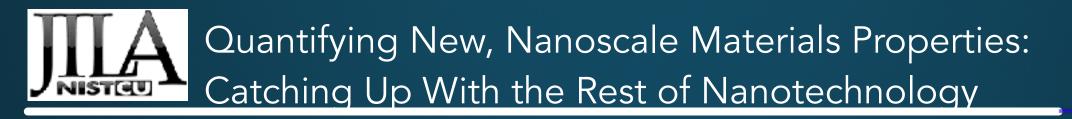






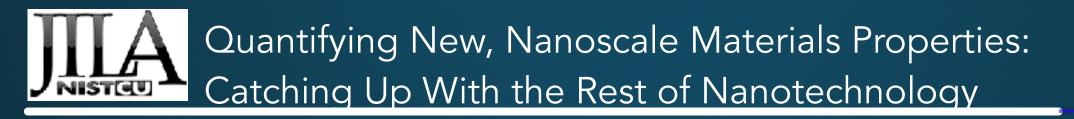
R. M. Karl, et al. Proc. CLEO, Post deadline, JTh5C.8. (2017)

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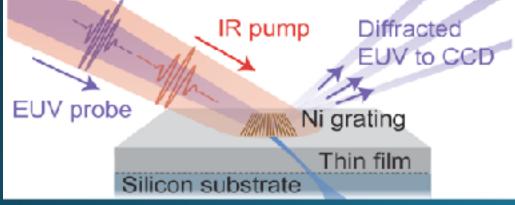
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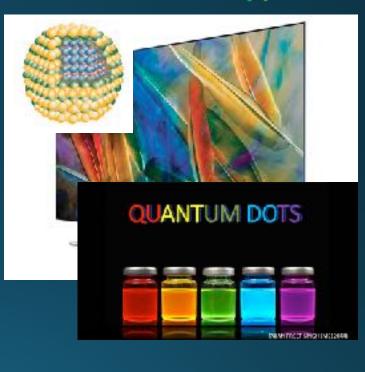


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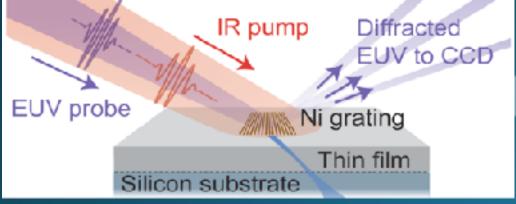
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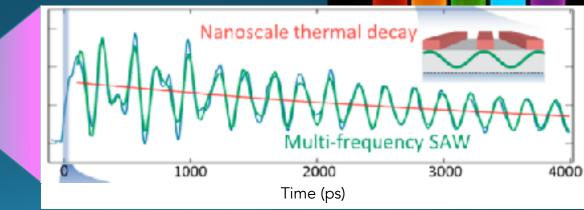




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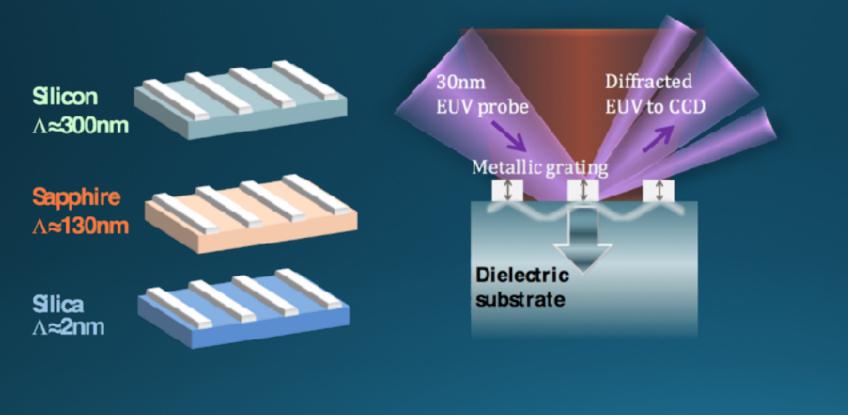




QUANTUM DOTS



• Novel electron transport properties illuminated by HHG!





Novel electron transport properties illuminated by HHG! • 10.4 m<sup>2</sup> K/W) 즈 sistivity Diffracted 30nm 200 400 Silicon EUV to CCD EUV probe ∆**≈300nm** iteracting multi-MFI ffective bou Metallic grating Sapphire Ú. ∆≈**130nm** Linewidth (nm) Dielectric substrate Silica ∆**≈2n**m

