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RPPR Final Report

as of 28-Sep-2021

Agency Code: 21XD

Proposal Number: 65314CHMUR

Agreement Number: W911NF-14-1-0383

INVESTIGATOR(S):

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DUNS Number: 124726725

EIN: 946002123

Report Date: 20-Jul-2021

Date Received: 27-Sep-2021

Final Report for Period Beginning 21-Jul-2014 and Ending 20-Apr-2021

Title: Post-Born-Oppenheimer Dynamics Using Isolated Attosecond Pulses

Begin Performance Period: 21-Jul-2014

End Performance Period: 20-Apr-2021

Report Term: 0-Other

Submitted By: Stephen Leone

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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 3

STEM Participants: 21

Major Goals: One of the most exciting developments in science during the last 20 years has been the invention of new techniques (high harmonic generation) to produce attosecond light pulses in the vacuum ultraviolet and soft x-ray regime (now providing useable pulses up to 300-500 eV in the X-ray region of the spectrum and as short as 50-100 attoseconds). Attosecond electron dynamics became a new field of investigation that goes beyond the typical femtosecond timescales of nuclear vibrational, rotational, or bond breaking motions in molecules. The investigation of such ultrashort attosecond timescales focuses on electron dynamics as the principal target. Since electrons are the primary driving force in all of chemistry, attosecond pulses offered the potential of a new frontier in molecular and chemical physics, in which one could directly track and manipulate electron dynamics. This MURI program was designed to address that frontier.

The projects involved both new pulse developments and fundamental science investigations in molecular dynamics. The program also balanced experimental research with theoretical developments and collaboration. The specific projects outlined in this MURI yielded new insights into the electron dynamics of novel energy transfer and charge transport mechanisms that are of broad interest to ARO, as well as technical and theoretical developments in lasers and chemical dynamics interpretations. In some cases, very short, subfemtosecond timescales were achieved, made possible by the subcycle nature of the attosecond pulse formation, as well as the locking of the carrier wave phase to the envelope of the driver pulses.

Experiments and theory addressed passage through conical intersections, electronic charge migration, electronic superpositions and vibrational superpositions, light-induced conical intersections, photoionization timescales and angular distributions. Many of the projects addressed chemical dynamics in regimes where the breakdown of the Born-Oppenheimer approximation was meaningful, achieving results where the electronic timescales were probed before, during, and after the critical nuclear motions relevant to the chemical dynamics.

Accomplishments: Overview details of the accomplishments are described in the attached file, and a brief summary of the most important findings are mentioned here. Refer to the annual reports for additional details during each annual project period.

This MURI program produced numerous results for attosecond pulse generation methods and a multitude of gas phase atomic and molecular measurements for the first time, each with enough time resolution to distinguish dynamical processes that were previously unobtainable. In some cases the successes were mainly experimental, in some primarily theoretical, and in many cases the work was combined experimental and theoretical.

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These include: third order dispersion compensation and optical gating methods to robustly produce attosecond pulses, passage through curve crossings and conical intersections in diatomic and polyatomic molecules, the formation of electronic coherences in atoms and molecules, the interplay between electronic and vibrational coherences in molecules, coherent electron dynamics triggered by core and inner valence ionization in molecules, state-resolved few-hundred attosecond delays in strong field ionization, formation of light-induced conical intersections.

Results showed the preservation of electronic coherent dynamics in the face of intramolecular vibrational relaxation that destroys the coherence, use of four wave mixing to analyze autoionizing dynamics, photoelectron angular distributions in inner shell ionization, development of attosecond X-ray pulses with longer wavelength driver pulses, application of soft X-rays to strong field ionization of nitrogen containing molecules, and gain mechanisms in air ionization. World record 53 attosecond pulses were validated.

Liquid phase photoelectron measurements were developed, and solid-state measurements of electron and hole carrier dynamics, as well as coherent phonon motions were accessed for the first time.

Training Opportunities: Seven research groups were involved in this MURI effort. In each laboratory and theoretical effort, graduate students, postdoctoral associates, visiting students, and related students are educated by both the principal investigators and the more advanced students and postdoctorals associated with the project. Training takes place on lasers, sophisticated principles of carrier envelope phase stabilization of short pulse lasers, pulse compression, the production of isolated attosecond pulses, and electron/ion and transient absorption detection. Theoretically students learn advanced computational codes for molecular dynamics and electron processes. Students and postdocs attend conferences and learn to disseminate their work orally and in written formats.

Students were given the opportunity to attend conferences and to present their work, in oral presentations and with posters. These include American Physical Society, American Chemical Society, Laser Science Conference, and a recollisions workshop related to high harmonic generation. Students were given the opportunity to design equipment and construct them in the shops and to develop codes for theory. Students interacted with mentors one on one and in group situations, such as group seminars and subgroup meetings, where they learned communication skills and techniques of asking and answering questions about research. Students developed individual study techniques to further their scientific knowledge. Students practiced and passed their oral qualifying exams with professors, and filed theses with faculty committees. There was additional training of graduate students and postdocs in petascale parallel computing (including workshops at the National Energy Research Scientific Computing Center).

Results Dissemination: Many publications were submitted and accepted; the detailed list of products enumerates over 100 publications. Some of the publications were reviews, which provided a perspective on the new field. A multitude of invited talks and contributed talks were given by the faculty members and students and postdocs involved in the MURI program. Those talks in many cases were high profile invited presentations at major conferences. One of the MURI participants will host the next major international conference on attosecond science. Outreach involved bringing high school teachers and high school students for laboratory visits and extended visits. Teaching modules were developed by those teachers regarding light-induced processes, short pulse lasers, and optical phenomena, which were provided to high school students after the visits. Laser companies were engaged about the needs for driver laser properties for the attosecond pulse production. One company started to provide carrier envelope phase stabilized lasers during the course of the MURI program, greatly benefitting the field.

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Honors and Awards: Corkum Schneider Medal, National Research Council Canada, 2017
Corkum Royal Medal, Royal Society, 2017
Chang UCF Luminary Award, 2017.
Corkum Awards 2018-2019:
Corkum 2019: Willis E. Lamb Award for Laser Science and Quantum Optics
Corkum 2018: The IOP's Isaac Newton Medal and Prize, the Society's highest award
Corkum 2018: SPIE Gold Medal, the Society's highest award
Corkum Honorary degrees 2018-2019: 2019: University of Rostock, 2018: Aix-Marseille University, France
Cederbaum: Honorary Doctorate from University of Debrecen, Hungary, 2016.
Chang UCF Trustee Chair Professor, 2016, 2021.
Chang UCF Pegasus Professor Award, 2016.
Corkum M.V. Lomonosov Gold Medal, Russian Academy of Sciences, 2015 (award shared with L. V. Keldysh)
Corkum Thomson Reuters Citation Laureate, 2015; (Citation shared with F. Krausz)
Corkum Honorary PhD from Université de Sherbrooke, 2015
Corkum Honorary PhD from the Université Laval, 2015
Corkum Honorary PhD from Aarhus University, 2016
Leone Honorary Doctorate Degree, 2016 University of Warwick, Warwick UK
Leone Ahmed Zewail Award in Ultrafast Science, American Chemical Society, 2016
Neumark Peter Debye Award in Physical Chemistry, American Chemical Society, 2019
Neumark Bourke Medal, Royal Society of Chemistry UK, 2015
Chang UCF University Excellence in Research Award, 2015.
Chang UCF Research Incentive Awards, 2015
Cederbaum: Elected Member of the Leopoldina National Academy of Sciences, 2015
Corkum Ives Award from the OSA – the highest award of the Optical Society (October 2014).
Corkum Appointed Distinguished University Professor at the University of Ottawa.
Sandhu UA Distinguished Scholar Award, Office of the Provost, University of Arizona, 2017
Cederbaum Honorary Doctorate from University of Debrecen, Hungary, 2016

Ongoing honors:
Neumark National Academy of Sciences
Leone National Academy of Sciences
Neumark American Academy of Arts and Sciences
Leone American Academy of Arts and Sciences
Corkum Foreign member of the Russian Academy of Sciences
Corkum Foreign member of the Austrian Academy of Sciences
Corkum Foreign member of the U.S. National Academy of Sciences
Corkum Fellow of the Royal Society of London
Corkum Fellow of the Royal Society of Canada

Protocol Activity Status:

Technology Transfer: There were extensive discussions with laser manufacturers, and one company, Coherent, decided to market a successful carrier envelope phase stabilized laser system. Short pulse methods developed in the MURI include the method called PASSAGE and the compensation for higher order dispersion by the use of KDP crystals.

