Attosecond Physics: A New Adventure with Tony in the Ultrafast World

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In celebration of Prof. Anthony Starace’s 70th Birthday
August 22, 2015
Outline

- **Attosecond physics**: the forefront of ultrafast science

- **My journey in the ultrafast world**: probing/controlling ultrafast electron dynamics with attosecond pulses

- **New adventure with Tony**: attosecond control in AMO physics

- **Summary**
What is an attosecond?

1 attosecond = $10^{-18}$ second

1 attosecond to a second is like 1 second to $\sim$32 billion years!
Why do we need attosecond pulses?

Why is the image blurring?

To “take a picture” of the electron motion inside an atom or molecule, one needs attosecond pulses!
ELI-HU site and science park

ELI-Atto

Operational in 2018

Cost: ~300M €

Budapest
165 km (on motorway)

Future
science city

Existing
shopping centre...

... planned extension

Existing
Student hostels

Downtown
5 km

~200M-$ in Asia

MPC-AS

Prof. F. Krausz, MPQ, Germany
Prof. J. Ullrich, MPI:NP, Germany
Prof. J.M. Rost, MPI:CS, Germany
Prof. M. Wolf, FHI, Germany
Prof. A. Cavalleri, CFEI, Germany

~50M-$

UCF

ASSP-GU [Brisbane]
Attosecond science is one of the missions for the proposed super-powerful laser facility at LLE.

* J. D. Zuegel et al., CLEO (2014)
D. D. Meyerhoffer et al., APS-DPP (2014)
The “attosecond-era” has been begun since the two important papers published in 2001.

**Observation of a Train of Attosecond Pulses from High Harmonic Generation**

P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Augé, Ph. Balcou, H. G. Muller, P. Agostini

In principle, the temporal beating of superposed high harmonics obtained by focusing a femtosecond laser pulse in a gas jet can produce a train of very short intensity spikes, depending on the relative phases of the harmonics. We present a method to measure such phases through two-photon, two-color photoionization. We found that the harmonics are locked in phase and form a train of 250-attosecond pulses in the time domain. Harmonic generation may be a promising source for attosecond time-resolved measurements.

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**Attosecond metrology**


The generation of ultrashort pulses is a key to exploring the dynamic behaviour of matter on ever-shorter timescales. Recent developments have pushed the duration of laser pulses close to its natural limit—the wave cycle, which lasts somewhat longer than one femtosecond (1 fs = 10⁻¹⁵ s) in the visible spectral range. Time-resolved measurements with these pulses are able to trace dynamics of molecular structure, but fail to capture electronic processes occurring on an attosecond (1 as = 10⁻¹⁸ s) timescale. Here we trace electronic dynamics with a time resolution of ≈ 150 as by using a subfemtosecond soft-X-ray pulse and a few-cycle visible light pulse. Our measurement indicates an attosecond response of the atomic system, a soft-X-ray pulse duration of 650 ± 150 as and an attosecond synchronism of the soft-X-ray pulse with the light field. The demonstrated experimental tools and techniques open the door to attosecond spectroscopy of bound electrons.
Some neat attosecond experiments in this "new-born" field

Time-resolved atomic inner-shell spectroscopy

The characteristic time constants of the relaxation dynamics of core-excited atoms have hitherto been inferred from the line-widths of electronic transitions measured by continuous-wave extreme ultraviolet or X-ray spectroscopy. Here we demonstrate that a laser-based sampling system, consisting of a few-femtosecond visible light pulse and a synchronized sub-femtosecond soft X-ray pulse, allows us to trace these dynamics directly on the time domain with an attosecond resolution. We have measured a lifetime of 7 fs of M-shell vacancies of krypton in such a pumpprobe experiment.