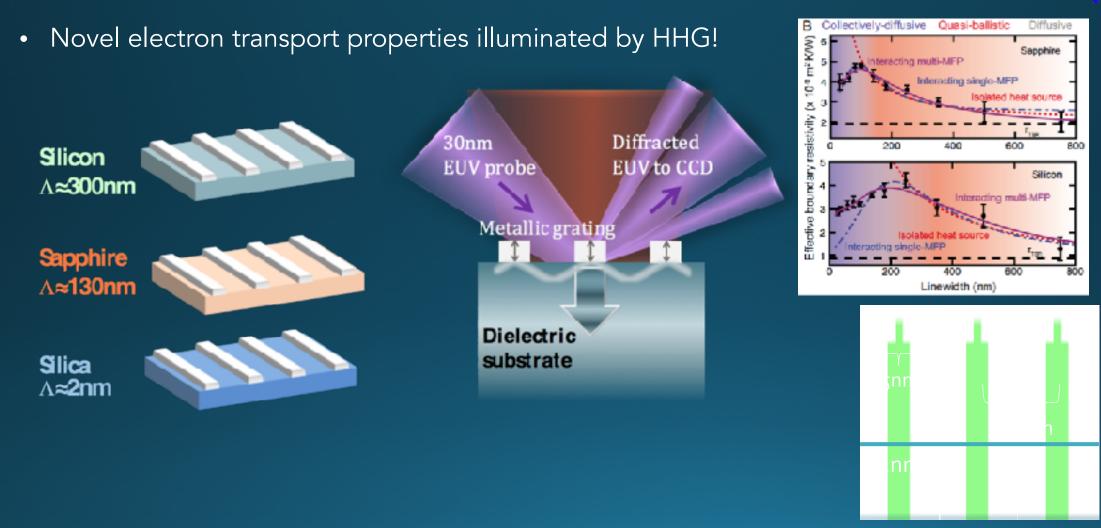
Sapphire

Silicon

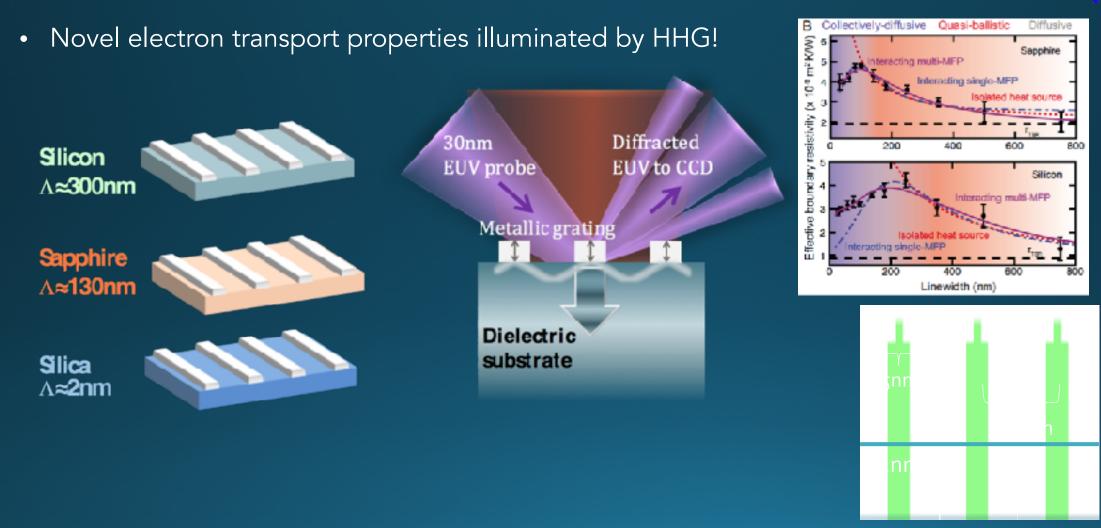
800

600

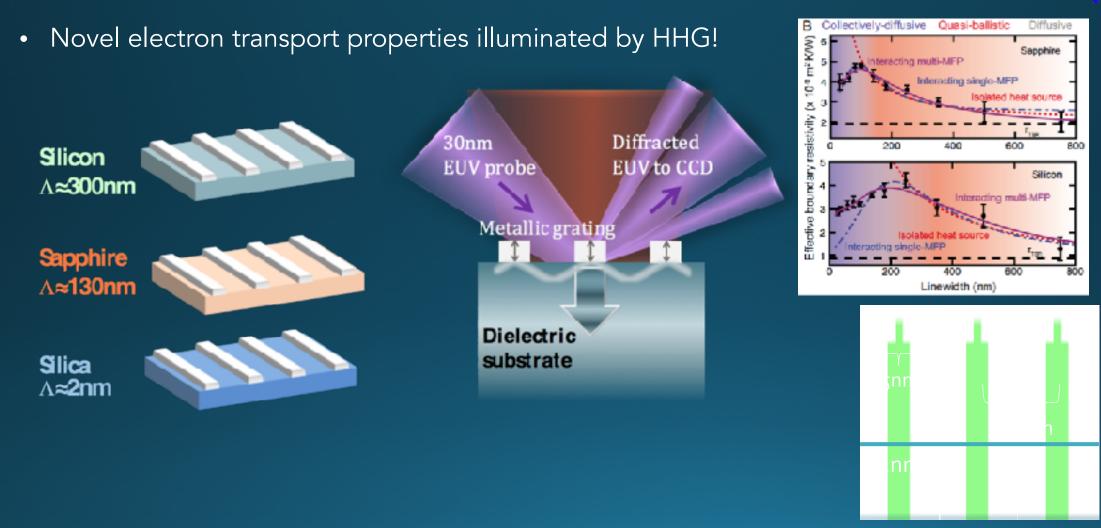




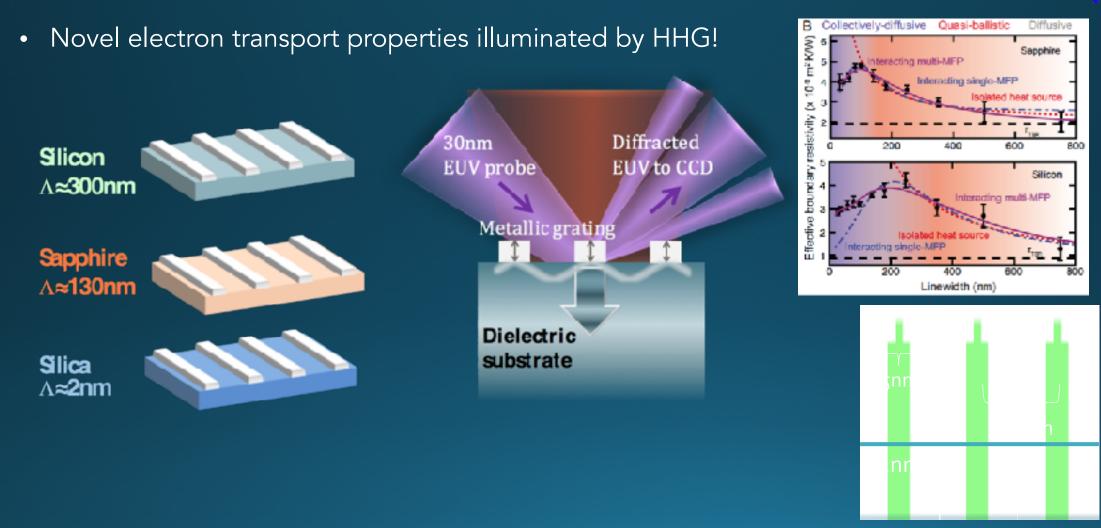










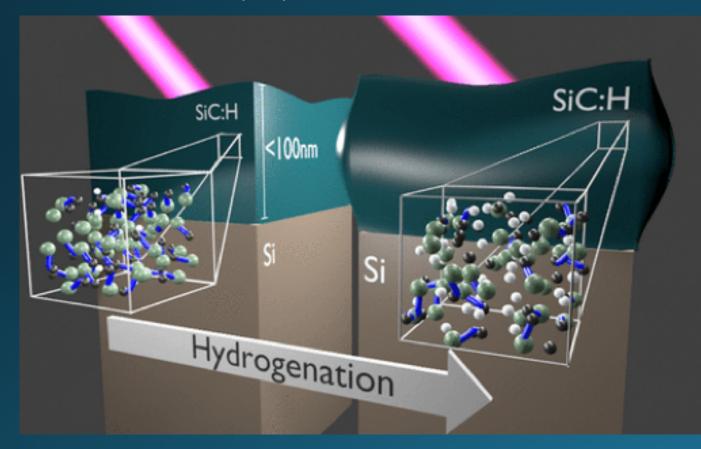




• New mechanical properties emerge for nanoscale thin films!

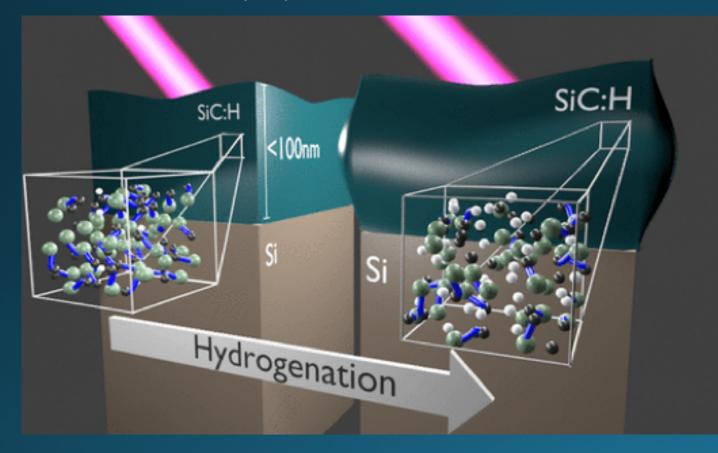


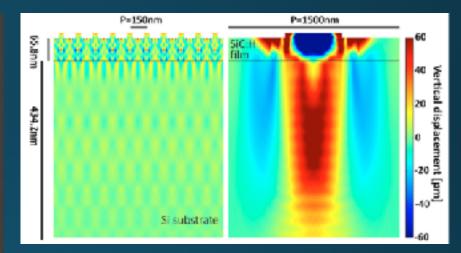
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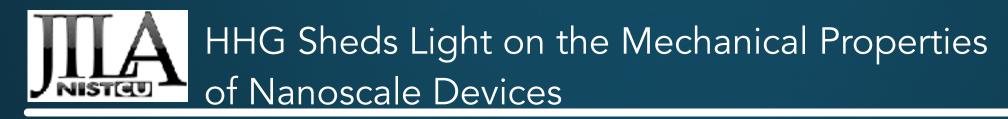




