Nobel Prize in Physics for Attosecond Physics

Stockholm U, Dec 8, 2023

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Anne L'Huillier (Lund)
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OSU red carpet, March 20, 2024

Pierre Agostini
Brutus

Feynman's lesson on Nature

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the outcome of a quantum process is dictated by the sum over all the quantum trajectories that contribute to it.

- we analyze based on these trajectories, but we don't necessarily measure them

attosecond clocking of strong field quantum trajectories

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menu:

- synopsis of strong field atomic physics
- semi-classical unified view
- quantum trajectory selector (QTS) concept
- building the QTS
- clocking rescattering

building a quantum trajectory selector for strong field physics

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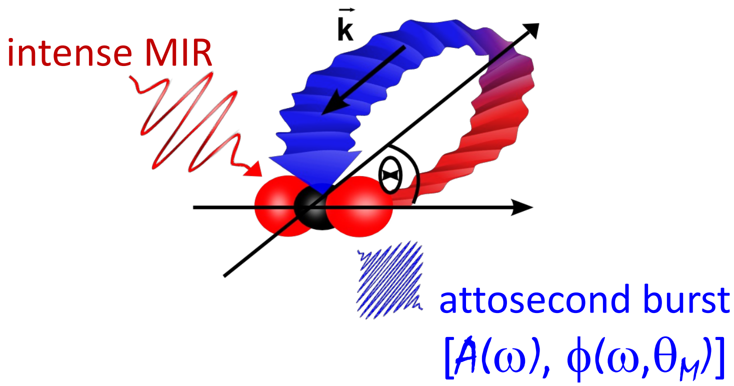
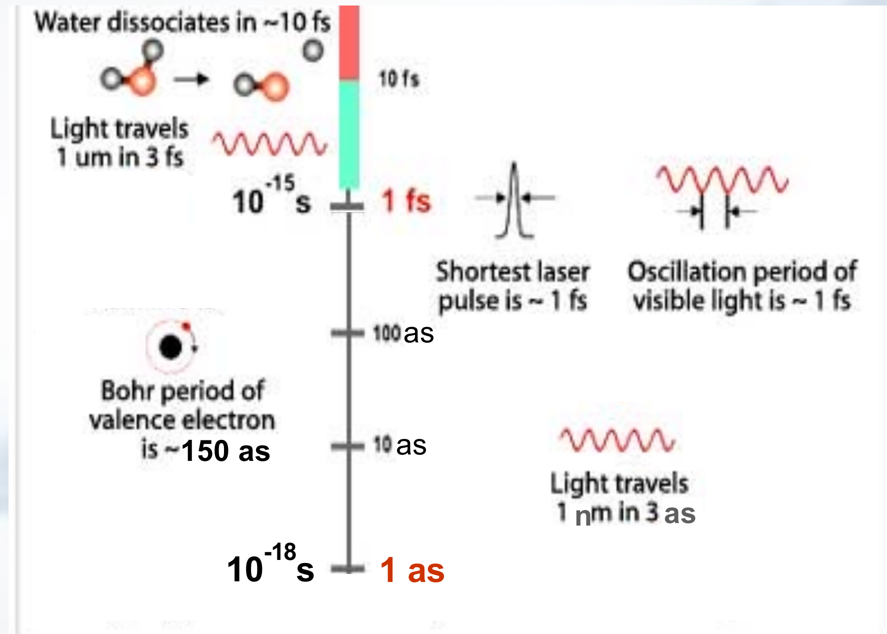
1993: rescattering physics
Kulander & Corkum



the birth of sub-cycle physics

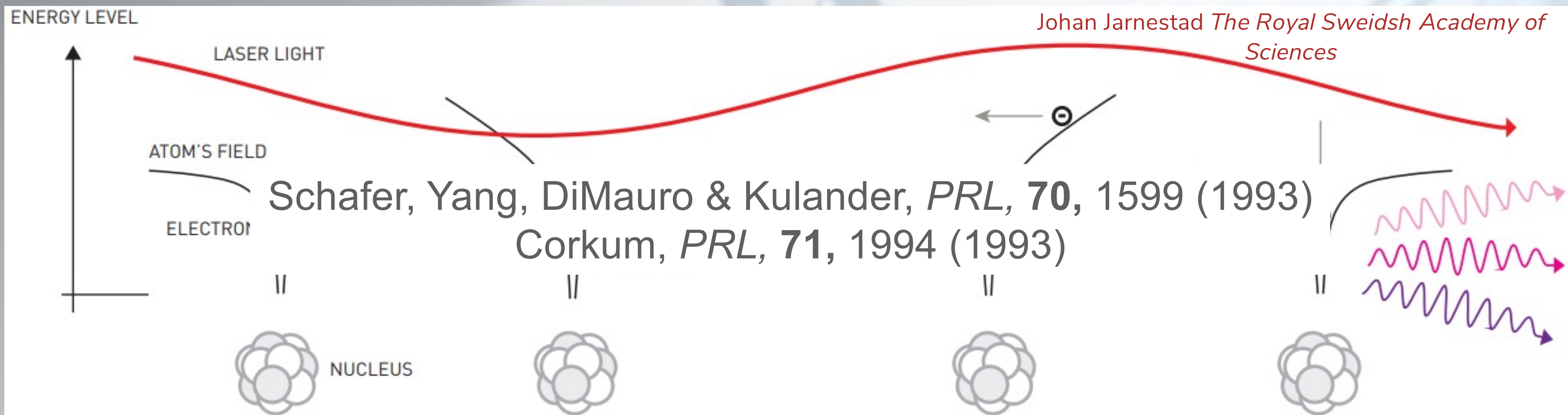
2001: Vienna & Paris attosecond era



can attosecond light teach us more about strong field physics?

strong field physics enabled attosecond science!