PARTICIPANTS:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Dong Hyuk Ko

Person Months Worked: 12.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

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Participant: Marko Haertelt

Person Months Worked: 12.00

Project Contribution:

National Academy Member: N

Funding Support:

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Person Months Worked: 12.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

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Person Months Worked: 12.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Mathew Britton

Person Months Worked: 6.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Patrick Laferriere

Person Months Worked: 6.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Martin Chiasson

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Aleksey Korobenko

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: T.J. Hammond

Person Months Worked: 2.00

Funding Support:

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as of 28-Sep-2021

Project Contribution:
National Academy Member: N

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National Academy Member: N

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Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
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Project Contribution:
National Academy Member: N

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Project Contribution:
National Academy Member: N

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Project Contribution:
National Academy Member: N

Participant Type: Co PD/PI
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Project Contribution:
National Academy Member: Y

RPPR Final Report
as of 28-Sep-2021

Participant Type: Co PD/PI
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Project Contribution:
National Academy Member: N
Funding Support:

Participant Type: Graduate Student (research assistant)
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Project Contribution:
National Academy Member: N
Funding Support:

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Project Contribution:
National Academy Member: N
Funding Support:

Participant Type: Graduate Student (research assistant)
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Project Contribution:
National Academy Member: N
Funding Support:

Participant Type: Graduate Student (research assistant)
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Project Contribution:
National Academy Member: N
Funding Support:

Participant Type: Graduate Student (research assistant)
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Project Contribution:
National Academy Member: N
Funding Support:

Participant Type: Graduate Student (research assistant)
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Project Contribution:
National Academy Member: N
Funding Support:

RPPR Final Report
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Participant Type: PD/PI

Participant: Stephen Leone

Person Months Worked: 1.00

Project Contribution:

National Academy Member: Y

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

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Project Contribution:

National Academy Member: N

Funding Support:

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Participant: Simona Scheid

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Jacquelin Fedyk

Person Months Worked: 12.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Co PD/PI

Participant: Lorenz Cederbaum

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Co PD/PI

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Person Months Worked: 1.00

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National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

RPPR Final Report
as of 28-Sep-2021

Participant: Loren Greenman

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Zachary Streeter

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Roger Bello Romero

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Fangjie Zhou

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Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Hanfu Kong

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Project Contribution:

National Academy Member: N

Funding Support:

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Funding Support:

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Project Contribution:
National Academy Member: N

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Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Xiaoming Ren

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Funding Support:

Project Contribution:

National Academy Member: N

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Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Co PD/PI

Participant: Zenghu Chang

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Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Co PD/PI

Participant: Daniel Neumark

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Funding Support:

Project Contribution:

National Academy Member: Y

Participant Type: Faculty

Participant: Ágnes Vibók

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Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Faculty

Participant: Ágnes Vibók

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Funding Support:

Project Contribution:

National Academy Member: N

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as of 28-Sep-2021

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National Academy Member: N

Funding Support:

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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)
Participant: Yuki Kobayashi
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Faculty
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
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Project Contribution:
National Academy Member: N

Funding Support:

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Participant: Xiaoming Ren
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
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Project Contribution:
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Funding Support:

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Funding Support:

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Participant: Michelle Haskins
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Other Professional
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Project Contribution:
National Academy Member: N

Funding Support:

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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

RPPR Final Report
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Project Contribution:
National Academy Member: N

Funding Support:

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Project Contribution:
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Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Tymur Parpiiev
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)
Participant: Bethany deRoulet
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Other (specify)
Participant: Johannes Weiss
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Undergraduate Student
Participant: Youn Jue (Eunice) Eunice
Person Months Worked: 3.00
Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Man-Nung Su
Person Months Worked: 6.00

Funding Support:

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Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student

Participant: Andrew Chew

Person Months Worked: 3.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Research Experience for Undergraduates (REU) Participant

Participant: Jaden Hochstetter

Person Months Worked: 1.00

Project Contribution:

National Academy Member: Y

Funding Support:

International Travel:

HUN 3 days

International Collaboration:

HUN

JPN

ARTICLES:

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Publication Identifier Type: Other

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Volume: 346

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First Page #: 1348

Date Submitted: 7/27/16 12:00AM

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Publication Location:

Article Title: Attosecond band-gap dynamics in silicon

Authors: Martin Schultze, Krupa Ramasesha, C.D. Pemmaraju, S.A. Sato, D. Whitmore, A. Gandman, James S. |

Keywords: silicon, band gap, attosecond, phonon, tunneling

Abstract: Electron transfer from valence to conduction band states in semiconductors is the basis of modern electronics. Here, attosecond extreme ultraviolet (XUV) spectroscopy is used to resolve this process in silicon in real time. Electrons injected into the conduction band by few-cycle laser pulses alter the silicon XUV absorption spectrum in sharp steps synchronized with the laser electric field oscillations. The observed ~450-attosecond step rise time provides an upper limit for the carrier-induced band-gap reduction and the electron-electron scattering time in the conduction band. This electronic response is separated from the subsequent band-gap modifications due to lattice motion, which occurs on a time scale of 60 ± 10 femtoseconds, characteristic of the fastest optical phonon. Quantum dynamical simulations interpret the carrier injection step as light-field-induced electron tunneling.

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Publication Identifier Type: DOI **Publication Identifier:** 10.1146/annurev-physchem-040215-112025
Volume: 67 **Issue:** 0 **First Page #:** 41
Date Submitted: 2/3/17 12:00AM **Date Published:** 2/12/16 4:00PM
Publication Location:
Article Title: Real-time probing of electron dynamics using attosecond time-resolved spectroscopy
Authors: Krupa Ramasesha, Stephen R. Leone, and Daniel M. Neumark
Keywords: ultrafast spectroscopy, electron dynamics, high harmonic generation, transient absorption, photoelectron spectroscopy
Abstract: Attosecond science has paved the way for direct probing of electron dynamics in gases and solids. This review provides an overview of recent attosecond measurements, focusing on the wealth of knowledge obtained by the application of isolated attosecond pulses in studying dynamics in gases and solid-state systems. Attosecond photoelectron and photoion measurements in atoms reveal strong-field tunneling ionization and a delay in the photoemission from different electronic states. These measurements applied to molecules shed light on ultrafast intramolecular charge migration. Similar approaches are used to understand photoemission processes from core and delocalized electronic states in metal surfaces. Attosecond transient absorption spectroscopy is used to follow the real-time motion of valence electrons and to measure the lifetimes of autoionizing channels in atoms. In solids, it provides the first measurements of bulk electron dynamics, revealing important phenomena such as the timesca
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Volume: 91 **Issue:** 5 **First Page #:** 51401
Date Submitted: 7/27/16 12:00AM **Date Published:**
Publication Location:
Article Title: Control of charge migration in molecules by ultrashort laser pulses
Authors: Nikolay V. Golubev, and Alexander I. Kuleff
Keywords: charge migration, laser control, hole, ultrashort pulses
Abstract: Due to electronic many-body effects, the ionization of a molecule can trigger ultrafast electron dynamics appearing as a migration of the created hole charge throughout the system. Here we propose a scheme to control the charge-migration dynamics with a single ultrashort laser pulse. We demonstrate by fully ab initio calculations on a molecule containing a chromophore and amine moieties that simple pulses can be used to stop the charge-migration oscillations and localize the charge on the desired site of the system. We argue that this control may be used to predetermine the follow-up nuclear rearrangement and thus the molecular reactivity.
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Publication Identifier: 10.1021/jz502468d

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Article Title: Direct Signature of Light-Induced Conical Intersections in Diatomics

Authors: G. J. Hala^{sz}, A[?]. Vibo^k, and L. S. Cederbaum

Keywords: conical intersection, nonadiabatic effects, lasers, deuterium ion

Abstract: Nonadiabatic effects are ubiquitous in physics, chemistry, and biology. They are strongly amplified by conical intersections (CIs), which are degeneracies between electronic states of triatomic or larger molecules. A few years ago it was revealed that CIs in molecular systems can be formed by laser light, even in diatomics. Because of the prevailing strong nonadiabatic couplings, the existence of such laser-induced conical intersections (LICIs) may considerably change the dynamical behavior of molecular systems. By analyzing the photodissociation process of the D₂⁺ molecule carefully, we found a robust effect in the angular distribution of the photofragments that serves as a direct signature of the LICI, providing undoubted evidence of its existence.

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Publication Identifier: 10.1063/1.4923441

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Date Submitted: 7/27/16 12:00AM

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Publication Location:

Article Title: Photodissociation of D₂⁺ induced by linearly chirped laser pulses

Authors: András Csehi, Gábor J. Halász, Lorenz S. Cederbaum, and Ágnes Vibók

Keywords: conical intersection, light induced conical intersection, deuterium molecule, laser, photodissociation, chirped pulse

Abstract: Recently, it has been revealed that so-called light-induced conical intersections (LICIs) can be formed both by standing or by running laser waves even in diatomic molecules. Due to the strong nonadiabatic couplings, the existence of such LICIs has significant impact on the dynamical properties of a molecular system. In our former studies, the photodissociation process of the D+2 molecule was studied initiating the nuclear dynamics both from individual vibrational levels and from the superposition of all the vibrational states produced by ionizing D₂. In the present work, linearly chirped laser pulses were used for initiating the dissociation dynamics of D+2. In contrast to the constant frequency (transform limited) laser fields, the chirped pulses give rise to LICIs with a varying position according to the temporal frequency change. To demonstrate the impact of these LICIs on the dynamical properties of diatomics, the kinetic energy release spectra, the total dissociation probability

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Publication Location:

Article Title: Attosecond transient absorption in dense gases: Exploring the interplay between resonant pulse propagation and laser-induced line-shape control

Authors: Chen-Ting Liao and Arvinder Sandhu

Keywords: Attosecond, helium, lineshape, laser, polarization, phase shift

Abstract: We investigate the evolution of extreme ultraviolet (XUV) spectral line shapes in an optically thick helium gas under near-infrared (IR) perturbation. In our experimental and theoretical work, we systematically vary the IR intensity, time-delay, gas density, and IR polarization parameters to study line-shape modifications induced by collective interactions in a regime beyond the single-atom response of a thin, dilute gas. In both experiment and theory, we find that specific features in the frequency-domain absorption profile, and their evolution with propagation distance, can be attributed to the interplay between resonant attosecond pulse propagation and IR-induced phase shifts. Our calculations show that this interplay also manifests itself in the time domain, with the IR pulse influencing the reshaping of the XUV pulse propagating in the resonant medium.