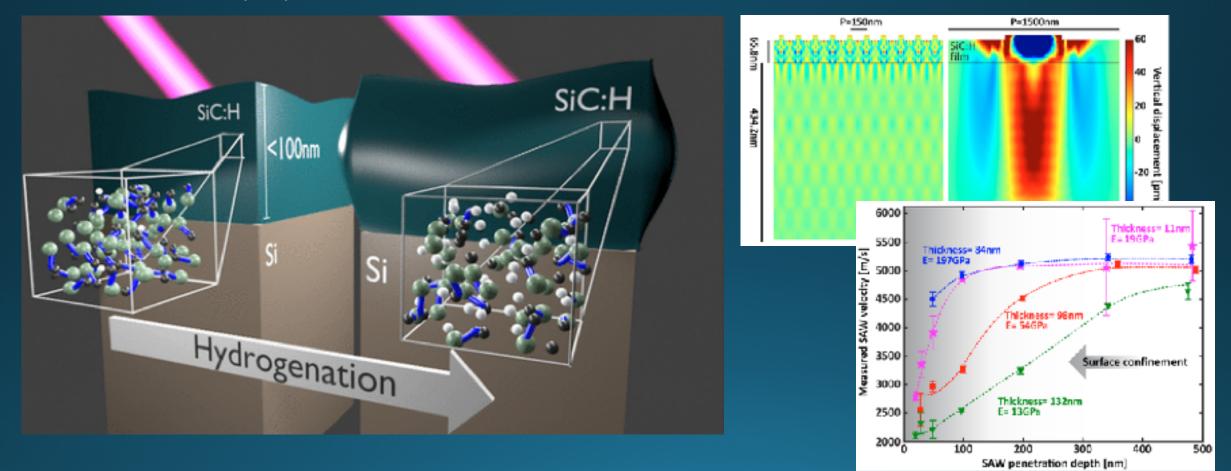
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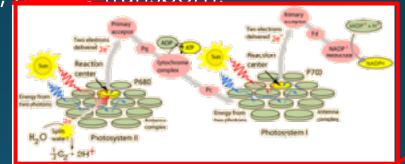
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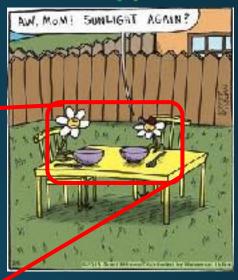


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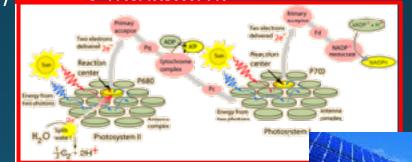


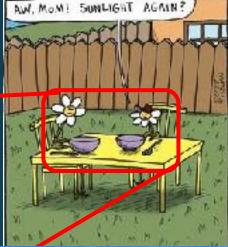
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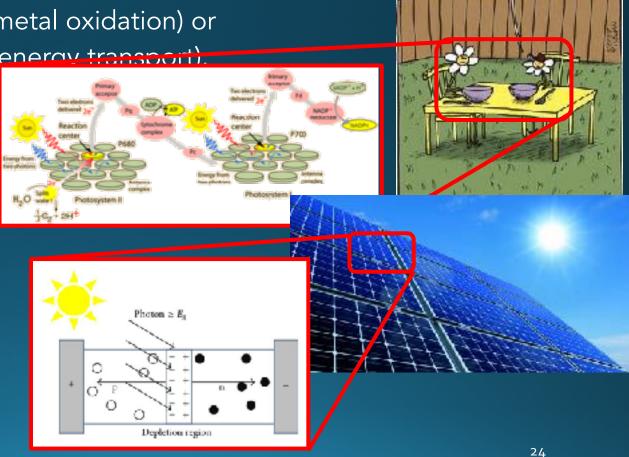


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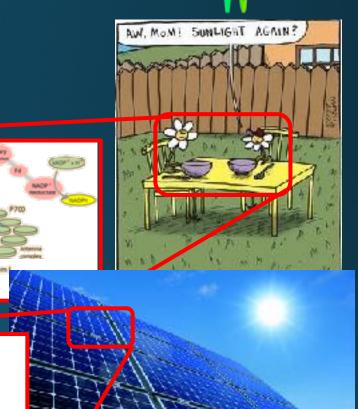


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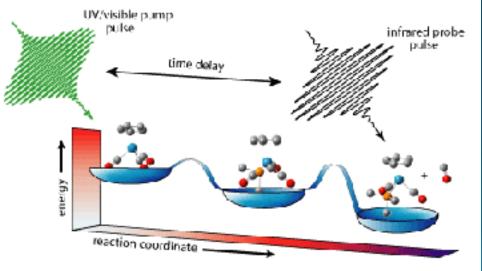
AW. MOM! SUNLIGHT AGAIN

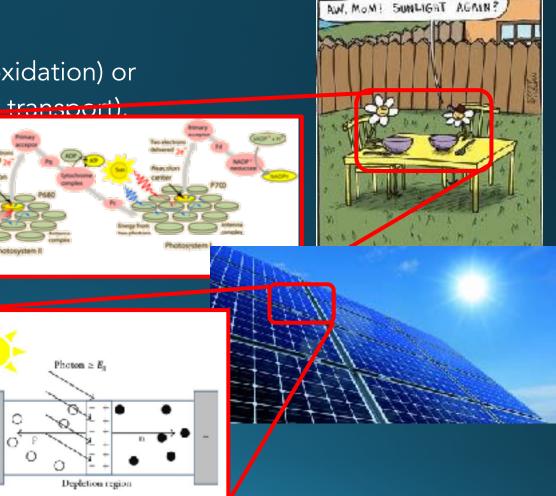
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Photon  $\geq E$ .

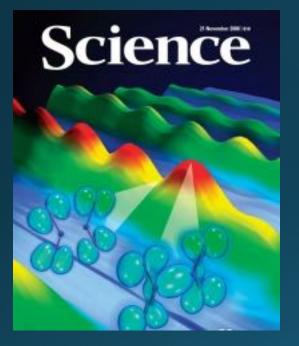
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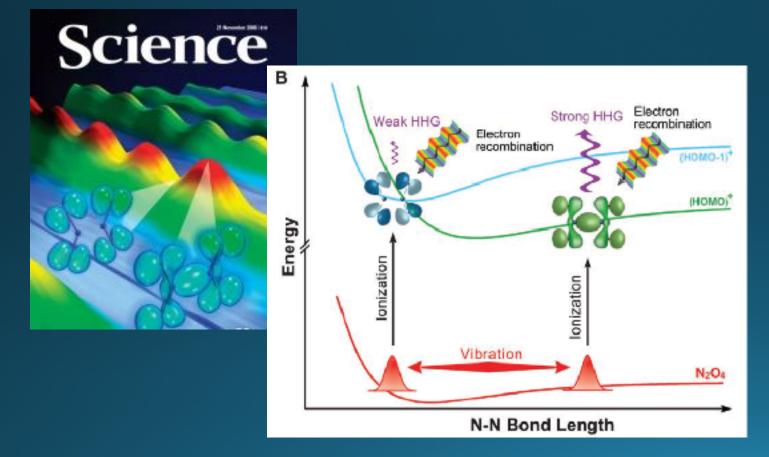