semi-classical physics: 3-step or rescattering model



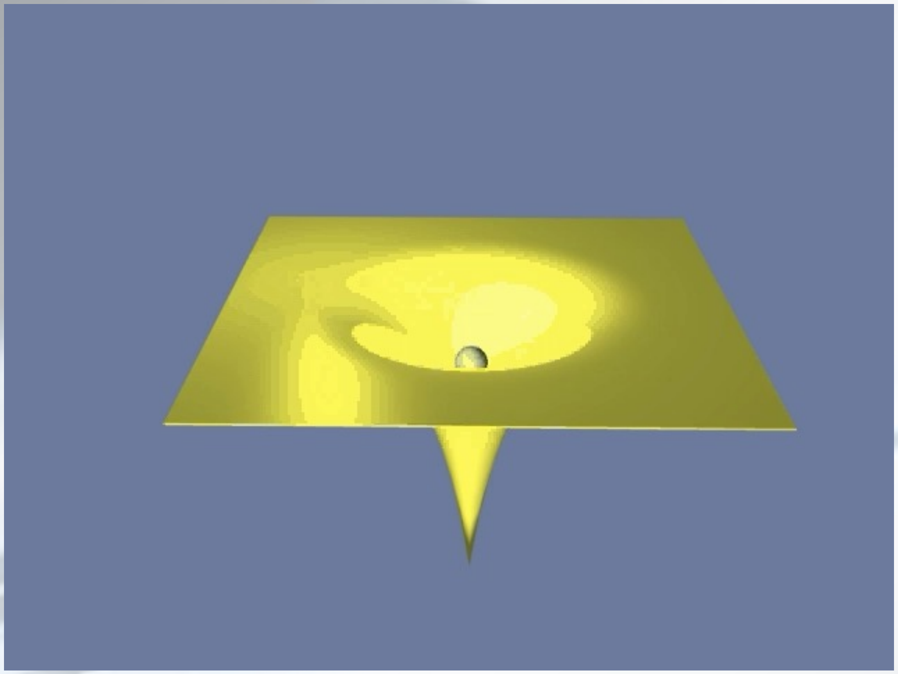
Schafer, Yang, DiMauro & Kulander, *PRL,* **70,** 1599 (1993)
Corkum, *PRL,* **71,** 1994 (1993)

1. Ionization 2. Propagation 3. Recombination

- ✓ recollision provides a simple, intuitive view of strong field interactions in the tunneling regime
- ✓ tunneled electron wave packet dynamics dominated by the field thus U_p (quiver energy) becomes a good ruler
- ✓ initial conditions defined by tunnel ionization

rescattering or 3-step model: the cartoon

Schafer, Yang, DiMauro & Kulander, *PRL,* **70,** 1599 (1993)
Corkum, *PRL,* **71,** 1994 (1993)



tunneling + propagation + interaction

- ✓ classical trajectories have different release time (phase), propagation time and harmonic emission times & energy
- ✓ maximum *return energy* is $3.17U_p$
- ✓ maximum *electron energy* $10U_p$
- ✓ physics is inherently sub-cycle

recollision heralds the attosecond science era

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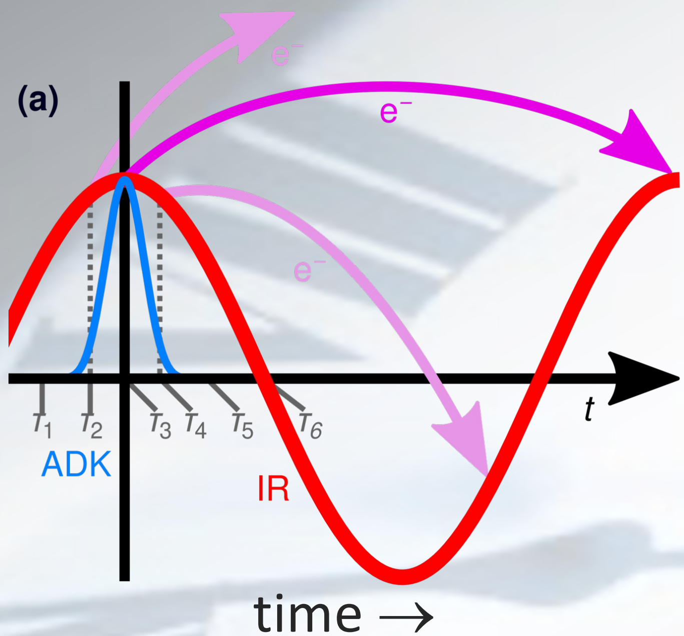

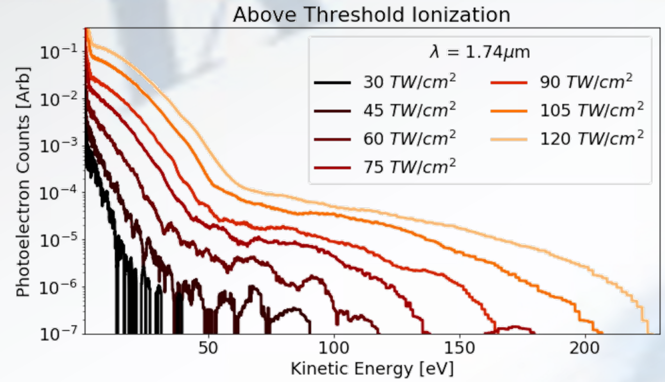

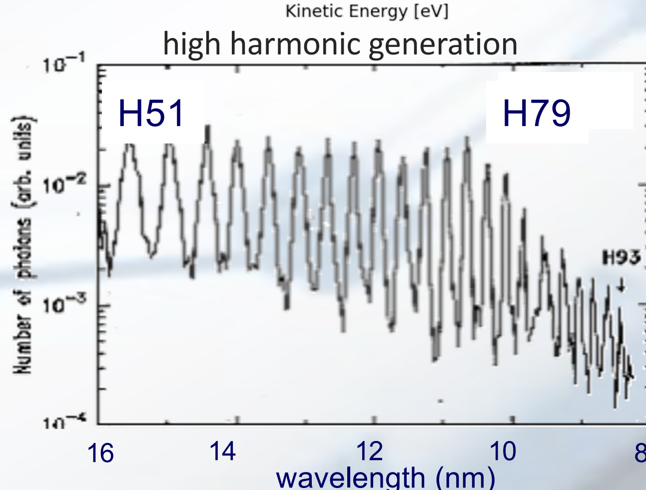

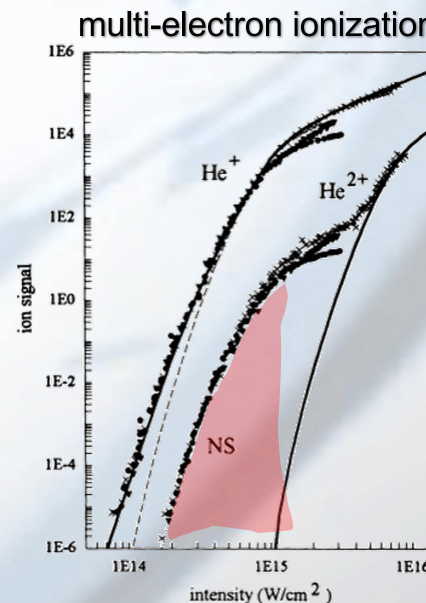

initial conditions defined by tunnel ionization ($\gamma << 1$)