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Date Submitted: 7/28/16 12:00AM

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Publication Location:

Article Title: Attosecond optics and technology: progress to date and future prospects

Authors: Z. Chang, P. B. Corkum, and S. R. Leone

Keywords: Ultrafast optics; Strong field laser physics; Multiphoton processes

Abstract: The milestones of attosecond optics research in the last 15 years are briefly reviewed, and the latest trends in applications in gaseous and condensed matter are introduced. An outlook on future development of attosecond soft x-ray sources and their application is provided.

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Publication Location:

Article Title: Polarization-assisted amplitude gating as a route to tunable, high-contrast attosecond pulses

Authors: H. Timmers, M. Sabbar, J. Hellwagner, D. M. Neumark, and S. R. Leone

Keywords: Ultrafast spectroscopy; Ultrafast nonlinear optics; X-rays, soft x-rays, extreme ultraviolet (EUV)

Abstract: Attosecond spectroscopy is a powerful technique for probing electron dynamics in fundamental systems. However, extending this method to cover a wide range of element-specific, core-hole transitions requires the availability of broadly tunable attosecond pulses. In this Letter, we present a new method for generating high-flux, high-contrast single attosecond pulses tunable across the range of 50–120 eV. The method is referred to as a Polarization ASSisted Amplitude GatE (PASSAGE) and uses a few-cycle driving pulse along with a partial polarization gate to extend the bandwidth of high harmonic emission in the temporally isolated, cut-off portion of the spectrum. The simplicity of this technique will help pave the way for implementing attosecond core-hole spectroscopy to probe more complex reactions and bring attosecond science into a multidisciplinary setting.

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Publication Location:

Article Title: Extreme ultraviolet transient absorption of solids from femtosecond to attosecond timescales

Authors: Lauren J. Borja, M. Zürich, C. D. Pemmaraju, Martin Schultze, Krupa Ramasesha, Andrey Gandman, Ja

Keywords: Multiharmonic generation; Spectroscopy, semiconductors; Ultrafast processes in condensed matter, including semiconductors

Abstract: High-harmonic generation (HHG) produces ultrashort pulses of extreme ultraviolet radiation (XUV), which can be used for pump-probe transient absorption spectroscopy in metal oxides, semiconductors, and dielectrics. Femtosecond transient absorption on iron and cobalt oxides identifies ligand-to-metal charge transfer as the main spectroscopic transition, rather than metal-to-metal charge transfer or d-d transitions, upon photoexcitation in the visible. In silicon, attosecond transient absorption reveals that electrons tunnel into the conduction band from the valence band under strong-field excitation, to energies as high as 6 eV above the conduction band minimum. Extensions of these experiments to other semiconductors, such as germanium, and other transition metal oxides, such as vanadium dioxide, are discussed. Germanium is of particular interest because it should be possible to follow both electron and hole dynamics in a single measurement using transient XUV absorption.

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Publication Location:

Article Title: Photodissociation of D₂ + induced by linearly chirped laser pulses

Authors: András Csehi, Gábor J. Halász, Lorenz S. Cederbaum, and Ágnes Vibók

Keywords: photodissociation dynamics, chirped laser pulses,

Abstract: Recently, it has been revealed that so-called light-induced conical intersections (LICIs) can be formed both by standing or by running laser waves even in diatomic molecules. Due to the strong nonadiabatic couplings, the existence of such LICIs has significant impact on the dynamical properties of a molecular system. In our former studies, the photodissociation process of the D₂ molecule was studied initiating the nuclear dynamics both from individual vibrational levels and from the superposition of all the vibrational states produced by ionizing D₂. In the present work, linearly chirped laser pulses were used for initiating the dissociation dynamics of D₂. In contrast to the constant frequency (transform limited) laser fields, the chirped pulses give rise to LICIs with a varying position according to the temporal frequency change.

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Date Submitted: 7/28/16 12:00AM

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Publication Location:

Article Title: Quantum oscillations between close-lying states mediated by the electronic continuum in intense high-frequency pulses

Authors: Philipp V. Demekhin, and Lorenz S. Cederbaum

Keywords: attosecond, ionization, Raman, theory

Abstract: The dynamics of neighboring states exposed to short intense laser pulses of carrier frequencies well above the ionization threshold of the system is investigated theoretically in the dipole approximation. It is shown that the ionizing pulse induces a time-dependent non-Hermitian coupling between these states determined by the Raman coupling and the direct ionization probability. This coupling induces quantum oscillations between the neighboring states while the strong pulse is on. The phenomenon opens the possibility to achieve a coherent control over the populations of neighboring states by short intense ionizing pulses. The present numerical results suggest exciting applications for free-electron lasers and attosecond experiments.

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Journal: Phys. Rev. A

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Publication Location:

Article Title: Dynamic interference in the photoionization of He by coherent intense high-frequency laser pulses: Direct propagation of the two-electron wave packets on large spatial grids

Authors: A. N. Artemyev, A. D. Müller, D. Hochstuhl, L. S. Cederbaum, and Ph. V. Demekhin

Keywords: attosecond, ionization, helium, theory

Abstract: The direct ionization of the helium atom by intense coherent high-frequency short laser pulses is investigated theoretically from first principles. To this end, we solve numerically the time-dependent Schrödinger equation for the two-electron wave packet and its interaction with the linearly polarized pulse by the efficient time-dependent restricted-active-space configuration-interaction method (TD-RASCI). In particular, we consider photon energies which are nearly resonant for the $1s \rightarrow 2p$ excitation in the He^+ ion. Thereby, we investigate the dynamic interference of the photoelectrons of the same kinetic energy emitted at different times along the pulse in the two-electron system. In order to enable observation of the dynamic interference in the computed spectrum, the electron wave packets were propagated on large spatial grids over long times.

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Volume: 144 **Issue:** **First Page #:** 074309
Date Submitted: 7/28/16 12:00AM **Date Published:** 2/19/16 4:09AM
Publication Location:

Article Title: Tracking the photodissociation probability of D₂⁺ induced by linearly chirped laser pulses

Authors: András Csehi, Gábor J. Halász, Lorenz S. Cederbaum, and Ágnes Vibók

Keywords: attosecond, dissociation, theory

Abstract: In the presence of linearly varying frequency chirped laser pulses, the photodissociation dynamics of D₂⁺ is studied theoretically after ionization of D₂. As a completion of our recent work [A. Csehi et al., J. Chem. Phys. 143, 014305 (2015)], a comprehensive dependence on the pulse duration and delay time is presented in terms of total dissociation probabilities. Our numerical analysis carried out in the recently introduced light-induced conical intersection (LICI) framework clearly shows the effects of the changing position of the LICI which is induced by the frequency modulation of the chirped laser pulses. This impact is presented for positively, negatively, and zero chirped short pulses.

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Publication Identifier Type: DOI **Publication Identifier:** DOI: 10.1039/c6fd00139d
Volume: 194 **Issue:** **First Page #:** 479
Date Submitted: 2/6/17 12:00AM **Date Published:** 7/7/16 5:32PM
Publication Location:

Article Title: Towards controlling the dissociation probability by light-induced conical intersections

Authors: A. Csehi, G. J. Halász, L. S. Cederbaum, and Á. Vibók

Keywords: conical intersection, theory, deuterium ion, light induced

Abstract: Light-induced conical intersections (LICIs) can be formed both by standing or by running laser waves. The position of a LICI is determined by the laser frequency while the laser intensity controls the strength of the nonadiabatic coupling. Recently, it was shown within the LICI framework that linearly chirped laser pulses have an impact on the dissociation dynamics of the D₂⁺ molecule (J. Chem. Phys., 143, 014305, (2015); J. Chem. Phys., 144, 074309, (2016)). In this work we exploit this finding and perform calculations using chirped laser pulses in which the time dependence of the laser frequency is designed so as to force the LICI to move together with the field-free vibrational wave packet as much as possible. Since nonadiabaticity is strongest in the vicinity of the conical intersection, this is the first step towards controlling the dissociation process via the LICI.

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Date Submitted: 2/6/17 12:00AM

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Article Title: Core ionization initiates sub-femtosecond charge migration in the valence shell

Authors: A. I. Kuleff, N. V. Kryzhevoi, M. Pernpointner, and L. S. Cederbaum

Keywords: ionization, ultrafast, attosecond

Abstract: After the ionization of a valence electron, the created hole can migrate ultrafast from one end of the molecule to another. Because of the advent of attosecond pulse techniques, the measuring and understanding of charge migration has become a central topic in attosecond science. Here, we pose the hitherto unconsidered question whether ionizing a core electron will also lead to charge migration. It is found that the created hole in the core stays put, but in response to this hole interesting electron dynamics takes place which can lead to intense charge migration in the valence shell. This migration is typically faster than that after the ionization of a valence electron and transpires on a shorter time scale than the natural decay of the core hole by the Auger process, making the subject very challenging to attosecond science.