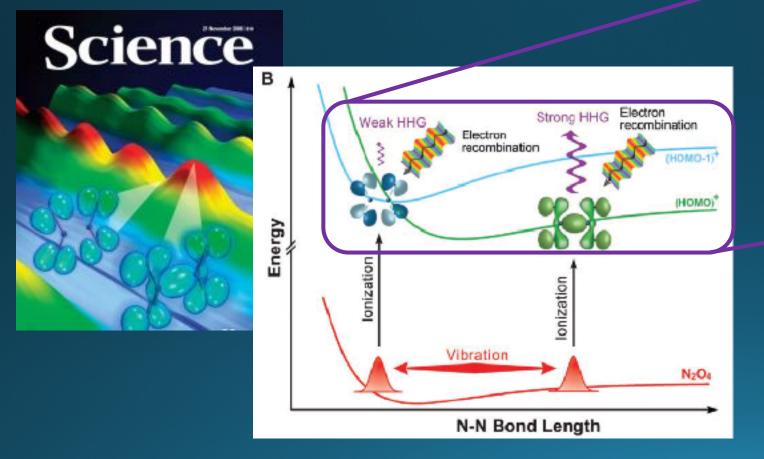


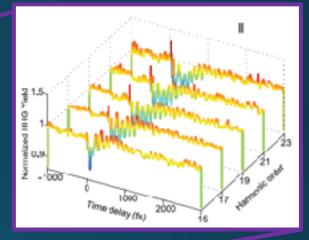




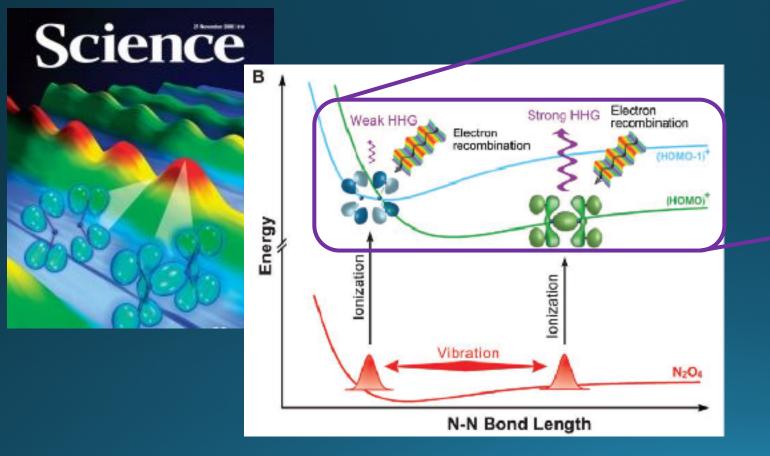


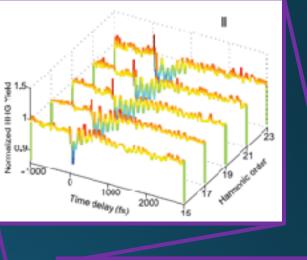
# Resolving Vibrational Dynamics of Molecules via High Harmonic Spectroscopy

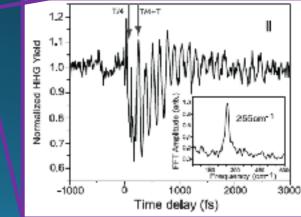




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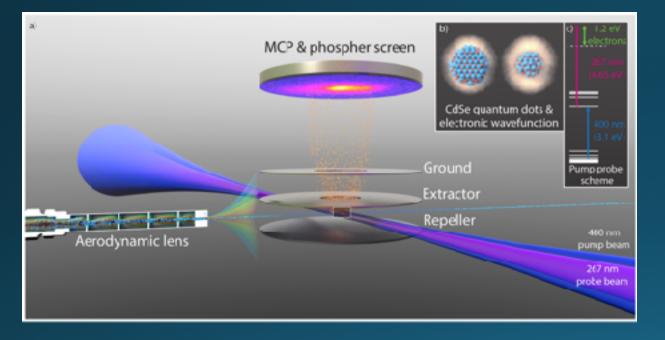