tunnel ionization in the optical regime

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Keldysh picture of strong field tunnel ionization

a sub-cycle view of ionization

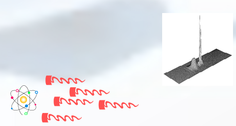
SOVIET PHYSICS JETP VOLUME 20, NUMBER 5 MAY, 1965

IONIZATION IN THE FIELD OF A STRONG ELECTROMAGNETIC WAVE

L. V. KELDYSH

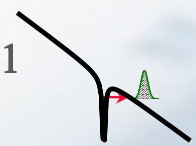
$$\gamma \equiv \frac{\text{optical frequency}}{\text{tunneling frequency}} = (IP/2U_p)^{1/2} \propto \omega/\sqrt{I}$$

$\gamma > 1$



"photon description"

$\gamma < 1$



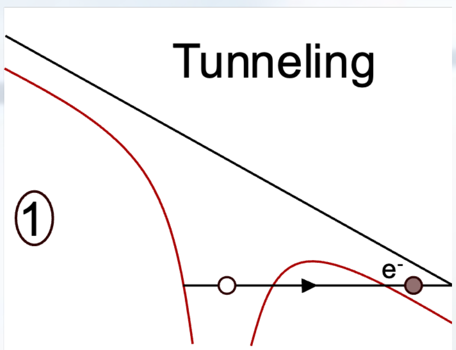
"dc-tunneling picture"

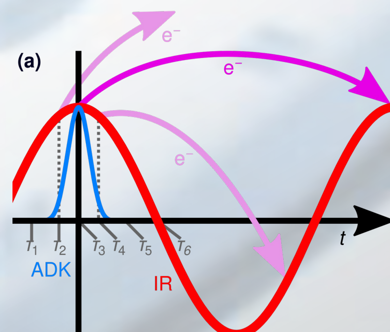
tunnel ionization: initial conditions

tunneling wave packet characteristics

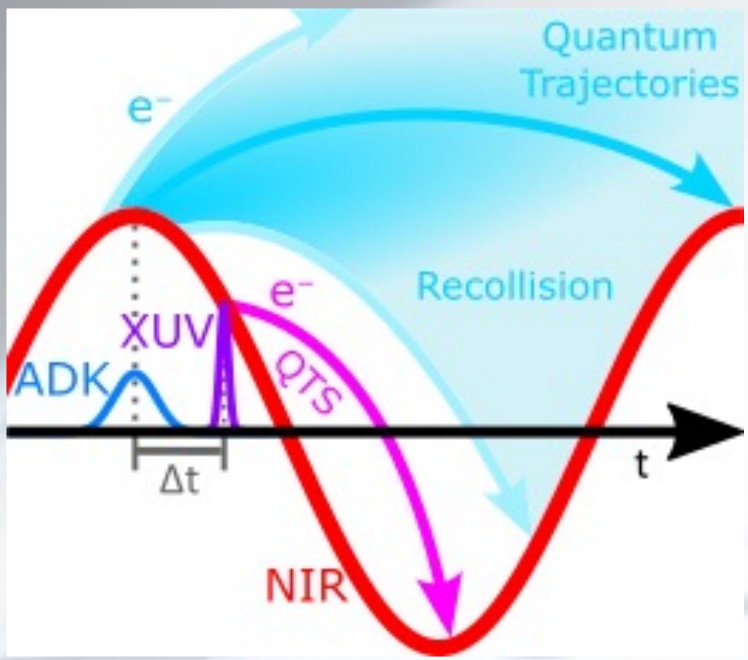
- ✓ all tunneling WP look the same
- ✓ born outside the potential well with near zero velocity
- ✓ ionization is highly nonlinear (exponential) ⇒ limits the intensity range for measurements
- ✓ wave packet (WP) born at peak of ac-field ⇒ initial phase fixed

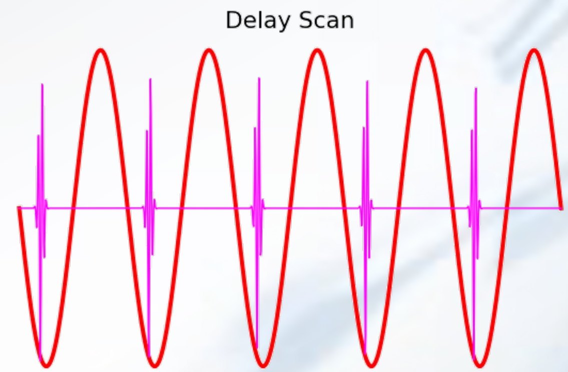
the experimentalist has limited control of the initial condition



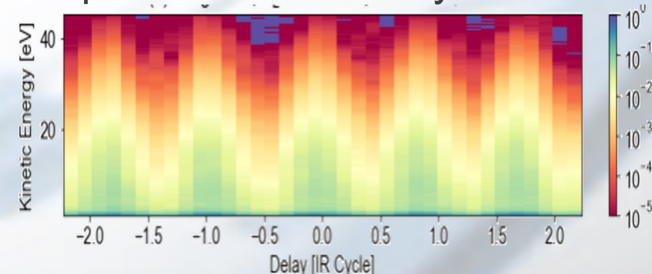


quantum trajectory selector: attosecond control





photoelectron delay scan



goal: use XUV to control the ionization time with attosecond precision and observe recollision

past efforts: control of HHG by APT seeding

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PHYSICAL REVIEW A **72**, 013411 (2005)

Large enhancement of macroscopic yield in attosecond pulse train–assisted harmonic generation

Mette B. Gaarde and Kenneth J. Schafer
Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803-4001, USA