Lorentz Meets Fano in Spectral Line Shapes: A Universal Phase and Its Laser Control

Symmetric Lorentzian and asymmetric Fano line shapes are fundamental spectroscopic signatures that quantify the structural and dynamical properties of nuclei, atoms, molecules, and solids. This study introduces a universal temporal-phase formulation, manifesting the Fano doubly-excited helium and the same-excited helium with

\[ W_{\text{fin}} \]

[diagram]

\[ W_{\text{bind}} \]

[diagram]

\[ W_{\text{Auger}} \]

[diagram]

\[ W_{\text{Core}} \]

[diagram]

\[ W_{\text{Valley}} \]

[diagram]

\[ W_{\text{Pit}} \]

[diagram]

\[ W_{\text{X-ray}} \]

[diagram]

\[ W_{\text{Probe}} \]

[diagram]
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- My journey in the ultrafast world: probing/controlling ultrafast electron dynamics with attosecond pulses
My “journey” of learning attosecond physics started in 2002 when I was a postdoc in Tony’s group.

Atomic gas-jet

\[ p_f = p_0 + eA(t_b) \]

\[ p_0 = \sqrt{2m(\hbar \omega_A - I_p)} \quad ; \quad E(t) = -\partial A(t)/\partial t \]

By measuring the final electron momentum at different time delays, one can directly “map” out the oscillating field strength of an ultrashort IR-pulse!
The TDSE calculations showed that one can indeed “map-out” the dressing IR-field.

“Great minds think alike”?
The exactly-same experiment was published in SCIENCE just two years later......
Attosecond pump-probe was proposed to explore the ultrafast electron motion in atoms in 2006.

**XUV-probe:**
- \( T = 250 \ \text{as} \)
- \( h\nu = 90 \ \text{eV} \)
- \( I = 3 \times 10^{15} \ \text{W/cm}^2 \)

**VUV-pump:**
- \( T = 1.5 \ \text{fs} \)
- \( h\nu = 23.7 \ \text{eV} \)
- \( I = 6 \times 10^{14} \ \text{W/cm}^2 \)
The ultrafast motion of electron wave-packet can be “mapped” out by time-delayed attosecond pulses.

The two-electron energy sharing also indicates the dynamic evolution of the wave packet.

Attosecond pump-probe proposed to explore the dance of electrons
by Todd Hanson

Electrons in atoms move in a choreographed motion on a time scale of attoseconds (one quadrillionth, or one billionth of a billionth of a second). To observe this ultrafast motion, physicists at Los Alamos have theoretically demonstrated an attosecond pump-probe technique that captures the steps in this intricate dance by exciting the atom at selected times. The development of this technique might someday allow scientists to actually see into a world of electron motion.

In research published recently in Physical Review Letters, Xuming Hu and Lub Fellow Les Collins, both of Atomic and Optical Theory (AOT), describe their work in modeling the dynamics of an attosecond probe, one of the first steps in building such a device, based on existing femtosecond (quadrillionth of a second) devices that use ultrashort laser pulses to capture the motion of atoms in molecules. An attosecond pump-probe would use extreme ultraviolet pulses to capture the motion of electrons in atoms.

According to Hu, the principal investigator for the project, "This generation of extremely short EUV pulses has shown great progress in the last few years. The attosecond pump-probe technique described in our paper could provide a substantial advance in the rapidly developing field of attoscience and could aid physicists, chemists and biologists in investigating and manipulating ultrafast motions of electrons in atoms, molecules, clusters and even nanomachines."

Working much like a strobe light that helps capture stop-action photographs of a falling drop of water, a current generation of femtosecond probes use laser pulses to capture the fast motion of atoms during chemical reactions. Using attosecond pulses of extreme ultraviolet radiation, Hu and Collins believe it may be possible to capture the complete motion of an atom within the incredibly short span of an attosecond.

Consequently, attosecond (APS) uncover...
The proposed pump-probe technique has been cited as a “drive-force” for producing intense attosecond pulses.
Instantaneous AC-Stark shift of excited states in He were probed with attosecond pulses (in collaboration with experimental group at UCF)\*.