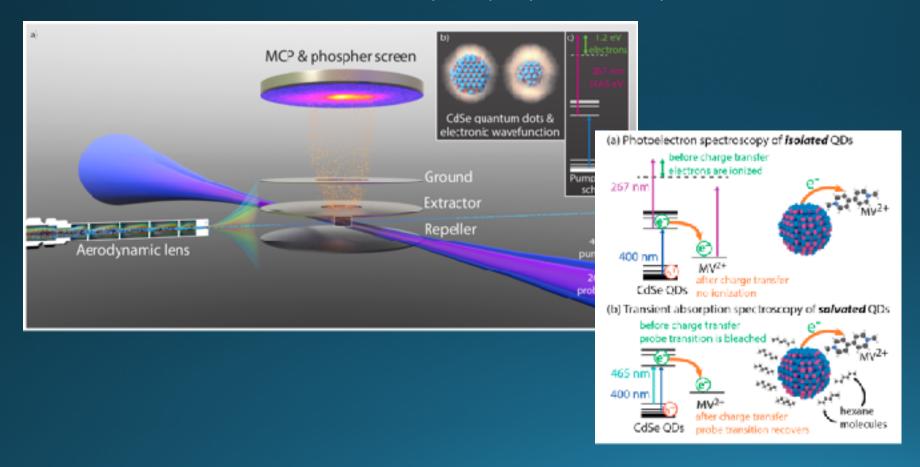


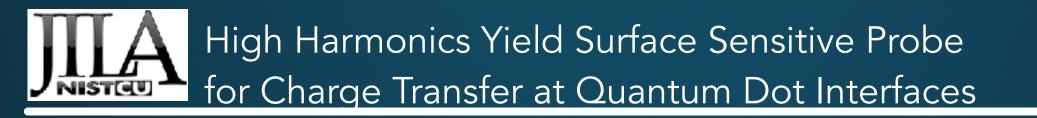


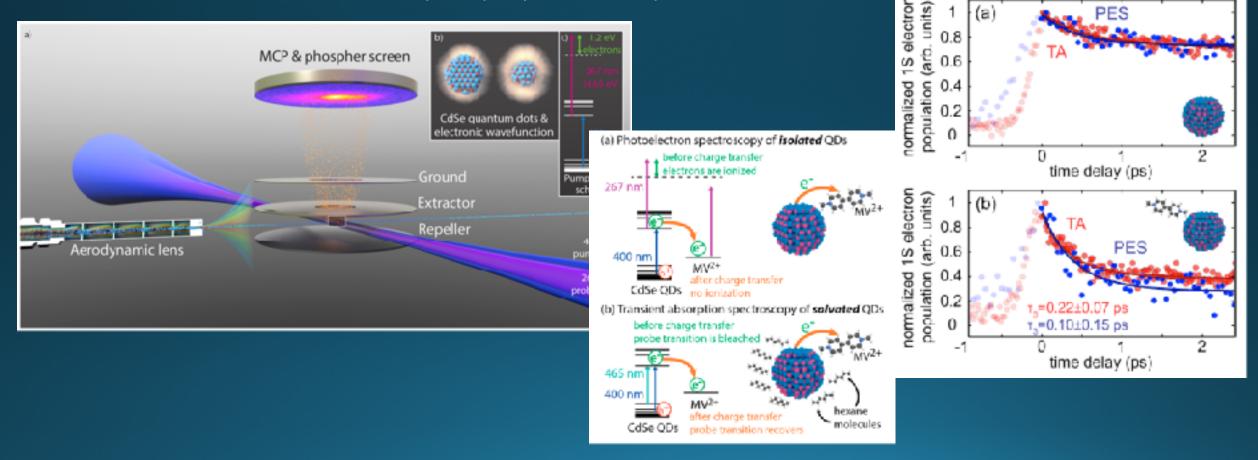












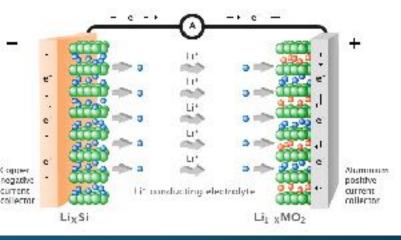
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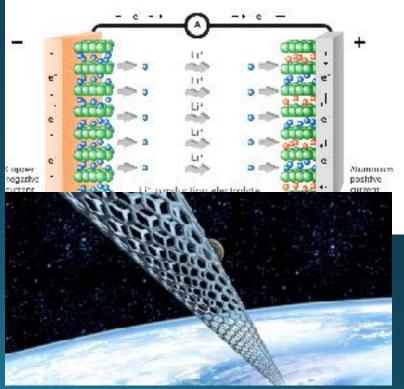
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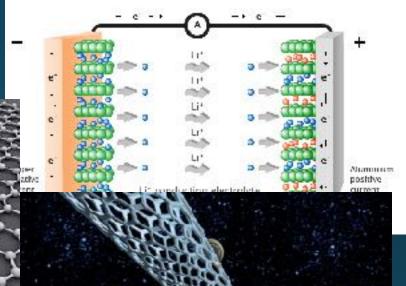
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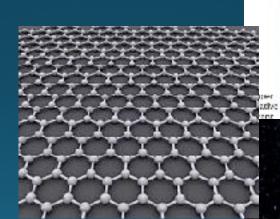
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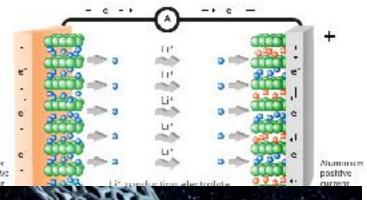


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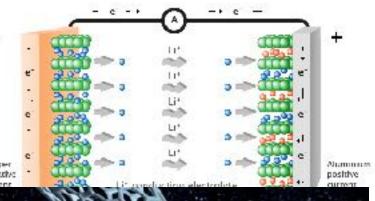




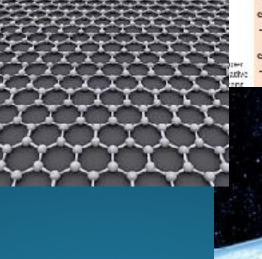


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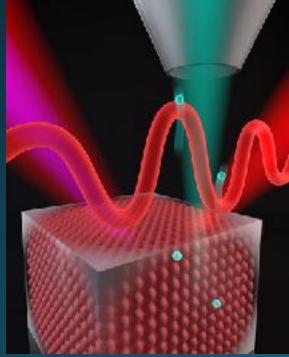


- Contrary to popular belief, photoionization is not an instantaneous process!
- It takes time for electrons to leave their home, and the time it takes depends upon where they live.



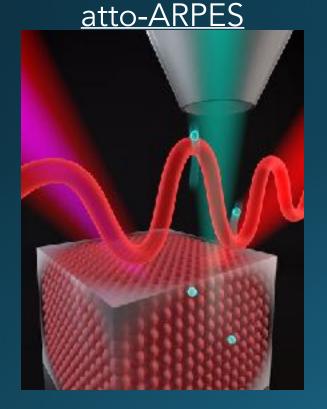
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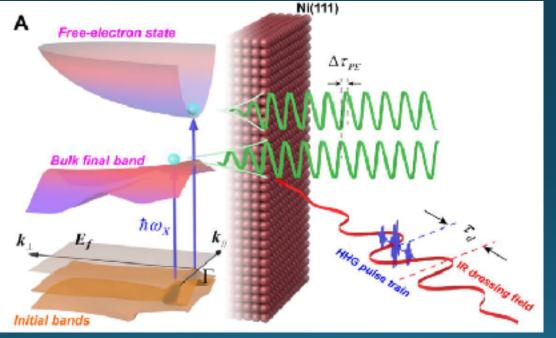






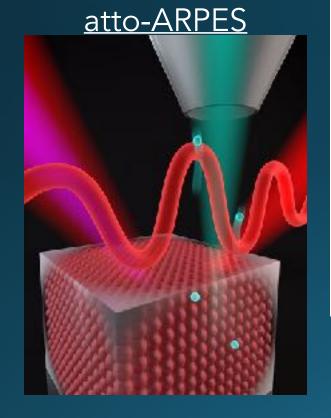
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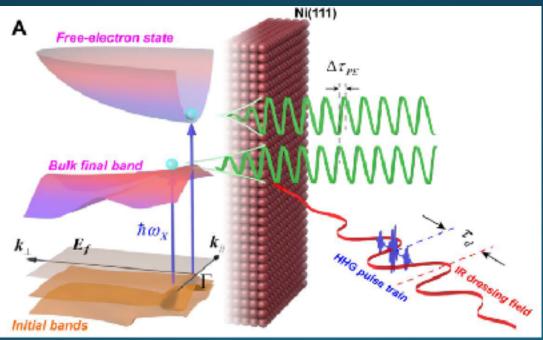


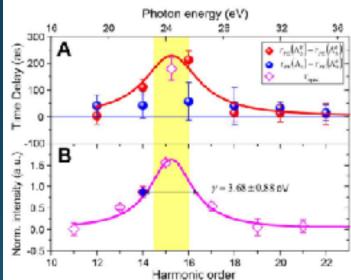




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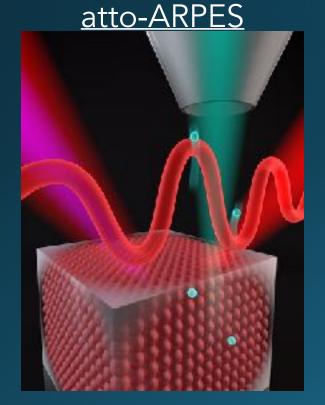


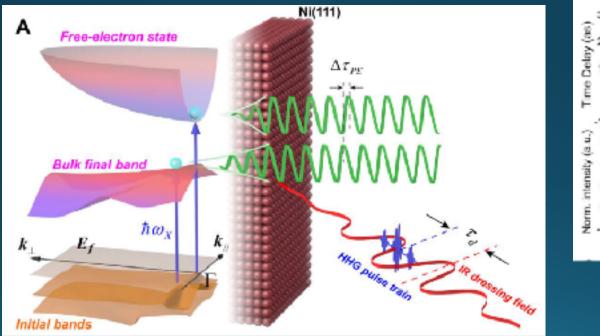


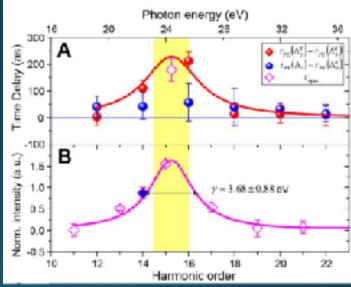




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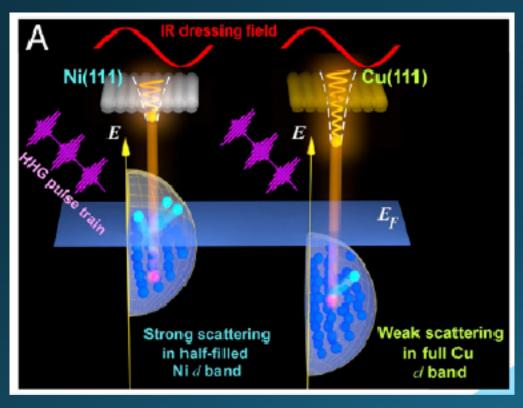
Shortest state-resolved lifetime measured... ~ 200 attoseconds!