Arne Heinrich, Jens Biegert, and Ursula Keller
ETH Zurich, Physics Department, Inst. of Quantum Electronics, CH-8093 Zürich, Switzerland

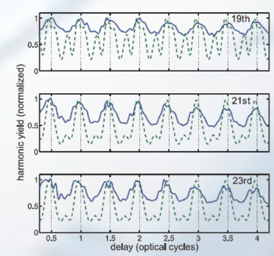
(Received 17 December 2004; published 28 July 2005)

“Control of high-order harmonic emission using attosecond pulse trains” JMO **53**, 87 (2006)

Attosecond control of electron–ion recollision in high harmonic generation

G Gademann[1,6], F Kelkensberg[1], W K Siu[1], P Johnsson[2], M B Gaarde[3,4], K J Schafer[3,4] and M J J Vrakking[1,5]

New J. Phys. **13**, 033002 (2011)



Self-probing spectroscopy of XUV photo-ionization dynamics in atoms subjected to a strong-field environment   Nat. Comm. 8 (2017)

Doron Azoury[1], Michael Krüger[1], Gal Orenstein[1], Henrik R. Larsson[2], Sebastian Bauch[3], Barry D. Bruner[1] & Nirit Dudovich[1]

quantum trajectory selector (QTS) method

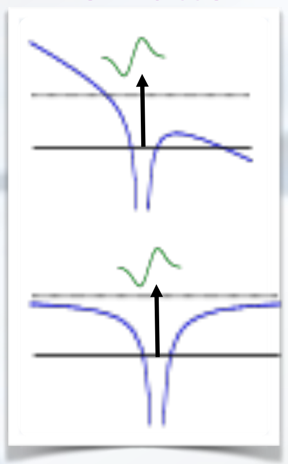
objective: study XUV recollision-induced strong field processes

- ✓ replace the strong field tunnel step with *atto* XUV ionization (step 1)
- ✓ use intense phase-locked MIR to drive propagation (steps 2 & 3)

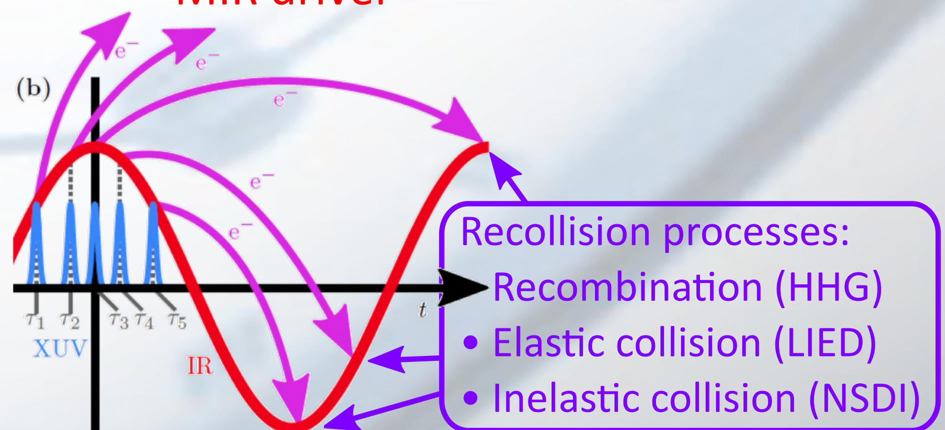
prerequisite: $(q\omega - I_p) \sim 0$, $U_p \rightarrow$ large, $R_{q\omega} >> R_{\omega} \sim 0$

solution: use the λ^2 scaling at longer wavelength

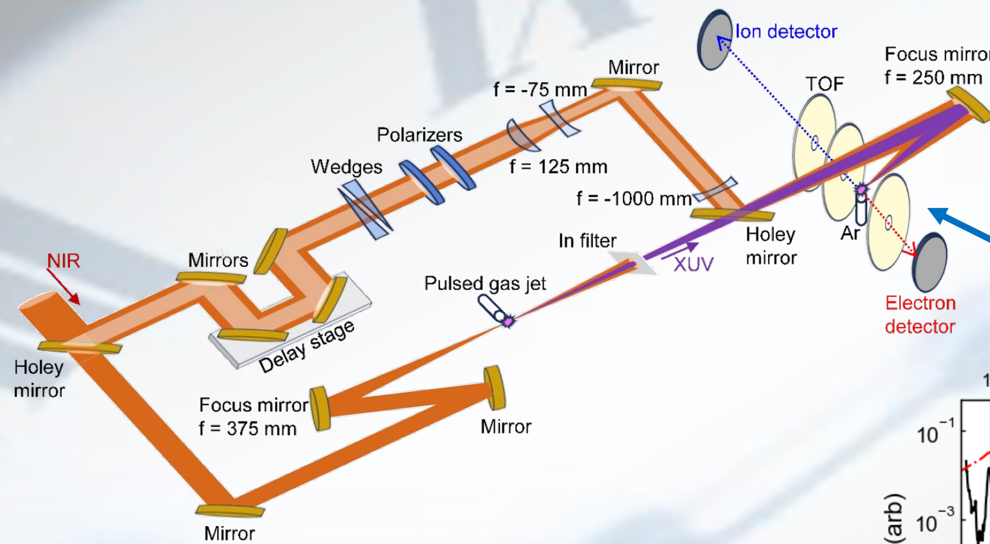
XUV atto

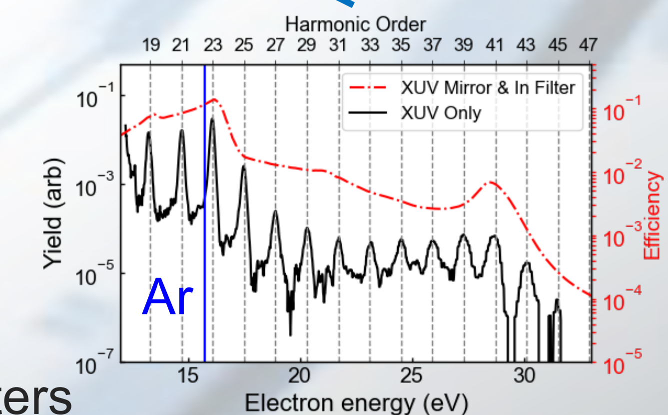


MIR driver



quantum trajectory selector apparatus





features:

1. fundamental field 1.8 – 2.4 μm
2. optimized bright XUV HHG
3. shaped APT spectrum: multi-layer mirrors & metal filters
4. control APT frequency: use both ω & 2ω/4ω schemes
5. temporal metrology uses RABBITT method
6. particle count: TOF e-ion coincidence capability

QTS: recollision (e,2e) double ionization of argon

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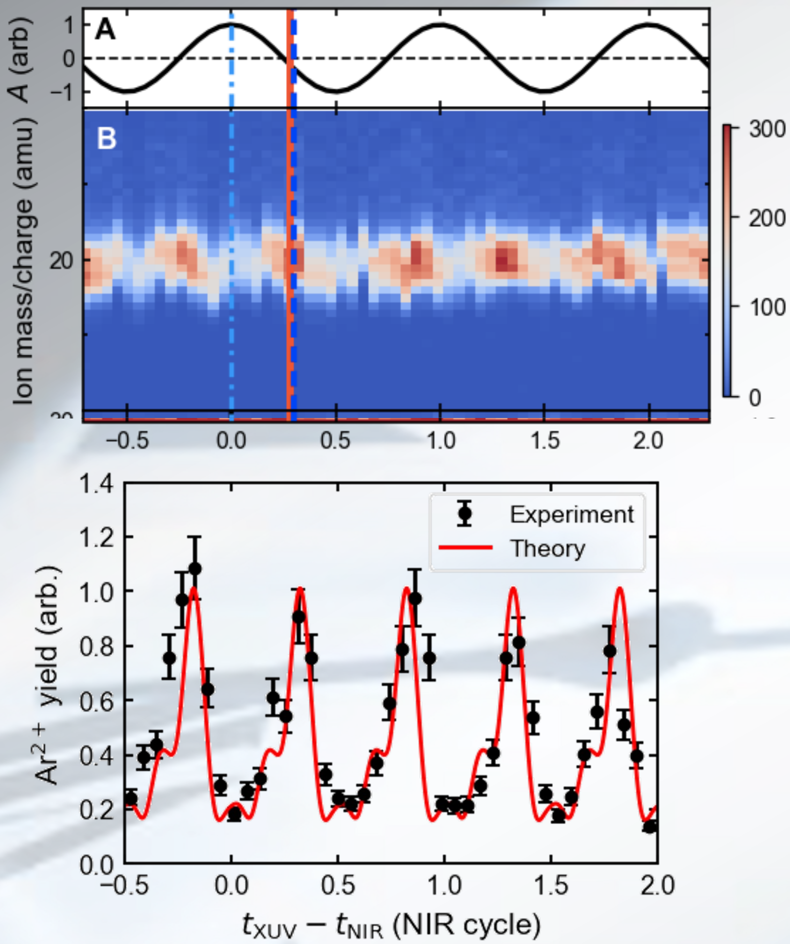
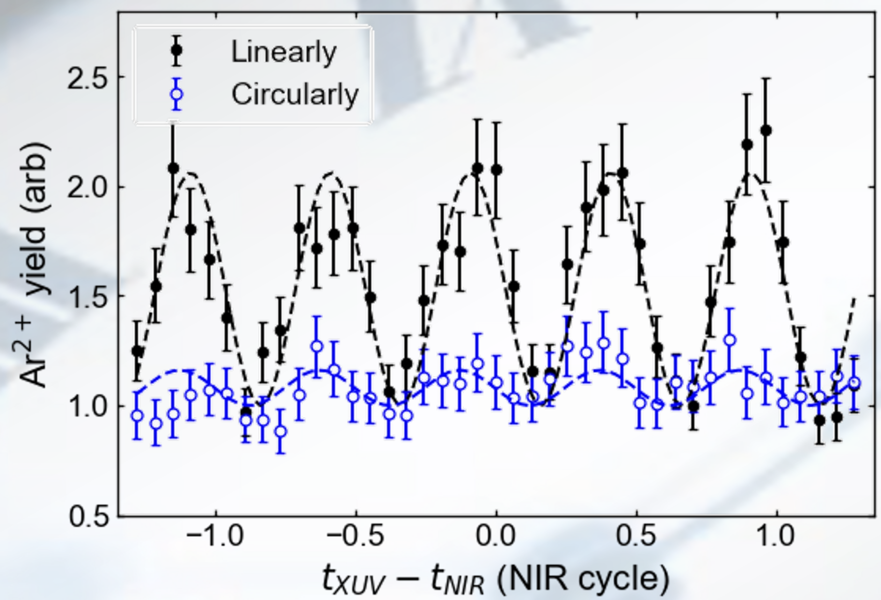


observations

- Ar^{2+} oscillate at 2ω = recollision
- MIR (1.7 μm) alone is < 5% dc
- $3U_p$ = 25 eV < $I_p(Ar^+)$ = 27.6 eV
- model based on recollision impact excitation/direct ionization cross-sections convoluted with RABBITT measurement yields good agreement

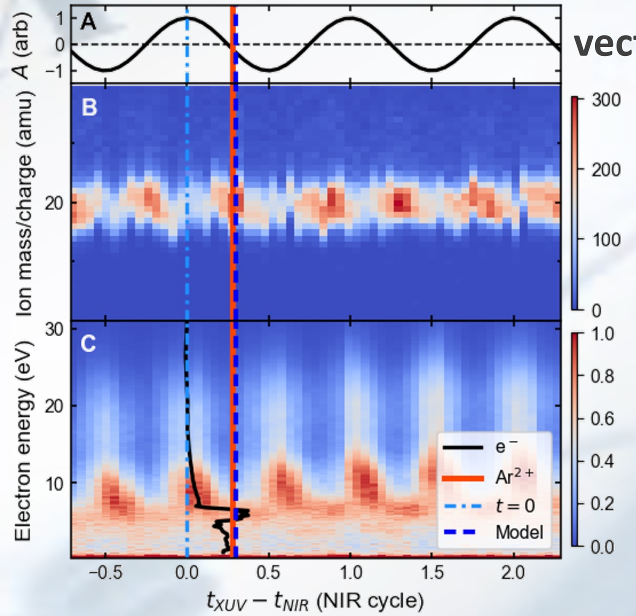
QTS: double ionization for LP & CP fundamental field



observations

- 2ω modulation of the Ar^{2+} yield decreases by factor of 8 for CP
- MIR alone: goes to zero for CP
- the residual 2-color background is independent of polarization