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Article Title: Attosecond pulses measured from the attosecond lighthouse

Authors: T. J. Hammond, Graham G. Brown, Kyung Taec Kim, D.M. Villeneuve, and P. B. Corkum

Keywords: attosecond, lighthouse

Abstract: The attosecond lighthouse is a method of using ultrafast wavefront rotation with high-harmonic generation to create a series of coherent, spatially separated attosecond pulses. Previously, temporal measurements by photoelectron streaking characterized isolated attosecond pulses created by manipulating the single-atom response^{1–4}. The attosecond lighthouse, in contrast, generates a series of pulses that spatially separate and become isolated by propagation. Here, we show that ultrafast wavefront rotation maintains the single-atom response (in terms of temporal character) of an isolated attosecond pulse over two octaves of bandwidth. Moreover, we exploit the unique property of the attosecond lighthouse—the generation of several isolated pulses—to measure the three most intense pulses. These pulses each have a unique spectrum and spectral phase.

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Volume: 40 **Issue:** 8 **First Page #:** 1768
Date Submitted: 7/28/16 12:00AM **Date Published:** 4/8/15 9:00PM
Publication Location:

Article Title: Controlling attosecond angular streaking with second harmonic radiation

Authors: T. J. Hammond, Kyung Taec Kim, Chunmei Zhang, D. M. Villeneuve, and P. B. Corkum

Keywords: Harmonic generation and mixing; Multiharmonic generation; Ultrafast nonlinear optics.

Abstract: High harmonic generation, which produces a coherent burst of radiation every half cycle of the driving field, has been combined with ultrafast wavefront rotation to create a series of spatially separated attosecond pulses, called the attosecond lighthouse. By adding a coherent second harmonic beam with polarization parallel to the fundamental, we decrease the generating frequency from twice per optical cycle to once. The increased temporal separation increases the pulse contrast. By scanning the carrier envelope phase, we see that the signal is 2 π periodic.

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Journal: Applied Physics Letters
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Date Submitted: 7/28/16 12:00AM **Date Published:** 6/7/16 4:31AM
Publication Location:

Article Title: Polarization Gating of High Harmonic Generation in the Water Window

Authors: Jie Li, Xiaoming Ren, Yanchun Yin, Yan Cheng, Eric Cunningham, Yi Wu, and Zenghu Chang

Keywords: attosecond, polarization gate, mid infrared

Abstract: We implement the polarization gating (PG) technique with a two-cycle, 1.7 μm driving field to generate an attosecond supercontinuum extending to the water window spectral region. The ellipticity dependence of the high harmonic yield over a photon energy range much broader than previous work is measured and compared with a semi-classical model. When PG is applied, the carrier-envelope phase (CEP) is swept to study its influence on the continuum generation. PG with one-cycle (5.7 fs) and two-cycle (11.3 fs) delay are tested, and both give continuous spectra spanning from 50 to 450 eV under certain CEP values, strongly indicating the generation of isolated attosecond pulses in the water window region.

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Journal: Optics Letters
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Volume: 41 **Issue:** 6 **First Page #:** 1142
Date Submitted: 7/28/16 12:00AM **Date Published:** 3/8/16 10:40PM
Publication Location:

Article Title: High-efficiency OPCPA in BiB3O6 for generation of 3 mJ, two-cycle, CEP-stable pulses at 1.7 μm

Authors: Yanchun Yin, Jie Li, Xiaoming Ren, Kun Zhao, Yi Wu, Eric Cunningham, Zenghu Chang

Keywords: Ultrafast lasers; Parametric oscillators and amplifiers.

Abstract: We produce a 3 mJ, two-cycle (11.4 fs), 1 kHz, carrier-envelope phase (CEP)-stable laser source at 1.7 μm via a three-stage Ti:sapphire-pumped optical parametric chirped pulse amplifier in BiB3O6. We achieve a pump-to-signal conversion efficiency of 18% in the last stage, which is, to the best of our knowledge, the highest yet achieved for near-octave bandwidth amplification. A f-to-2f measurement shows a CEP instability of 165 mrad over 1 h. This is an ideal light source for generating isolated attosecond pulses in the soft x-ray region.

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Volume: 93 **Issue:** **First Page #:** 033405
Date Submitted: 7/28/16 12:00AM **Date Published:** 3/5/16 6:51AM
Publication Location:

Article Title: Attosecond transient absorption in dense gases: Exploring the interplay between resonant pulse propagation and laser-induced line-shape control

Authors: Chen-Ting Liao, Arvinder Sandhu, Seth Camp, Kenneth J. Schafer, and Mette B. Gaarde

Keywords: line shape, attosecond, optical thickness

Abstract: We investigate the evolution of extreme ultraviolet (XUV) spectral line shapes in an optically thick helium gas under near-infrared (IR) perturbation. In our experimental and theoretical work, we systematically vary the IR intensity, time-delay, gas density, and IR polarization parameters to study line-shape modifications induced by collective interactions in a regime beyond the single-atom response of a thin, dilute gas. In both experiment and theory, we find that specific features in the frequency-domain absorption profile, and their evolution with propagation distance, can be attributed to the interplay between resonant attosecond pulse propagation and IR-induced phase shifts. Our calculations show that this interplay also manifests itself in the time domain, with the IR pulse influencing the reshaping of the XUV pulse propagating in the resonant medium.

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Volume: 154 **Issue:** **First Page #:** 234301
Date Submitted: 6/15/21 12:00AM **Date Published:** 6/15/21 7:57PM
Publication Location:

Article Title: Mapping wave packet bifurcation at a conical intersection in CH₃I by attosecond XUV transient absorption spectroscopy

Authors: K. F. Chang, H. Wang, S. M. Poullain, D. Prendergast, D. M. Neumark, and S. R. Leone

Keywords: XUV, Spectroscopy, Attosecond, Molecules

Abstract: Extreme ultraviolet (XUV) transient absorption spectroscopy has emerged as a sensitive tool for mapping the real-time structural and electronic evolution of molecules. Here, attosecond XUV transient absorption is used to track dynamics in the A-band of methyl iodide (CH₃I). Gaseous CH₃I molecules are excited to the A-band by a UV pump (277 nm, ~20 fs) and probed by attosecond XUV pulses targeting iodine I(4d) core-to-valence transitions. Owing to the excellent temporal resolution of the technique, passage through a conical intersection is mapped through spectral signatures of nonadiabatic wave packet bifurcation observed to occur at 15 ± 4 fs following UV photoexcitation. The observed XUV signatures and time dynamics are in agreement with previous simulations [H. Wang, M. Odelius, and D. Prendergast, J. Chem. Phys. 151, 124106 (2019)]. Due to the short duration of the UV pump pulse, coherent vibrational motion in the CH₃I ground state along the C–I stretch mode (538 ± 7 cm⁻¹) launched

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Volume: 6 **Issue:** **First Page #:** 26671
Date Submitted: 7/29/16 12:00AM **Date Published:** 5/27/16 11:25PM
Publication Location:

Article Title: Full characterization of an attosecond pulse generated using an infrared driver

Authors: Chunmei Zhang, Graham G. Brown, Kyung Taec Kim, D. M. Villeneuve & P. B. Corkum

Keywords: attosecond, mid infrared, spatial isolation

Abstract: The physics of attosecond pulse generation requires using infrared driving wavelength to reach the soft X-rays. However, with longer driving wavelength, the harmonic conversion efficiency drops significantly. It makes the conventional attosecond pulse measurement using streaking very difficult due to the low photoionization cross section in the soft X-rays region. In-situ measurement was developed for precisely this purpose. We use in-situ measurement to characterize, in both space and time, an attosecond pulse produced by ultrafast wavefront rotation of a 1.8 micron fundamental beam. We confirm what models suggest – that each beamlet is an isolated attosecond pulse in the time domain. We get almost constant flat wavefront curvature through the whole photon energy range. The measurement method is scalable to the soft X-ray spectral region.

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Volume: 24 **Issue:** 22 **First Page #:** 24989
Date Submitted: 2/6/17 12:00AM **Date Published:** 11/1/16 6:45PM
Publication Location:

Article Title: High-energy two-cycle pulses at 3.2 micron by a broadband-pumped dual-chirped optical parametric amplification

Authors: YANCHUN YIN, JIE LI, XIAOMING REN, YANG WANG, ANDREW CHEW, AND ZENGHU CHANG

Keywords: optical parametric amplifier, lithium niobate,

Abstract: A design for efficient generation of mid-infrared pulses at 3.2 micron is presented, which is based on numerical simulations of the broadband-pumped dual-chirped optical parametric amplification (DC-OPA) in LiNbO₃ doped with 5 mol.% MgO (MgO:LiNbO₃). The broadband seed can be generated by difference frequency generation in KTA using spectrally-broadened Ti:Sapphire lasers. The broad DC-OPA phase-matching bandwidth-spanning from 2.4 micron to 4.0 micron-is achieved by chirping both the broadband Ti:Sapphire pump pulses and the seed pulses in such a way that the individual temporal slice of pump spectrum is able to phase match that of seed spectrum. This phase matching scheme allows the use of longer crystals without gain narrowing or loss of conversion efficiency. The theoretical conversion efficiency from the pump to the idler reaches 19.1 %, enabling generation of a few hundred mJ of mid-IR energy with an available large-aperture MgO:LiNbO₃ crystal.