• The time it takes an electron to leave a material is highly dependent on the local environment...



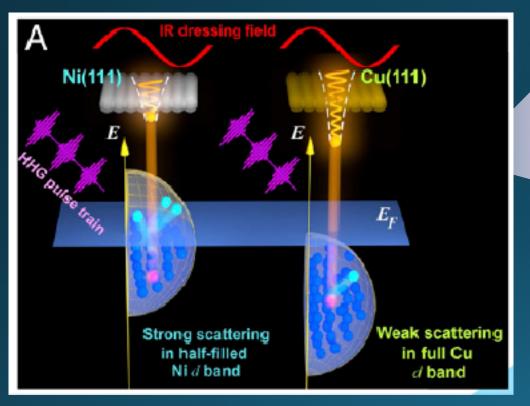
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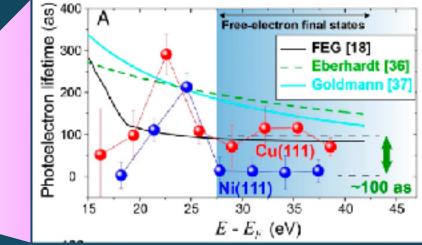




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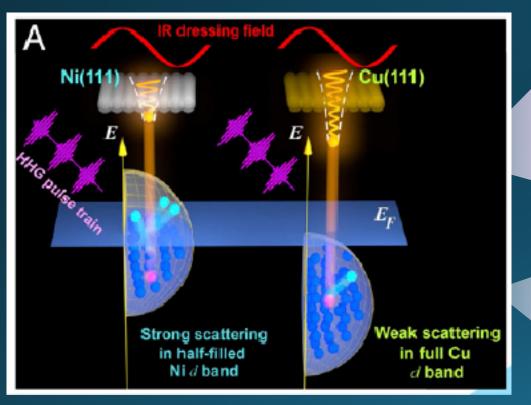


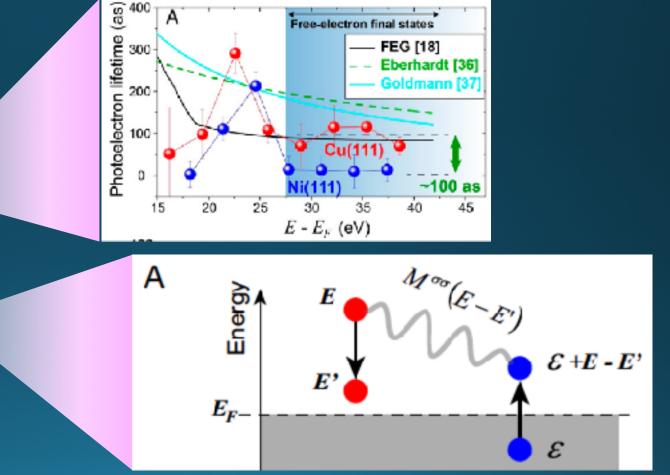




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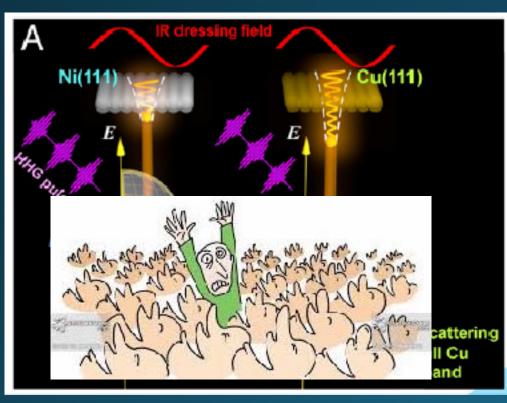


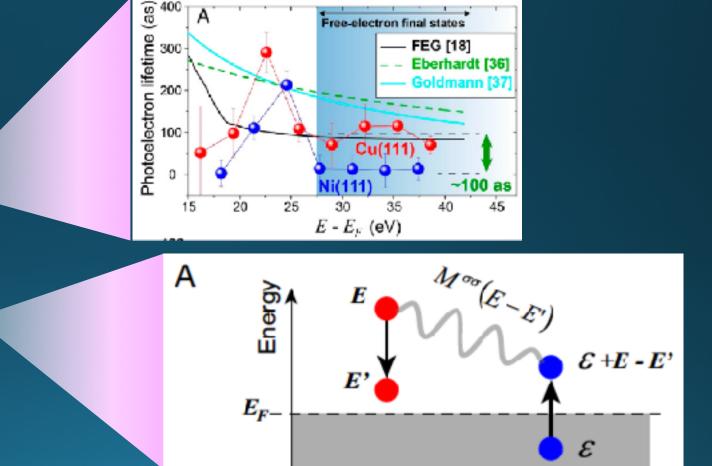




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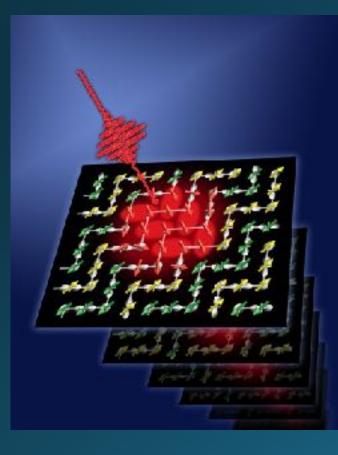




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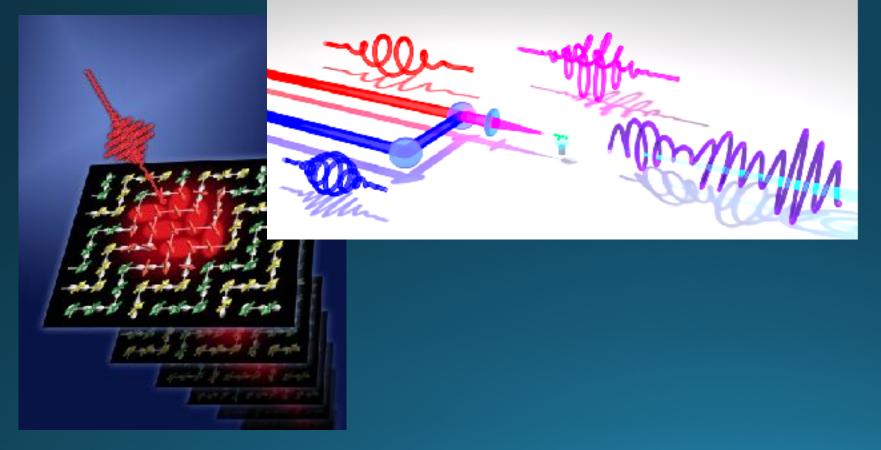