QTS: streak clocking the double ions



vector potential

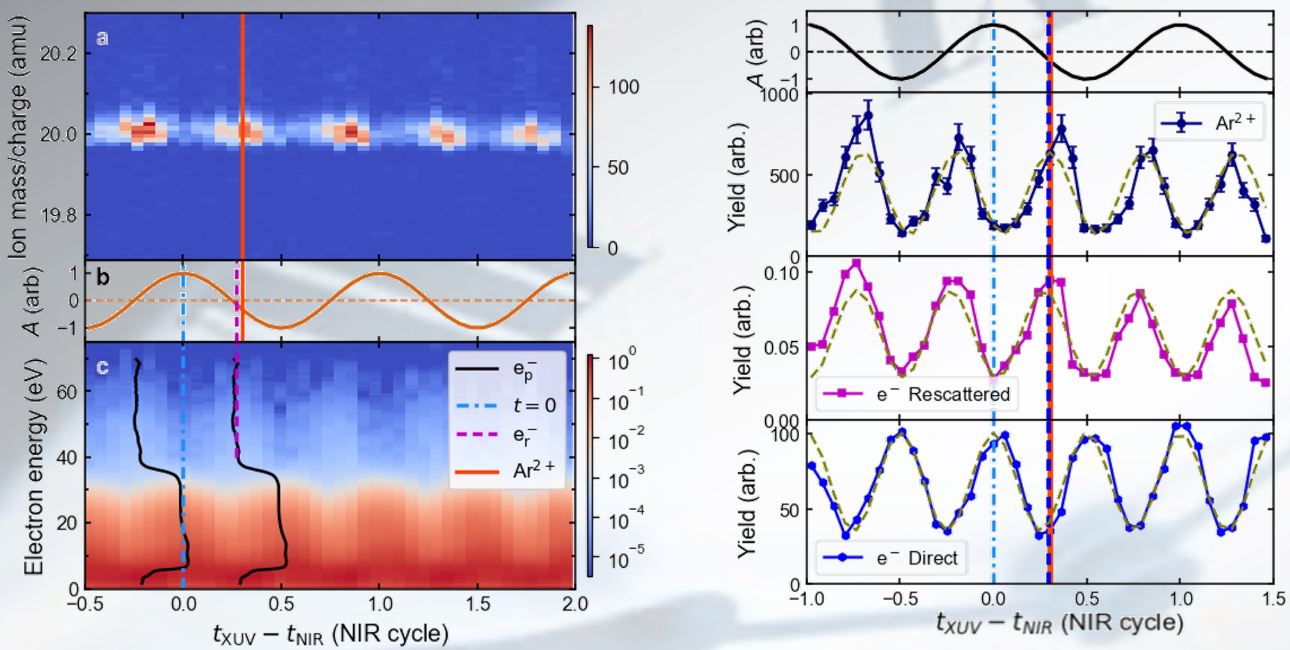
argon double ion delay spectrogram

argon electron delay spectrogram

- electron & double ion delay scans are recorded in parallel
- calibration: maximum streaked energy ($2U_p$) occurs at E-field node
- Ar^{2+} max occurs ~ 0.30 (0.02 [118 as]) MIR cycles after E-field node

3.17U_p classical phase is 0.297 MIR cycle!

QTS: streak clocking the Ar^{2+} & rescattered electrons



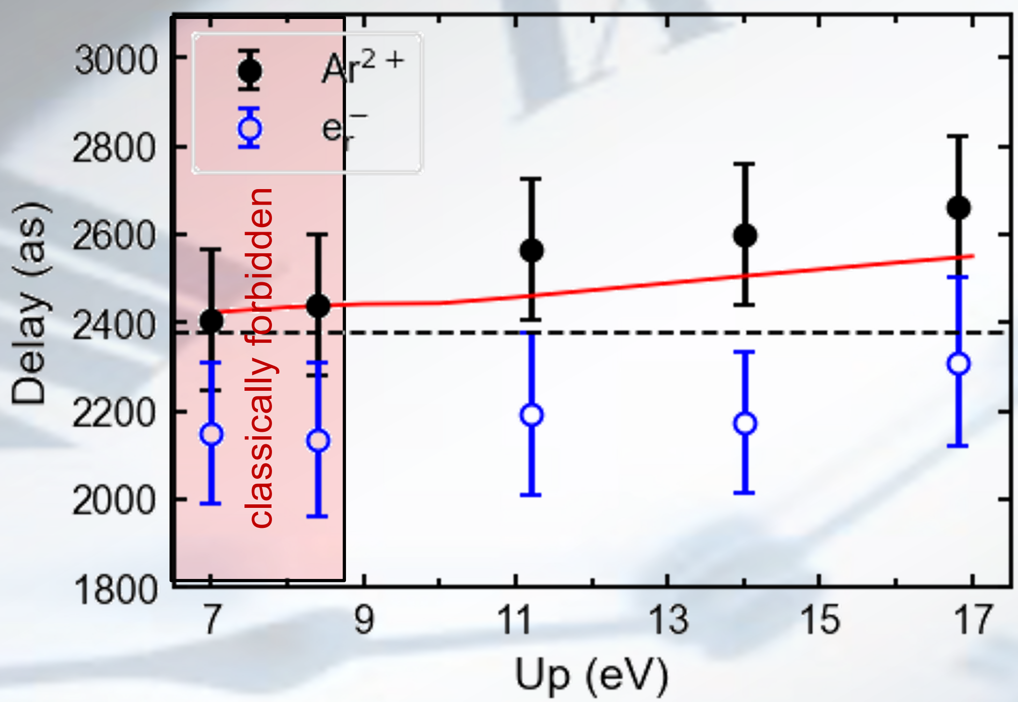
Ar^{2+}: 0.30±0.02 cycle

plateau e-: 0.27±0.02 cycle

clock the rescattered plateau electrons at 0.27±0.02 NIR cycle

classical phase for $10U_p$ electrons & $3.17U_p$: ~0.28 & 0.30 cycles!