140-as XUV

6-fs NIR

Can one control the electron correlation to boost photo-absorption in double-ionization (DI) of He?
The momentum distribution of He-DI manifests the enhanced photo-absorption due to the strong e-e correlation.

\[ E_1[E_2] = 3.2U_p + 12h\nu_{IR} \approx 36 + 18 = 54\text{-eV} \]

\[ E_2[E_1] = h\nu_{EUV} - I_p + 12h\nu_{IR} \approx 41 - 54.4 + 18 = 3.6\text{-eV} \]
The probability of energetic electrons oscillates with the time delay by half-period of the IR field\(^*\)

\[ (E_1 + E_2 = 60 \text{-eV}) \]

Due to excessive IR-photon absorption induced by the strong e-e correlation

“Double-slit” interference patterns can be used to infer the inter-nucleus distance at the instant of the attosecond pulse striking*.

\[ \tau = 250 \text{ asec.} \]

\[ h\nu = 350\text{-eV} \quad h\nu = 630\text{-eV} \]

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Illuminating molecules from within

Marc J. J. Vrakking
FOM Institute for Atomic and Molecular Physics (AMOLF), Science Park 113, Amsterdam, 1098 XG, The Netherlands
Published August 31, 2009

Calculations show that with new short pulse x-ray light sources, it should be possible to use photoelectric emission to make movies of changes in molecular structure.

Subject Areas: Atomic and Molecular Physics, Optics

A Viewpoint on:
Attosecond photoelectron microscopy of $\text{H}_2^+$
S. X. Hu, L. A. Collins and B. I. Schneider

Much of our knowledge about molecular structure and reactivity is based on interpreting how molecules interact with light. In particular, time-resolved pump-probe have been observed previously at synchrotron the photoionization of CO molecules by Le workers [2].

FIG. 1: (Top) In Young’s double-slit experiment light falls onto two slits and is detected on a screen placed behind the slits. An interference pattern is observed, because one cannot determine whether the light passed through the right or left slit. (Bottom) In the experiment proposed by Hu, Collins, and Schneider, x-ray photons eject the single electron from $\text{H}_2^+$. 
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Electron correlation plays an important role in single ionization of He in the vicinity of autoionization resonance*

Enhanced asymmetry in few-cycle attosecond pulse ionization of He in the vicinity of autoionizing resonances

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Asymmetries in production of He⁺(n = 2) with an intense few-cycle attosecond pulse

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By solving the two-electron time-dependent Schrödinger equation, we study carrier-envelope phase (CEP) effects on ionization plus excitation of He to He⁺(n = 2) states by a few-cycle attosecond pulse with a carrier frequency of 51 eV. For most CEPs the asymmetries in the photoelectron angular distributions with excitation of He⁺(2s) or He⁺(2p) have opposite signs and are two orders of magnitude larger than for ionization without excitation. These results indicate that attosecond pulse CEP effects may be significantly amplified in correlated two-electron ionization processes.
Controlling double-ionization of He by an elliptically-polarized attosecond pulse has been demonstrated.

Nonlinear Dichroism in Back-to-Back Double Ionization of He by an Intense Elliptically Polarized Few-Cycle Extreme Ultraviolet Pulse


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Control of double ionization of He by means of the polarization and carrier-envelope phase (CEP) of an intense, few-cycle extreme ultraviolet (XUV) pulse is demonstrated numerically by solving the six-dimensional two-electron, time-dependent Schrödinger equation for He interacting with an elliptically polarized XUV pulse. Guided by perturbation theory (PT), we predict the existence of a nonlinear dichroic effect (α f5/2) that is sensitive to the CEP, ellipticity, peak intensity I, and temporal duration of the pulse. This dichroic effect (i.e., the difference of the two-electron angular distributions for opposite helicities of the ionizing XUV pulse) originates from interference of first- and second-order PT amplitudes, allowing one to probe and control S- and D-wave channels of the two-electron continuum. We show that the back-to-back in-plane geometry with unequal energy sharing is an ideal one for observing this dichroic effect that occurs only for an elliptically polarized, few-cycle attosecond pulse.

FIG. 2 (color online). The TDP $W(\hat{r}, e)$ [Eq. (6)] (in units of $10^{-5}$ a.u.) vs. $\varphi$ [cf. Fig. 1(b)] for DPI of He by a three-cycle XUV pulse (with $\omega = 65$ eV, $I = 2$ PW/cm², $T = 190.9$ as, a cos² envelope, and an ellipticity $\eta = \pm 0.5$ or $\xi = \pm 0.8$) for four CEPs: (a) $\varphi = 0$, (b) $\varphi = \pi/3$, (c) $\varphi = \pi/2$, (d) $\varphi = 5\pi/6$. All results are for the back-to-back geometry and unequal energy sharing (UES); $E_1 = 0.7$ eV and $E_2 = 3.3$ eV. In (c) we give for comparison $W(L=1)(\hat{r}, e)$; see text for discussion.
Matter-wave vortices can be produced in photoionization by delayed circularly-polarized attosecond pulses*