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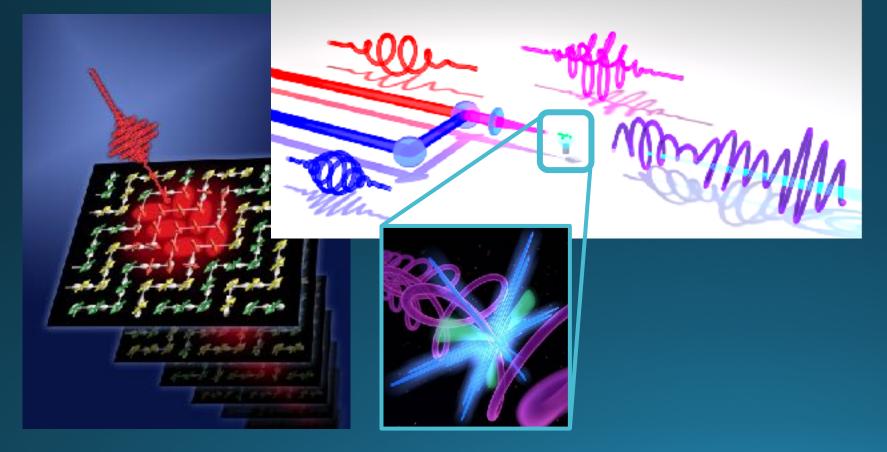


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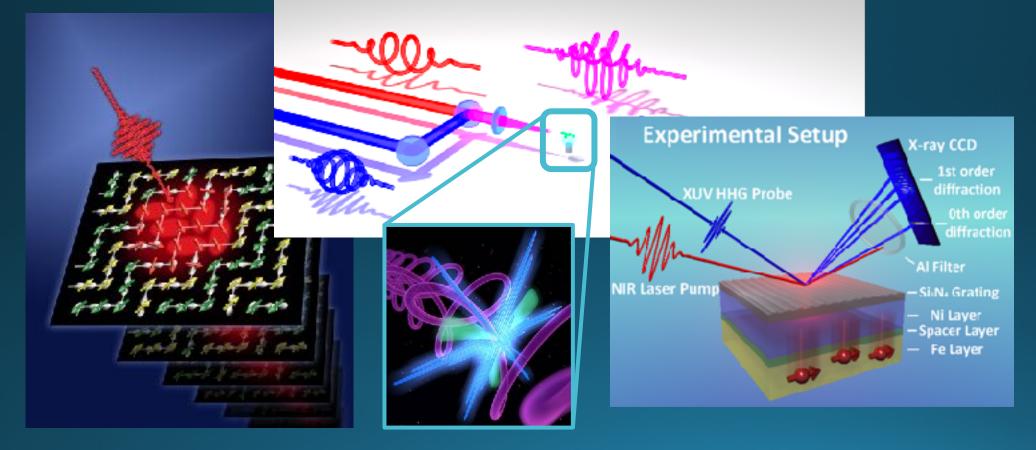


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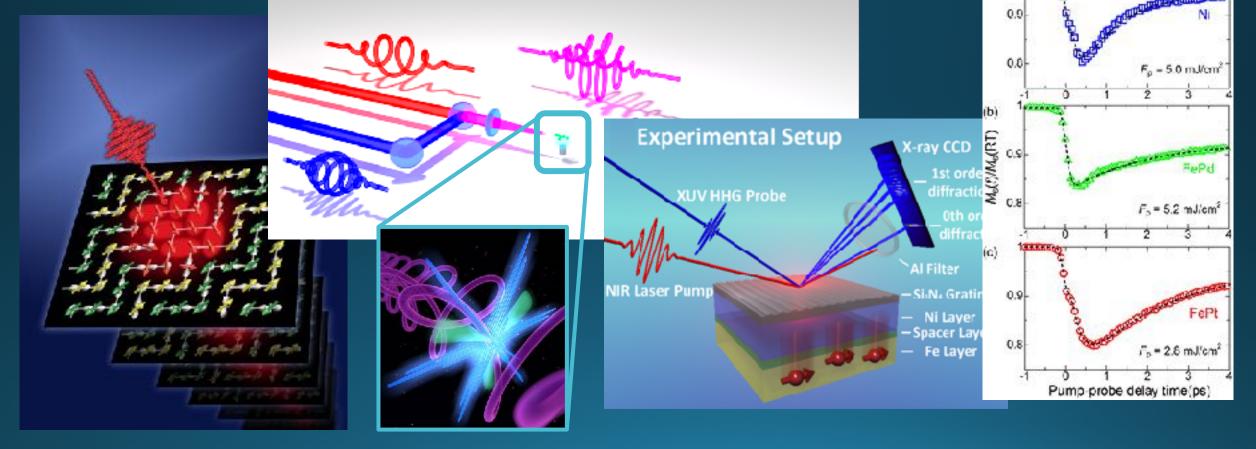


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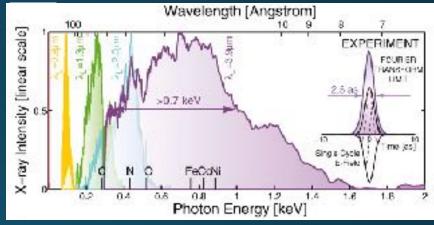
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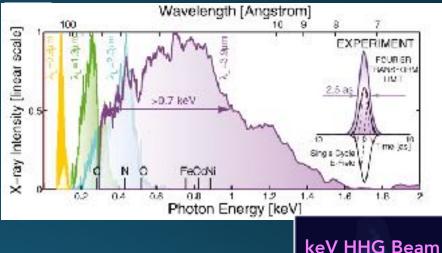
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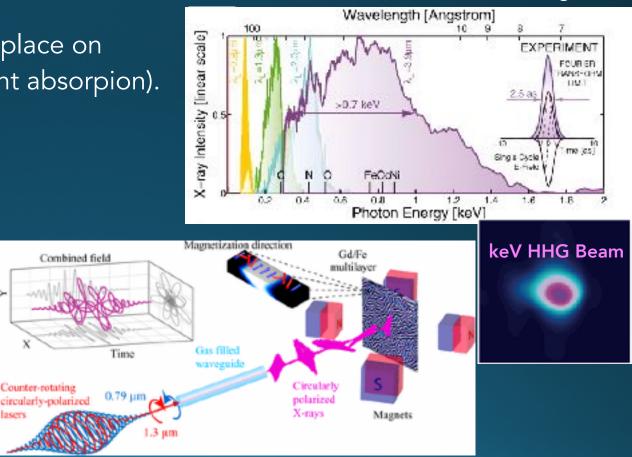




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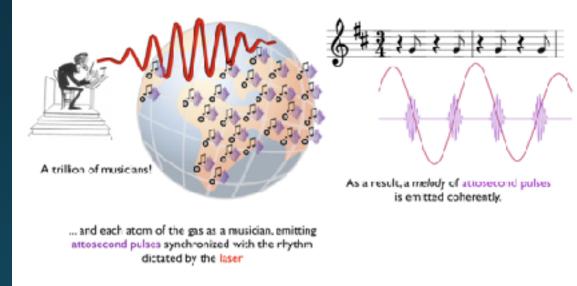
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31

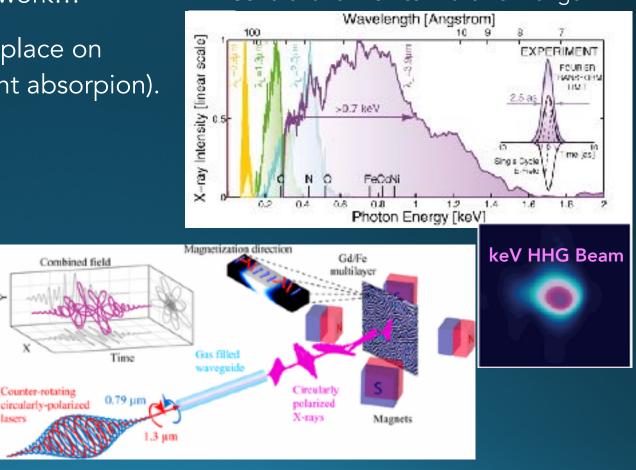


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Laser as the orchestra director...