QTS: intensity dependence of rescattering process



- (e,2e) & elastic scattering persist above- & below-threshold
- (e,2e) & elastic scattering show different intensity dependence (50-200 as)
- modeling predicts an intensity dependent delay
- model shows that collisional impact excitation & *direct* (*e,2e*) ionization important

QTS: a future paradigm for strong field physics

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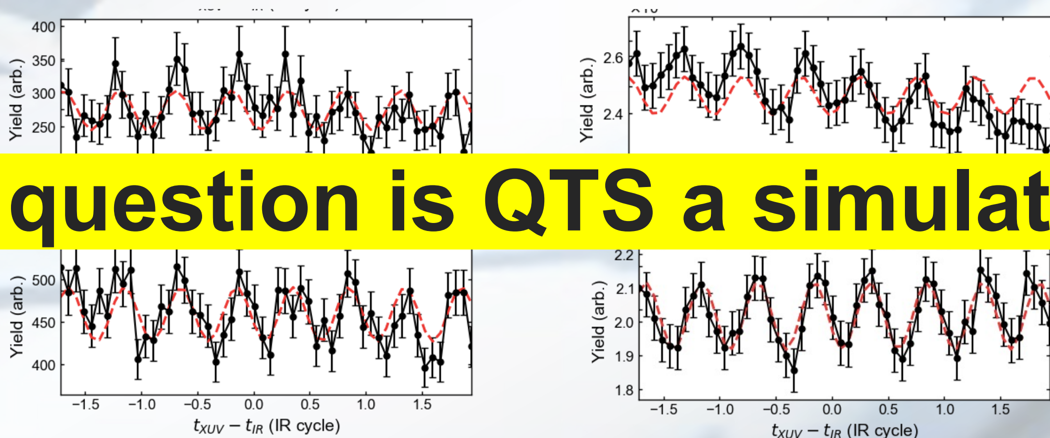
- ✓ robust & generalized control of initial & propagation conditions
- ✓ focus on non-classical behavior
- ✓ SF physics via excitation from core states
- ✓ wave packet holography
- ✓ clocking dynamics and structure in the molecular frame

CO_2^{2+} CO^+



C^+ O^+

big question is QTS a simulator?

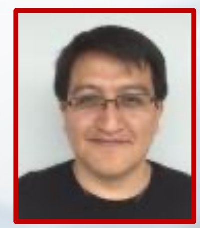
Agostini/DiMauro group

OSU:
- Pierre Agostini
- Andrew Piper
- Joey Liu
- Yaguo Tang
- Dietrich Kiesewetter
- Abraham Camacho-Garibay

LSU:
- Kenneth Schafer
- Jens Bakhoj

experiment

theory



NSF Mid-Scale Research Infrastructure (RI-1 & 2)

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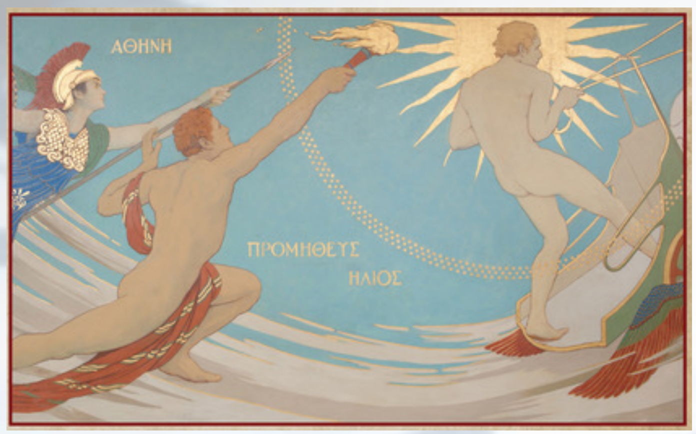
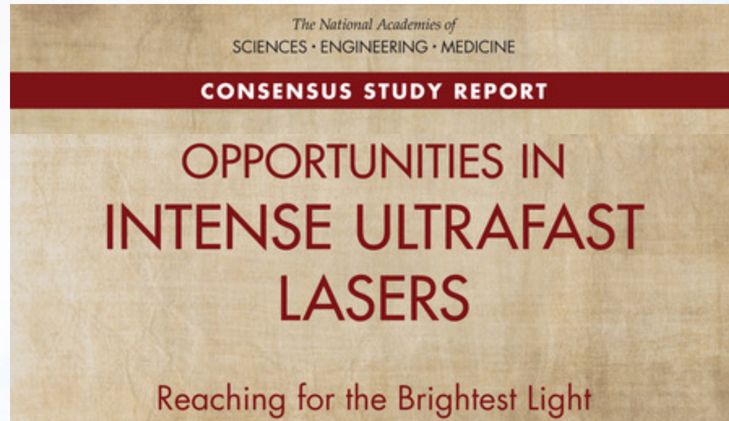
NSF RI program addressed the NAS Recommendations for US Laser Infrastructure





"The United States was the leading innovator and dominant user of high-intensity laser technology when it was developed in the 1990s, but Europe and Asia have now grown to dominate this sector through coordinated national and regional research and infrastructure programs. In Europe, this has stimulated the emergence of the Extreme Light Infrastructure (ELI) program."

Opportunities in Intense Ultrafast Lasers: Reaching for the Brightest Light, NAS, 2018