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Date Submitted: 2/6/17 12:00AM

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Article Title: Reconstruction of an excited-state molecular wave packet with attosecond transient absorption spectroscopy

Authors: Yan Cheng, Michael Chini, Xiaowei Wang, Alberto Gonz'alez-Castrillo, Alicia Palacios, Luca Argenti, Fe

Keywords: Attosecond, coherent superposition, spectroscopy

Abstract: Attosecond science promises to allow new forms of quantum control in which a broadband isolated attosecond pulse excites a molecular wave packet consisting of a coherent superposition of multiple excited electronic states. This electronic excitation triggers nuclear motion on the molecular manifold of potential energy surfaces and can result in permanent rearrangement of the constituent atoms. Here, we demonstrate attosecond transient absorption spectroscopy (ATAS) as a viable probe of the electronic and nuclear dynamics initiated in excited states of a neutral molecule by a broadband vacuum ultraviolet pulse. Owing to the high spectral and temporal resolution of ATAS, we are able to reconstruct the time evolution of a vibrational wave packet within the excited B $1\ u^+$ electronic state of H₂ via the laser-perturbed transient absorption spectrum.

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Article Title: Generating high-contrast, near single-cycle waveforms with third-order dispersion compensation

Authors: Henry Timmers, Yuki Kobayashi, Kristina F. Chang, Maurizio Reduzzi, Daniel M. Neumark, Stephen R. J.

Keywords: Ultrafast spectroscopy; Ultrafast nonlinear optics; X-rays, soft x-rays, extreme ultraviolet (EUV)

Abstract: Femtosecond laser pulses lasting only a few optical periods hold the potential for probing and manipulating the electronic degrees of freedom within matter. However, the generation of high-contrast, few-cycle pulses in the high power limit still remains nontrivial. In this Letter, we present the application of ammonium dihydrogen phosphate (ADP) as an optical medium for compensating for the higher-order dispersion of a carrier-envelope stable few-cycle waveform centered at 735 nm. The ADP crystal is capable of removing the residual third-order dispersion present in the spectral phase of an input pulse, resulting in near-transform-limited 2.9 fs pulses lasting only 1.2 optical cycles in duration. By utilizing these high-contrast, few-cycle pulses for high-harmonic generation, we are able to produce nanojoule-scale, isolated attosecond pulses.

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Journal: Physical Review A
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevA.95.031401
Volume: 95 **Issue:** 3 **First Page #:** 031401
Date Submitted: 7/12/17 12:00AM **Date Published:** 3/2/17 12:00AM
Publication Location:

Article Title: Attosecond transient-absorption dynamics of xenon core-excited states in a strong driving field

Authors: Yuki Kobayashi, Henry Timmers, Mazyar Sabbar, Stephen R. Leone, Daniel M. Neumark

Keywords: xenon, Floquet, attosecond, transient absorption, core-excited

Abstract: We present attosecond transient-absorption experiments on xenon 4d⁹ 6p core-level states resonantly driven by intense ($1.6 \times 10^{14} \text{ W/cm}^2$) few-cycle near-infrared laser pulses. In this strongly driven regime, broad induced absorption features with half-cycle (1.3-fs) delay-dependent modulation are observed over the range of 58–65 eV, predicted as a signature of the breakdown of the rotating-wave approximation in strong-field driving of Autler-Townes splitting [A. N. Pfeiffer and S. R. Leone, Phys. Rev. A 85, 053422 (2012)]. Relevant atomic states are identified by a numerical model involving three electronic states, and the mechanism behind the broad induced absorption is discussed in the Floquet formalism. These results demonstrate that a near-infrared field well into the tunneling regime can still control the optical properties of an atomic system over a several-electron-volt spectral range and with attosecond precision.

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Optics Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1364/OL.41.005365
Volume: 41 **Issue:** 22 **First Page #:** 5365
Date Submitted: 7/12/17 12:00AM **Date Published:** 11/1/16 2:00PM
Publication Location:

Article Title: Simultaneous generation of sub-5-femtosecond 400 nm and 800 nm pulses for attosecond extreme ultraviolet pump-probe spectroscopy

Authors: Hung-Tzu Chang, Michael Zurch, Peter M. Kraus, Lauren J. Borja, Daniel M. Neumark, Stephen R. Leone

Keywords: Ultrafast spectroscopy; Pulse compression; Ultrafast nonlinear optics

Abstract: Few-cycle laser pulses with wavelengths centered at 400 nm and 800 nm are simultaneously obtained through wavelength separation of ultrashort, spectrally broadened Vis–NIR laser pulses spanning 350–1100 nm wavelengths. The 400 nm and 800 nm pulses are separately compressed, yielding pulses with 4.4 fs and 3.8 fs duration, respectively. The pulse energy exceeds 5 μJ for the 400 nm pulses and 750 μJ for the 800 nm pulses. Intense 400 nm few-cycle pulses have a broad range of applications in nonlinear optical spectroscopy, which include the study of photochemical dynamics, semiconductors, and photovoltaic materials on few-femtosecond to attosecond time scales. The ultrashort 400 nm few-cycle pulses generated here not only extend the spectral range of the optical pulse for NIR–XUV attosecond pump–probe spectroscopy but also pave the way for two-color, three-pulse, multidimensional optical–XUV spectroscopy experiments.

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Volume: 194

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Date Submitted: 7/12/17 12:00AM

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Publication Location:

Article Title: Attosecond science in atomic, molecular, and condensed matter physics

Authors: Stephen R. Leone, Daniel M. Neumark

Keywords: attosecond, time-resolved, high harmonic, transient absorption

Abstract: Attosecond science represents a new frontier in atomic, molecular, and condensed matter physics, enabling one to probe the exceedingly fast dynamics associated with purely electronic dynamics in a wide range of systems. This paper presents a brief discussion of the technology required to generate attosecond light pulses and gives representative examples of attosecond science carried out in several laboratories. Attosecond transient absorption, a very powerful method in attosecond science, is then reviewed and several examples of gas phase and condensed phase experiments that have been carried out in the Leone/Neumark laboratories are described.

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Publication Location:

Article Title: State-resolved attosecond reversible and irreversible dynamics in strong optical fields

Authors: Mazyar Sabbar, Henry Timmers, Yi-Jen Chen, Allison K. Pymer, Zhi-Heng Loh, Scott G. Sayres, Stefar

Keywords: xenon, attosecond, transient absorption, state-resolved

Abstract: Strong-field ionization (SFI) is a key process for accessing real-time quantum dynamics of electrons on the attosecond timescale. The theoretical foundation of SFI was pioneered in the 1960s, and later refined by various analytical models. While asymptotic ionization rates predicted by these models have been tested to be in reasonable agreement for a wide range of laser parameters, predictions for SFI on the sub-laser-cycle timescale are either beyond the scope of the models or show strong qualitative deviations from full quantum-mechanical simulations. Here, using the unprecedented state specificity of attosecond transient absorption spectroscopy, we follow the real-time SFI process of the two valence spin-orbit states of xenon. The results reveal that the irreversible tunnelling contribution is accompanied by a reversible electronic population that exhibits an observable spin-orbit-dependent phase delay. A detailed theoretical analysis attributes this observation to transient ground

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Date Submitted: 7/12/17 12:00AM

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Publication Location:

Article Title: Ultrafast carrier thermalization and trapping in silicon-germanium alloy probed by extreme ultraviolet transient absorption spectroscopy

Authors: Michael Z?rch, Hung-Tzu Chang, Peter M. Kraus, Scott K. Cushing, Lauren J. Borja, Andrey Gandman,

Keywords: silicon germanium, core level, attosecond, electron, hole

Abstract: Semiconductor alloys containing silicon and germanium are of growing importance for compact and highly efficient photonic devices due to their favorable properties for direct integration into silicon platforms and wide tunability of optical parameters. Here, we report the simultaneous direct and energy-resolved probing of ultrafast electron and hole dynamics in a silicon-germanium alloy with the stoichiometry Si_{0.25}Ge_{0.75} by extreme ultraviolet transient absorption spectroscopy. Probing the photoinduced dynamics of charge carriers at the germanium M_{4,5}-edge (~ 30 eV) allows the germanium atoms to be used as reporter atoms for carrier dynamics in the alloy. The photoexcitation of electrons across the direct and indirect band gap into conduction band (CB) valleys and their subsequent hot carrier relaxation are observed and compared to pure germanium, where the Ge direct (DEgap; Ge; direct $\frac{1}{4}$ 0:8 eV) and Si_{0.25}Ge_{0.75} indirect gaps (DEgap; Si_{0.25}Ge_{0.75}; indirect $\frac{1}{4}$ 0:95 eV) are comparable in energy.