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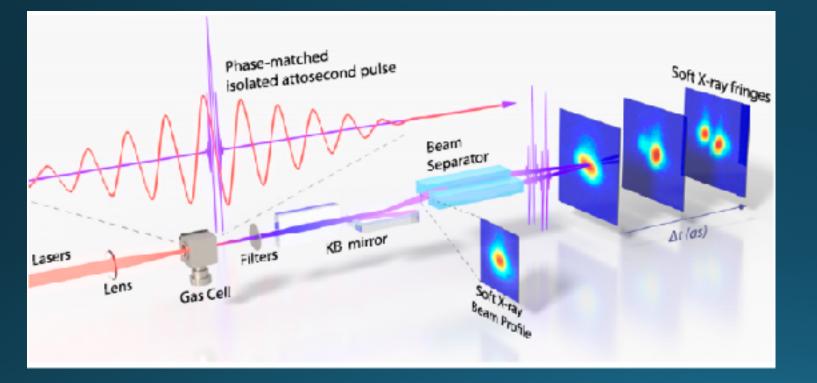




• Carefully changing the "tune" of the HHG process yields isolated attosecond pulses!

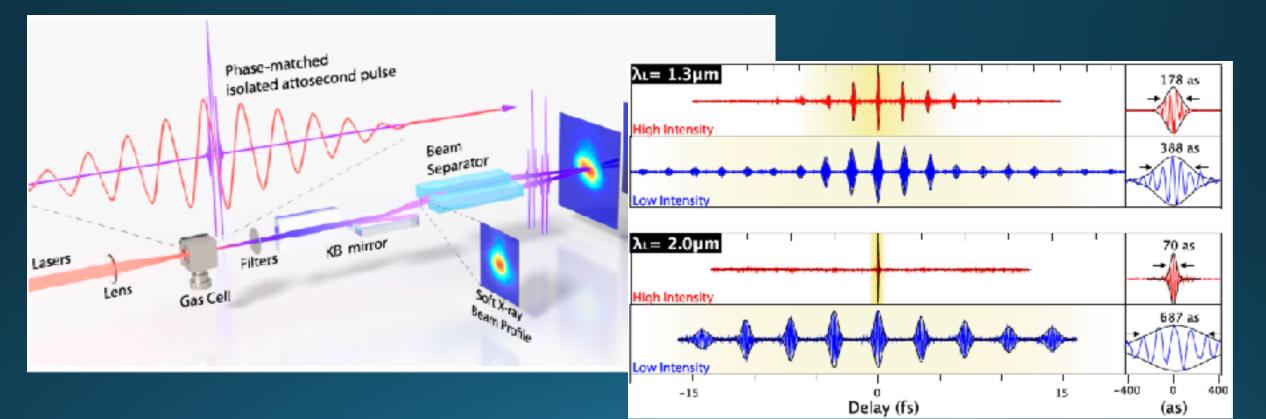


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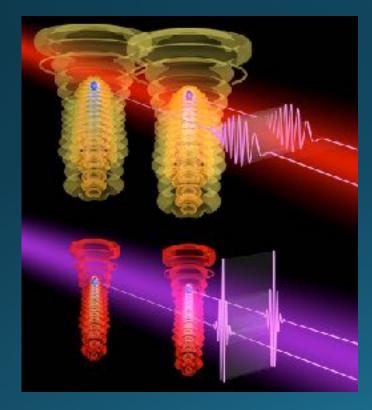




• Driving the HHG process with UV light yield bright, narrowband harmonics spanning many elemental absorption edges!

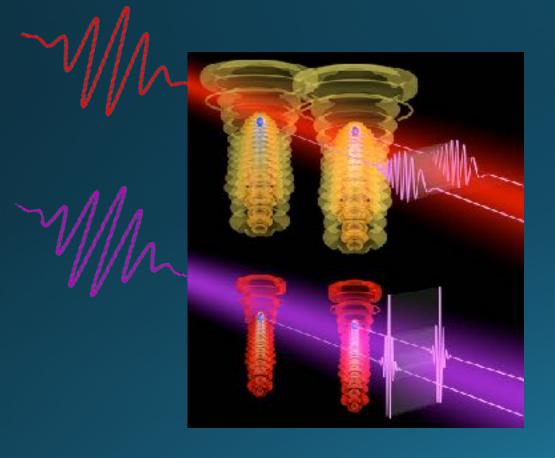


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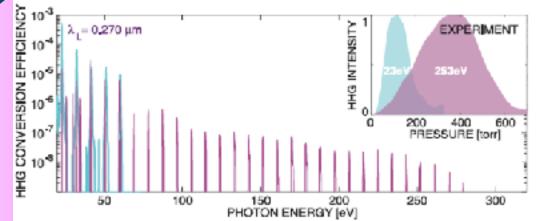


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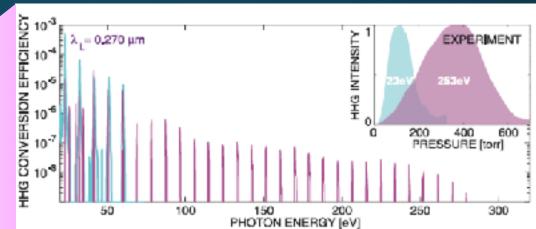
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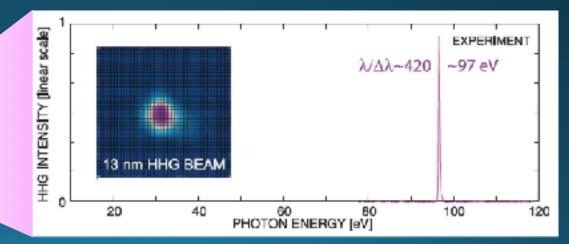
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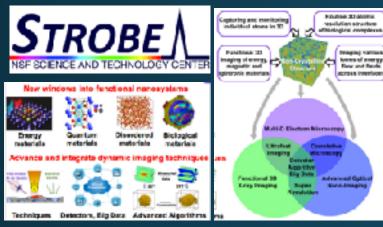
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## Future of Ultrafast Light and Materials Science: Probing Nature at Its Fundamental Limits NISTEU

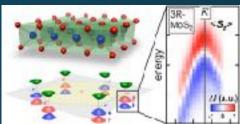
## **Real Time Functional Imaging**



# **Attosecond Dynamics**

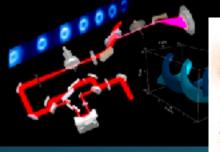
## <u> Tracking the Ultrafast Migration of</u> **Charge in Correlated Materials**

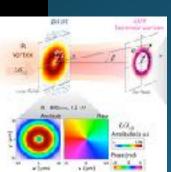
Momentury



Mott insulator

#### **Structured Light Beams for** Advanced Spectroscopies





### New Sources of Ultrafast Laser