The NeXUS Facility at Ohio State University

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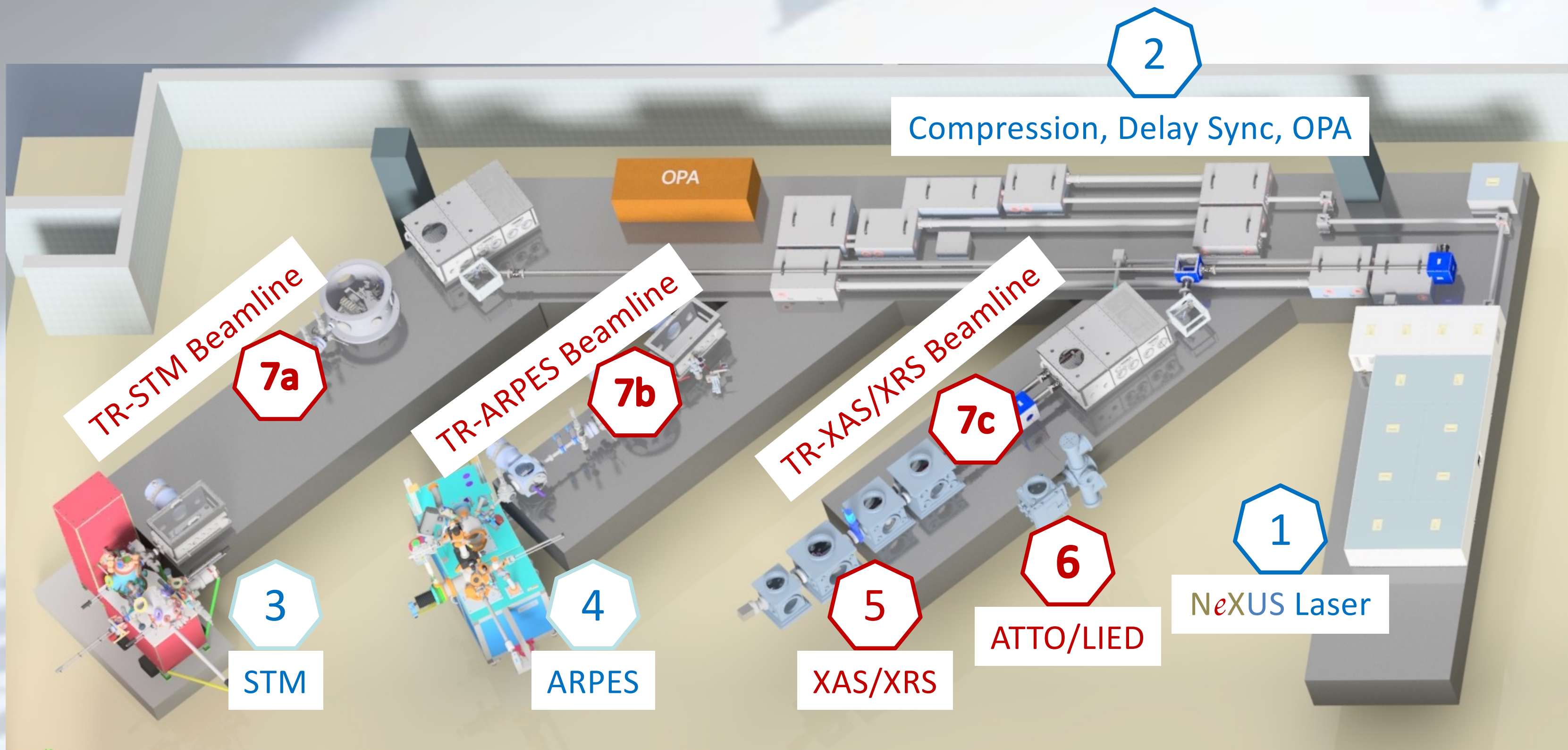
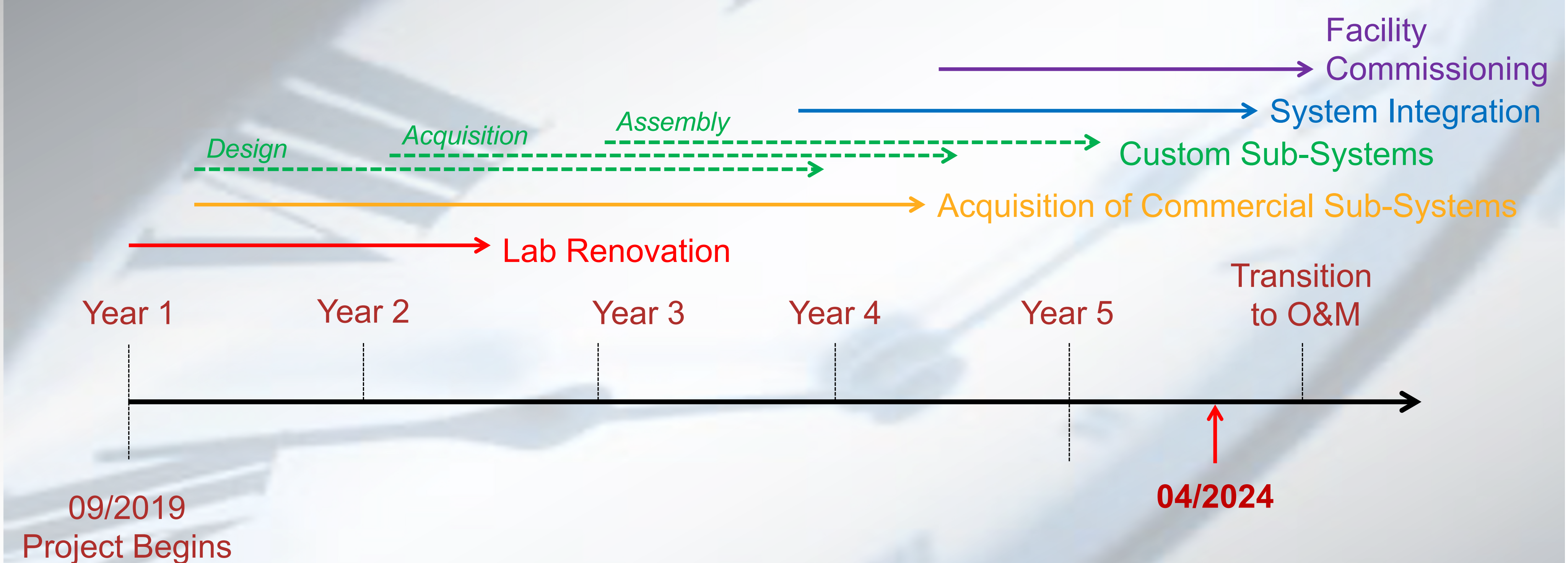
NeXUS Overview

- kW-class Ultrafast Laser:
  8 mJ at 100 kHz or 0.8 mJ at 1 MHz, pulse duration down to 8 fs
- drive attosecond and femtosecond XUV and soft X-ray generation
- supply XUV light to the following experimental end stations
  - ✓ X-ray absorption / X-ray reflection spectroscopy (TR-XAS/XRS)
  - ✓ Angle-resolved photoelectron spectroscopy (TR-ARPES)
  - ✓ Element-specific scanning tunneling microscopy (TR-STM)
  - ✓ Attosecond science / Laser induced electron diffraction (ATTO / LIED)

The NeXUS Facility at Ohio State University



NeXUS project timeline and progress



Questions?