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Publication Location:

Article Title: Direct and simultaneous observation of ultrafast electron and hole dynamics in germanium

Authors: Michael Z?rch, Hung-Tzu Chang, Lauren J. Borja, Peter M. Kraus, Scott K. Cushing, Andrey Gandman,

Keywords: germanium, hole, electron, attosecond, transient absorption

Abstract: Understanding excited carrier dynamics in semiconductors is crucial for the development of photovoltaics and efficient photonic devices. However, overlapping spectral features in optical pump-probe spectroscopy often render assignments of separate electron and hole carrier dynamics ambiguous. Here, ultrafast electron and hole dynamics in germanium nanocrystalline thin films are directly and simultaneously observed by ultrafast transient absorption spectroscopy in the extreme ultraviolet at the germanium M_{4,5} edge. We decompose the spectra into contributions of electronic state blocking and photo-induced band shifts at a carrier density of $8 \times 10^{20} \text{ cm}^{-3}$. Separate electron and hole relaxation times are observed as a function of hot carrier energies. A first-order electron and hole decay of 1 ps suggests a Shockley-Read-Hall recombination mechanism. The simultaneous observation of electrons and holes with extreme ultraviolet transient absorption spectroscopy paves the way for investigating

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Journal: Journal of the Optical Society of America B

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Date Submitted: 7/12/17 12:00AM

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Publication Location:

Article Title: Extreme ultraviolet transient absorption of solids from femtosecond to attosecond timescales

Authors: Lauren J. Borja, M. Zurch, C. D. Pemmaraju, Martin Schultze, Krupa Ramasesha, Andrey Gandman, Ja

Keywords: Multiharmonic generation; Spectroscopy, semiconductors; Ultrafast processes in condensed matter, including semiconductors

Abstract: High-harmonic generation (HHG) produces ultrashort pulses of extreme ultraviolet radiation (XUV), which can be used for pump-probe transient absorption spectroscopy in metal oxides, semiconductors, and dielectrics. Femtosecond transient absorption on iron and cobalt oxides identifies ligand-to-metal charge transfer as the main spectroscopic transition, rather than metal-to-metal charge transfer or d-d transitions, upon photoexcitation in the visible. In silicon, attosecond transient absorption reveals that electrons tunnel into the conduction band from the valence band under strong-field excitation, to energies as high as 6 eV above the conduction band minimum. Extensions of these experiments to other semiconductors, such as germanium, and other transition metal oxides, such as vanadium dioxide, are discussed. Germanium is of particular interest because it should be possible to follow both electron and hole dynamics in a single measurement using transient XUV absorption.

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Publication Location:

Article Title: Attosecond pulses measured from the attosecond lighthouse

Authors: T. J. Hammond, Graham G. Brown, Kyung Taec Kim, D. M. Villeneuve, P. B. Corkum

Keywords: attosecond, lighthouse, pulse characterization

Abstract: The attosecond lighthouse is a method of using ultrafast wavefront rotation with high-harmonic generation to create a series of coherent, spatially separated attosecond pulses. Previously, temporal measurements by photoelectron streaking characterized isolated attosecond pulses created by manipulating the single-atom response 1–4. The attosecond lighthouse, in contrast, generates a series of pulses that spatially separate and become isolated by propagation. Here, we show that ultrafast wavefront rotation maintains the single-atom response (in terms of temporal character) of an isolated attosecond pulse over two octaves of bandwidth. Moreover, we exploit the unique property of the attosecond lighthouse—the generation of several isolated pulses—to measure the three most intense pulses. These pulses each have a unique spectrum and spectral phase.

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Volume: 49

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Publication Location:

Article Title: Time delay in molecular photoionization

Authors: P Hockett, E Frumker, D M Villeneuve, P B Corkum

Keywords: attosecond, photoionization, Wigner delay, angle-resolved, molecular

Abstract: Time-delays in the photoionization of molecules are investigated. As compared to atomic ionization, the time-delays expected from molecular ionization present a much richer phenomenon, with a strong spatial dependence due to the anisotropic nature of the molecular scattering potential. We investigate this from a scattering theory perspective, and make use of molecular photoionization calculations to examine this effect in representative homonuclear and heteronuclear diatomic molecules, nitrogen and carbon monoxide. We present energy and angle-resolved maps of the Wigner delay time for single-photon valence ionization, and discuss the possibilities for experimental measurements.

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Publication Location:

Article Title: Full characterization of an attosecond pulse generated using an infrared driver

Authors: Chunmei Zhang, Graham G. Brown, Kyung Taec Kim, D. M. Villeneuve, P. B. Corkum

Keywords: attosecond pulse, characterization, X-ray, streaking, attosecond lighthouse

Abstract: The physics of attosecond pulse generation requires using infrared driving wavelength to reach the soft X-rays. However, with longer driving wavelength, the harmonic conversion efficiency drops significantly. It makes the conventional attosecond pulse measurement using streaking very difficult due to the low photoionization cross section in the soft X-rays region. In-situ measurement was developed for precisely this purpose. We use in-situ measurement to characterize, in both space and time, an attosecond pulse produced by ultrafast wavefront rotation of a 1.8 μ m fundamental beam. We confirm what models suggest – that each beamlet is an isolated attosecond pulse in the time domain. We get almost constant flat wavefront curvature through the whole photon energy range. The measurement method is scalable to the soft X-ray spectral region.

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Publication Location:

Article Title: Attosecond optics and technology: progress to date and future prospects [Invited]

Authors: Zenghu Chang, Paul B. Corkum, Stephen R. Leone

Keywords: Ultrafast optics; Strong field laser physics; Multiphoton processes

Abstract: The milestones of attosecond optics research in the last 15 years are briefly reviewed, and the latest trends in applications in gaseous and condensed matter are introduced. An outlook on future development of attosecond soft x-ray sources and their application is provided.

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Date Submitted: 7/12/17 12:00AM **Date Published:** 8/1/16 9:00PM
Publication Location:

Article Title: Core Ionization Initiates Subfemtosecond Charge Migration in the Valence Shell of Molecules

Authors: Alexander I. Kuleff, Nikolai V. Kryzhevoi, Markus Pernpointner, Lorenz S. Cederbaum

Keywords: charge migration, valence orbital, core hole, attosecond

Abstract: After the ionization of a valence electron, the created hole can migrate ultrafast from one end of the molecule to another. Because of the advent of attosecond pulse techniques, the measuring and understanding of charge migration has become a central topic in attosecond science. Here, we pose the hitherto unconsidered question whether ionizing a core electron will also lead to charge migration. It is found that the created hole in the core stays put, but in response to this hole interesting electron dynamics takes place which can lead to intense charge migration in the valence shell. This migration is typically faster than that after the ionization of a valence electron and transpires on a shorter time scale than the natural decay of the core hole by the Auger process, making the subject very challenging to attosecond science.

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Journal: Faraday Discuss.
Publication Identifier Type: DOI **Publication Identifier:** 10.1039/C6FD00139D
Volume: 194 **Issue:** **First Page #:** 479
Date Submitted: 7/12/17 12:00AM **Date Published:** 7/8/16 4:00AM
Publication Location:

Article Title: Towards controlling the dissociation probability by light-induced conical intersections

Authors: Andras Csehi, Gabor J. Halasz, Lorenz S. Cederbaum, Agnes Vibok

Keywords: conical intersection, light induced, D2+

Abstract: Light-induced conical intersections (LICIs) can be formed both by standing or by running laser waves. The position of a LICI is determined by the laser frequency while the laser intensity controls the strength of the nonadiabatic coupling. Recently, it was shown within the LICI framework that linearly chirped laser pulses have an impact on the dissociation dynamics of the D2+ molecule (J. Chem. Phys., 143, 014305, (2015); J. Chem. Phys., 144, 074309, (2016)). In this work we exploit this finding and perform calculations using chirped laser pulses in which the time dependence of the laser frequency is designed so as to force the LICI to move together with the field-free vibrational wave packet as much as possible. Since nonadiabaticity is strongest in the vicinity of the conical intersection, this is the first step towards controlling the dissociation process via the LICI. Our showcase example is again the D2+ molecular ion. To demonstrate the impact of the LICIs on the dynamical properties

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Publication Location:

Article Title: Electronic decay through carbon chains

Authors: Alexander I. Kuleff

Keywords: Non-local electronic decay Multielectron wave-packet propagation Fluorinated cumulenones

Abstract: Using the multielectron wave-packet propagation method the electronic decay of O2s vacancy in fluorinated cumulenones, OCnF2, containing a chain of up to five carbons is traced in time and space. It is shown that in all studied cases this state decays non-locally by emitting an electron from the remote fluorines. Even in the pentatetraenone case, where the oxygen and the fluorines are more than 7 Å apart, this non-local decay is extremely efficient, with a time constant of about 5 fs. The process can be viewed as an ultrafast energy transfer through the carbon chain and thus our systematic study allows to shed some light on the dependence of the time scale of the electron-correlation driven energy transfer through a medium.

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Article Title: Impact of two-electron dynamics and correlations on high-order-harmonic generation in He

Authors: Anton N. Artemyev, Lorenz S. Cederbaum, Philipp V. Demekhin

Keywords: high harmonic, helium, electron correlation, ionization

Abstract: The interaction of a helium atom with an intense, short, 800-nm, laser pulse is studied theoretically beyond the single-active-electron approximation. For this purpose, the time-dependent Schrödinger equation for a two electron wave packet driven by a linearly polarized infrared pulse is solved by the time-dependent restricted-active-space configuration-interaction method in the dipole velocity gauge. By systematically extending the space of active configurations, we investigate the role of the collective two-electron dynamics in the strong-field ionization and high-order-harmonic generation processes. Our numerical results demonstrate that allowing both electrons in He to be dynamically active results in a considerable extension of the computed high-order-harmonic generation spectrum.