FIG. 2. (color online). Triply differential probability (TDP) \( d^3W/d^3p \) [see Eqs. (2), (4), and (5)] in the polarization plane for He ionization by (a,b,c) right/left and (d) left/right circularly-polarized attosecond pulses. Top row results: time delay \( \tau = 0 \); bottom row results: \( \tau = 500 \) as. In (a), \( \phi_1 = \phi_2 = 0 \); in (b,c,d), \( \phi_1 = 0, \phi_2 = \pi/2 \). In all panels \( \omega = 36 \) eV, \( \pi_p = 3 \) cycles, \( I = 10^{14} \) W/cm\(^2\), and the magnitudes in a.u. of the TDPs are indicated by the color scales.
Summary

- Attosecond physics has been an active field of study for the past decade.

- My “journey” in exploring the ultrafast world started with the inspiration from Tony!

- New adventure with Tony in attosecond AMO physics is bringing fun to us!!
Best wishes from Suxing & Lishan’s Family

Happy 70th Birthday to Tony!
Thank You
“The shortest pulse ever created” to date is ~67-attoseconds generated by gas-HHG using ~7-fs laser at UCF

Tailoring a 67 attosecond pulse through advantageous phase-mismatch

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A single isolated attosecond pulse of 67 as was composed from an extreme UV supercontinuum covering 55–130 eV generated by the double optical gating technique. Phase mismatch was used to exclude the single-atom cutoff of the spectrum that possesses unfavorable attochirp, allowing the positive attochirp of the remaining spectrum to be compensated by the negative dispersion of a zirconium foil. Two algorithms, PROOF and FROG-CRAB, were employed to retrieve the pulse from the experimental spectrum, yielding nearly identical results. © 2012 Optical Society of America
The classical double-slit picture becomes valid in APM only when $n\lambda_e < 0.65 \times R_0$ [independent slits: $R_0\sin(\phi_n) = n\lambda_e$]

Comparison of the double-slit interference angles of the “classical” double-slit (DS) formula and our TDSE calculations as the photon energy ($\hbar\omega$) varies

<table>
<thead>
<tr>
<th>$\hbar\omega$ (eV)</th>
<th>$\lambda_e$ (Bohr)</th>
<th>Classical DS angles ($\phi_n$)(°)</th>
<th>TDSE angles ($\phi_n$)(°)</th>
</tr>
</thead>
<tbody>
<tr>
<td>170</td>
<td>1.9587</td>
<td>$\phi_1 \approx 75.3^\circ$</td>
<td>$\phi_1 \approx 52.0^\circ$</td>
</tr>
<tr>
<td>210</td>
<td>1.7274</td>
<td>$\phi_1 \approx 58.5^\circ$</td>
<td>$\phi_1 \approx 49.5^\circ$</td>
</tr>
<tr>
<td>250</td>
<td>1.5258</td>
<td>$\phi_1 \approx 50.5^\circ$</td>
<td>$\phi_1 \approx 46.5^\circ$</td>
</tr>
<tr>
<td>300</td>
<td>1.4104</td>
<td>$\phi_1 \approx 44.1^\circ$</td>
<td>$\phi_1 \approx 42.3^\circ$</td>
</tr>
<tr>
<td>350</td>
<td>1.2955</td>
<td>$\phi_1 \approx 39.7^\circ$</td>
<td>$\phi_1 \approx 39.9^\circ$</td>
</tr>
<tr>
<td>430</td>
<td>1.1588</td>
<td>$\phi_1 \approx 34.9^\circ$</td>
<td>$\phi_1 \approx 34.8^\circ$</td>
</tr>
<tr>
<td>630</td>
<td>0.9462</td>
<td>$\phi_1 \approx 28.0^\circ$</td>
<td>$\phi_1 \approx 28.4^\circ$</td>
</tr>
</tbody>
</table>

By measuring the diffraction angles, one can infer the transient structure of molecules.