Nobel Prize in Physics for Attosecond Physics

Stockholm U, Dec 8, 2023

Ferenc Krausz (MPQ)
Anne L’Huillier (Lund)
Pierre Agostini (OSU)

OSU red carpet, March 20, 2024

Pierre Agostini
Brutus
Feynman’s lesson on Nature

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
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

1993: rescattering physics
Kulander & Corkum

intense MIR

attosecond burst
$[A(\omega), \phi(\omega, \theta_\mu)]$

the birth of sub-cycle physics

2001: Vienna & Paris attosecond era

Water dissociates in ~10 fs

Bohr period of valence electron
is ~150 as

10^{-18} s

1 as

$10^{-18}$ s

1 as

strong field physics enabled attosecond science?

strong field physics can teach us more about strong field physics!
semi-classical physics: 3-step or rescattering model

- 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)

tunneling + propagation + interaction

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

initial conditions defined by tunnel ionization ($\gamma \ll 1$)
Keldysh picture of strong field tunnel ionization
a sub-cycle view of ionization

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

\( \gamma > 1 \) \quad \text{“photon description”}

\( \gamma < 1 \) \quad \text{“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) $\Rightarrow$ limits the intensity range for measurements
✓ wave packet (WP) born at peak of ac-field $\Rightarrow$ initial phase fixed

the experimentalist has limited control of the initial condition
quantum trajectory selector: attosecond control

goal: use XUV to control the ionization time with attosecond precision and observe recollision
past efforts: control of HHG by APT seeding

"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 Gademann1,6, F Kelkensberg1, W K Siu1, P Johnsson2, M B Gaarde3,4, K J Schafer3,4 and M J J Vrakking1,5


Self-probing spectroscopy of XUV photo-ionization dynamics in atoms subjected to a strong-field environment

Nat. Comm. 8 (2017)

Doron Azoury1, Michael Krüger1, Gal Orenstein1, Henrik R. Larsson2, Sebastian Bauch3, Barry D. Bruner1

& Nirit Dudovich1
deconstructing the strong field processes

quantum trajectory selector (QTS) method

objective: study XUV recollision-induced strong field processes

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

prerequisite: \((q \omega - l_p) \sim 0\), \(U_p \rightarrow \text{large}\), \(R_{q\omega} \gg R_{\omega} \sim 0\)

solution: use the \(\lambda^2\) scaling at longer wavelength

\[\begin{align*}
\text{XUV atto} & \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \季度

Recollision processes:
- Recombination (HHG)
- Elastic collision (LIED)
- Inelastic collision (NSDI)
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 \( \omega \) & \( 2\omega/4\omega \) schemes
5. temporal metrology uses RABBITT method
6. particle count: TOF e-ion coincidence capability
QTS: recollision (e,2e) double ionization of argon

Observations

- Ar$^{2+}$ oscillate at 2\(\omega\) = recollision
- MIR (1.7 µm) alone is < 5% dc
- 3\(U_p\) = 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**

- $2\omega$ 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

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
- \(\text{Ar}^{2+}\) max occurs \(\sim 0.30\) (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

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!

Ar\(^{2+}\): 0.30±0.02 cycle

plateau e-: 0.27±0.02 cycle
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

- 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

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
"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
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

1. NeXUS Laser
2. Compression, Delay Sync, OPA
3. TR-STM Beamline
4. TR-ARPES Beamline
5. TR-XAS/XRS Beamline
6. ATTO/LIED
7a. STM
7b. ARPES
7c. XAS/XRS

The NeXUS Facility is a state-of-the-art research facility at Ohio State University, equipped with various beamlines for different types of measurements including transmission electron microscopy (STM), time-resolved spectroscopy (TR-ARPES), and X-ray absorption spectroscopy (XAS/XRS). The NeXUS Laser system provides the necessary coherent light for these experiments, enabling precise and detailed scientific investigations.
NeXUS project timeline and progress

- **09/2019**: Project Begins
- **Year 1**: Lab Renovation
- **Year 2**: Design
- **Year 3**: Acquisition
- **Year 4**: Assembly
- **Year 5**: System Integration, Facility Commissioning, Custom Sub-Systems, Acquisition of Commercial Sub-Systems, Transition to O&M

**04/2024**: Assembly
Questions?