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Article Title: Competition between Light-Induced and Intrinsic Nonadiabatic Phenomena in Diatomics

Authors: Andras Csehi, Gabor J. Halasz, Lorenz S. Cederbaum, Agnes Vibok

Keywords: conical intersection, sodium iodide, LICI, light induced conical intersection

Abstract: Nonadiabatic effects arise due to avoided crossings or conical intersections that are either present naturally in field-free space or induced by a classical laser field in a molecule. Recently, it was demonstrated that nonadiabatic effects in diatomics can also be created in an optical cavity. Here, the quantized radiation field mixes the nuclear and electronic degrees of freedom. We show the equivalence of using the cavity's quantized field and the classical laser field as usually done for molecules. This is demonstrated for NaI, which exhibits a pronounced natural (intrinsic) avoided crossing that competes with the avoided crossing induced by the field. Furthermore, rotating molecules exhibit lightinduced conical intersections (LICIs) in classical laser light, and we also investigate the impact of these intersections. For NaI, we undoubtedly demonstrate a significant difference between the impact of the laser-induced avoided crossing and that of the LICI on the dynamics of the molecu

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Publication Location:

Article Title: Probing autoionizing states of molecular oxygen with XUV transient absorption: Electronic-symmetry-dependent line shapes and laser-induced modifications

Authors: Chen-Ting Liao, Xuan Li, Daniel J. Haxton, Thomas N. Rescigno, Robert R. Lucchese, C. William McCu

Keywords: Atomic and molecular processes in external fields, Attosecond XUV spectroscopy, Optical transient phenomenon, Configuration interaction, Strong-field induced spectra

Abstract: We used extreme ultraviolet (XUV) transient absorption spectroscopy to study the autoionizing Rydberg states of oxygen in an electronically- and vibrationally-resolved fashion. XUV pulse initiates molecular polarization and near-infrared pulse perturbs its evolution. Transient absorption spectra show positive optical-density (OD) change in the case of ns²g and nd²g autoionizing states of oxygen and negative OD change for nd²g states. Multiconfiguration time-dependent Hartree-Fock (MCTDHF) calculations are used to simulate the transient absorption and the resulting spectra and temporal evolution agree with experimental observations. We model the effect of near infrared perturbation on molecular polarization and find that the laser-induced phase-shift model agrees with the experimental and MCTDHF results, while the laser-induced attenuation model does not. We relate the electronic-state-symmetry-dependent sign of the OD change to the Fano parameters of the static absorption line shapes.

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Publication Location:

Article Title: XUV Transient Absorption Spectroscopy: Probing Laser-Perturbed Dipole Polarization in Single Atom, Macroscopic, and Molecular Regimes

Authors: Chen-Ting Liao, Arvinder Sandhu

Keywords: High-harmonic generation, Atomic and molecular processes in external fields, Attosecond XUV spectroscopy, Optical transient phenomenon, Strong-field induced spectra

Abstract: We employ an extreme ultraviolet (XUV) pulse to impulsively excite dipole polarization in atoms or molecules, which corresponds to coherently prepared superposition of excited states. A delayed near infrared (NIR) pulse then perturbs the fast-evolving polarization, and the resultant absorbance change is monitored in dilute helium, dense helium, and sulfur hexafluoride (SF₆) molecules. We observe and quantify the time-dependence of various transient phenomena in helium atoms, including laser-induced phase (LIP), time-varying (AC) Stark shift, quantum path interference, and laser-induced continuum structure. In the case of dense helium targets, we discuss nonlinear macroscopic propagation effects pertaining to LIP and resonant pulse propagation, which account for the appearance of new spectral features in transient lineshapes. We then use tunable NIR photons to demonstrate the wavelength dependence of the transient laser induced effects. In the case of molecular polarization experiment i

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Article Title: Laser waveform control of extreme ultraviolet high harmonics from solids

Authors: Yong Sing You, Mengxi Wu, Yanchun Yin, Andrew Chew, Xiaoming Ren, Shima Gholam-Mirzaei, Dana

Keywords: Ultrafast processes in condensed matter, including semiconductors; Ultrafast phenomena; Ultrafast nonlinear optics

Abstract: Solid-state high-harmonic sources offer the possibility of compact, high-repetition-rate attosecond light emitters. However, the time structure of high harmonics must be characterized at the sub-cycle level. We use strong two-cycle laser pulses to directly control the time-dependent nonlinear current in single-crystal MgO, leading to the generation of extreme ultraviolet harmonics. We find that harmonics are delayed with respect to each other, yielding an atto-chirp, the value of which depends on the laser field strength. Our results provide the foundation for attosecond pulse metrology based on solid-state harmonics and a new approach to studying sub-cycle dynamics in solids.

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Article Title: Towards Terawatt Sub-Cycle Long-Wave Infrared Pulses via Chirped Optical Parametric Amplification and Indirect Pulse Shaping

Authors: Yanchun Yin, Andrew Chew, Xiaoming Ren, Jie Li, Yang Wang, Yi Wu, Zenghu Chang

Keywords: optical parametric amplifier, sub-cycle, infrared, laser

Abstract: We present an approach for both efficient generation and amplification of 4–12 μm pulses by tailoring the phase matching of the nonlinear crystal Zinc Germanium Phosphide (ZGP) in a narrowband-pumped optical parametric chirped pulse amplifier (OPCPA) and a broadband-pumped dual-chirped optical parametric amplifier (DC-OPA), respectively. Preliminary experimental results are obtained for generating 1.8–4.2 μm super broadband spectra, which can be used to seed both the signal of the OPCPA and the pump of the DC-OPA. The theoretical pump-to-idler conversion efficiency reaches 27% in the DC-OPA pumped by a chirped broadband Cr²⁺: ZnSe/ZnS laser, enabling the generation of Terawatt-level 4–12 μm pulses with an available large-aperture ZGP. Furthermore, the 4–12 μm idler pulses can be compressed to sub-cycle pulses by compensating the tailored positive chirp of the idler pulses using the bulk compressor NaCl, and by indirectly controlling the higher-order idler phase through tuning the signal (2

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Publication Location:

Article Title: Attosecond pulse generation isolated with an asymmetric polarization gating

Authors: Gao Chen, Eric Cunningham, Zenghu Chang

Keywords: High harmonic generation; isolated attosecond pulse production

Abstract: High harmonics generated using the polarization gating technique are simulated under the strong-field approximation for the cases of equal and unequal ratios between the amplitudes of the two counter-rotating pulses. The effect of the field asymmetry is observed on the high harmonic yield and cut-off in the frequency domain and on the attosecond pulse duration and satellite peak contrast in the time domain. The case where the strong pulse comes later is more favourable in terms of generating a higher cut-off and a shorter pulse duration. It is predicted that thin metal filters can reduce the impact of satellite pulses while simultaneously reducing the pulse duration.

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Publication Location:

Article Title: High-energy two-cycle pulses at 3.2 micron by a broadband-pumped dual-chirped optical parametric amplification

Authors: Yanchun Yin, Jie Li, Xiaoming Ren, Yang Wang, Andrew Chew, Zenghu Chang

Keywords: Ultrafast lasers; Parametric oscillators and amplifiers

Abstract: A design for efficient generation of mid-infrared pulses at 3.2 μm is presented, which is based on numerical simulations of the broadband-pumped dual-chirped optical parametric amplification (DC-OPA) in LiNbO₃ doped with 5 mol.% MgO (MgO:LiNbO₃). The broadband seed can be generated by difference frequency generation in KTA using spectrally-broadened Ti:Sapphire lasers. The broad DC-OPA phase-matching bandwidth-spanning from 2.4 μm to 4.0 μm -is achieved by chirping both the broadband Ti:Sapphire pump pulses and the seed pulses in such a way that the individual temporal slice of pump spectrum is able to phase match that of seed spectrum. This phase matching scheme allows the use of longer crystals without gain narrowing or loss of conversion efficiency. The theoretical conversion efficiency from the pump to the idler reaches 19.1 %, enabling generation of a few hundred mJ of mid-IR energy with an available large-aperture MgO:LiNbO₃ crystal. Furthermore, the commercially available acousto

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Journal: Scientific Reports

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Publication Identifier: 10.1038/s41598-017-11652-9

Volume: 7

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Publication Location:

Article Title: Generation of octave-spanning mid-infrared pulses from cascaded second-order nonlinear processes in a single crystal

Authors: Yanchun Yin, Xiaoming Ren, Andrew Chew, Jie Li, Yang Wang, Fengjiang Zhuang, Yi Wu, Zenghu Cha

Keywords: mid infrared, laser, difference frequency generation. octave spanning

Abstract: We report on experimental generation of a 6.8 μJ laser pulse spanning from 1.8 to 4.2 μm from cascaded second-order nonlinear processes in a 0.4-mm BiB₃O₆ (BIBO) crystal. The nonlinear processes are initiated by intra-pulse difference frequency generation (DFG) using spectrally broadened Ti:Sapphire spectrum, followed by optical parametric amplification (OPA) of the DFG pulse. The highest energy, 12.6 μJ , is achieved in a 0.8-mm BIBO crystal with a spectrum spanning from 1.8 to 3.5 μm . Such cascaded nonlinear processes are enabled by the broadband pump and the coincident phase matching angle of DFG and OPA. The spectrum is initiated from the DFG process and is thus expected to have passive stable carrier-envelope phase, which can be used to seed either a chirped pulse amplifier (CPA) or an optical parametric chirped pulse amplifier (OPCPA) for achieving high-energy few-cycle midinfrared pulses. Such cascaded second-order nonlinear processes can be found in many other crystals such as K

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Publication Location:

Article Title: 53-Attosecond X-ray Pulses Reach the Carbon K-edge

Authors: Jie Li, Xiaoming Ren, Yanchun Yin, Kun Zhao, Andrew Chew, Yan Cheng, Eric Cunningham, Yang War

Keywords: attosecond pulse, x-ray, high harmonic

Abstract: The motion of electrons in the microcosm occurs on a time scale set by the atomic unit of time—24 attoseconds. Attosecond pulses at photon energies corresponding to the fundamental absorption edges of matter, which lie in the soft X-ray regime above 200 eV, permit the probing of electronic excitation, chemical state, and atomic structure. Here we demonstrate a soft X-ray pulse duration of 53 as and single pulse streaking reaching the carbon K-absorption edge (284 eV) by utilizing intense two-cycle driving pulses near 1.8- μ m center wavelength. Such pulses permit studies of electron dynamics in live biological samples and next-generation electronic materials such as diamond.

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Journal: Physical Review A

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Publication Location:

Article Title: Variational treatment of electron-polyatomic molecule scattering calculations using adaptive overset grids

Authors: Loren Greenman, Robert R. Lucchese, and C. William McCurdy

Keywords: Kohn variational, electron scattering, methane

Abstract: The complex Kohn variational method for electron–polyatomic-molecule scattering is formulated using an overset-grid representation of the scattering wave function. The overset grid consists of a central grid and multiple dense atom-centered subgrids that allow the simultaneous spherical expansions of the wave function about multiple centers. Scattering boundary conditions are enforced by using a basis formed by the repeated application of the free-particle Green's function and potential $\hat{G}+0V$ on the overset grid in a Born-Arnoldi solution of the working equations. The theory is shown to be equivalent to a specific Padé approximant to the T matrix and has rapid convergence properties, in both the number of numerical basis functions employed and the number of partial waves employed in the spherical expansions. The method is demonstrated in calculations on methane and CF₄ in the static-exchange approximation and compared in detail with calculations performed with the numerical Schwinger

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Journal: Phys. Chem. Chem. Phys.
Publication Identifier Type: DOI **Publication Identifier:** 10.1039/c7cp02164j
Volume: 19 **Issue:** **First Page #:** 19656
Date Submitted: 6/25/18 12:00AM **Date Published:** 5/4/17 7:00AM
Publication Location:

Article Title: Intrinsic and light-induced nonadiabatic phenomena in the NaI molecule

Authors: A. Csehi, G. J. Halász, L. S. Cederbaum, and Á. Vibók

Keywords: light induced, conical intersection, sodium iodide

Abstract: Nonadiabatic effects play a very important role in controlling chemical dynamical processes. They are strongly related to avoided crossings (AC) or conical intersections (CIs) which can either be present naturally or induced by classical laser light in a molecular system. The latter are named as “light-induced avoided crossings” (LIACs) and “light-induced conical intersections” (LICIs). By performing one or two dimensional quantum dynamical calculations LIAC and LICI situations can easily be created even in diatomic molecules. Applying such calculations for the NaI molecule, which is a strongly coupled diatomic in field free case, significant differences between the impact of the LIAC and LICI on the ground state population dynamics were observed. Moreover, obtained results undoubtedly demonstrate that the effect of the LIAC and LICI on the dynamics strongly depends on the intensity and the frequency of the applied laser field as well as the permanent dipole moments of the molecule

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Journal: Nature Communication
Publication Identifier Type: DOI **Publication Identifier:** 10.1038/s41467-017-01133-y
Volume: 8 **Issue:** **First Page #:** 1018
Date Submitted: 6/27/18 12:00AM **Date Published:** 10/18/17 2:00PM
Publication Location:

Article Title: Few-femtosecond passage of conical intersections in the benzene cation

Authors: M. C. E. Galbraith, S. Scheit, N. V. Golubev, G. Reitsma, N. Zhavoronkov, V. Despré, F. Lépine, A. I. Ku

Keywords: conical intersection, benzene cation,

Abstract: Observing the crucial first few femtoseconds of photochemical reactions requires tools typically not available in the femtochemistry toolkit. Such dynamics are now within reach with the instruments provided by attosecond science. Here, we apply experimental and theoretical methods to assess the ultrafast nonadiabatic vibronic processes in a prototypical complex system—the excited benzene cation. We use few-femtosecond duration extreme ultraviolet and visible/near-infrared laser pulses to prepare and probe excited cationic states and observe two relaxation timescales of 11 ± 3 fs and 110 ± 20 fs. These are interpreted in terms of population transfer via two sequential conical intersections. The experimental results are quantitatively compared with state-of-the-art multi-configuration time-dependent Hartree calculations showing convincing agreement in the timescales. By characterising one of the fastest internal conversion processes studied to date, we enter an extreme regime of ultrafast

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Journal: J. Phys. B: Roadmap for Ultrafast X-ray Atomic and Molecular Physics

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Publication Location:

Article Title: Attosecond science: from transient absorption to multidimensional spectroscopies

Authors: Yuki Kobayashi, Maurizio Reduzzi, Kristina F. Chang, Henry Timmers, Daniel M. Neumark, Stephen R. Leone

Keywords: attosecond, transient absorption, roadmap

Abstract: X-ray free electron lasers (XFELs) and tabletop sources of x-rays based upon high harmonic generation (HHG) have revolutionized the field of ultrafast x-ray atomic and molecular physics largely due to an explosive growth in capabilities in the past decade. XFELs now provide unprecedented intensities (1020 W/cm²) of monochromatic x-rays at wavelengths down to ~1 Ångström, and, HHG provides unprecedented time resolution (~60 attoseconds) and a correspondingly large coherent bandwidth at longer wavelengths. For context, timescales can be referenced to the Bohr orbital period in hydrogen atom of 150 attoseconds and the hydrogen-molecule vibrational period of 8 femtoseconds; wavelength scales can be referenced to the chemically significant carbon K-edge at a photon energy of ~280 eV (44 Ångströms) and the bond length in methane of ~1 Ångström. With these modern x-ray sources one now has the ability to focus on individual atoms, even when embedded in a complex ...

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Date Submitted: 6/25/18 12:00AM

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Article Title: Femtosecond tracking of carrier relaxation in germanium with extreme ultraviolet transient reflectivity

Authors: Christopher J. Kaplan, Peter M. Kraus, Andrew D. Ross, Michael Zürch, Scott K. Cushing, Marieke F. J. van der Vliet

Keywords: Extreme ultraviolet, XUV, transient reflectivity, germanium

Abstract: Extreme ultraviolet (XUV) transient reflectivity around the germanium M_{4,5} edge (3d core-level to valence transition) at 30 eV is advanced to obtain the transient dielectric function of crystalline germanium [100] on femtosecond to picosecond time scales following photoexcitation by broadband visible-to-infrared (VIS/NIR) pulses. By fitting the transient dielectric function, carrier-phonon induced relaxations are extracted for the excited carrier distribution. The measurements reveal a hot electron relaxation rate of 3.2 ± 0.2 ps attributed to the X-L intervalley scattering and a hot hole relaxation rate of 600 ± 300 fs ascribed to intravalley scattering within the heavy hole (HH) band, both in good agreement with previous work. An overall energy shift of the XUV dielectric function is assigned to a thermally induced band gap shrinkage by formation of acoustic phonons, which is observed to be on a timescale of 4–5 ps, in agreement with previously measured optical phonon lifetimes. The

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Article Title: The ultrafast X-ray spectroscopic revolution in chemical dynamics

Authors: Peter M. Kraus, Michael Zürrich, Scott K. Cushing, Daniel M. Neumark, Stephen R. Leone

Keywords: review, x-ray, time dynamics, chemical dynamics, attosecond, femtosecond

Abstract: The past two decades have seen rapid developments in short- pulse X- ray sources, which have enabled the study of nuclear and electronic dynamics by ultrafast X- ray spectroscopies with unprecedented time resolution ranging from nanoseconds to attoseconds. In this Perspective, we discuss some of the major achievements in the study of nuclear and electronic dynamics with X- ray pulses produced by high- harmonic, free- electron-laser and synchrotron sources. The particular dynamic processes probed by X- ray radiation highlighted in this Perspective are electronic coherences on attosecond to femtosecond timescales, chemical reactions, such as dissociations, and pericyclic ring- openings, spincrossover dynamics, ligand- exchange dynamics and structural deformations in excited states. X- ray spectroscopic probing of chemical dynamics holds great promise for the future owing to the ongoing developments of new spectroscopies, such as four- wave mixing, and the continuous improvements in emerg

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Volume: 11 **Issue:** 9 **First Page #:** 594
Date Submitted: 6/27/18 12:00AM **Date Published:** 8/22/17 4:00AM
Publication Location:

Article Title: Integrating solids and gases for attosecond pulse generation

Authors: T. J. Hammond, Sylvain Monchocé, Chunmei Zhang, Giulio Vampa, Dennis Klug, A. Yu. Naumov, D. M.

Keywords: attosecond, few-cycle, high harmonic

Abstract: Control of the field of few-cycle optical pulses has had an enormous impact on attosecond science. Subcycle pulses open the potential for non-adiabatic phase matching while concentrating the electric field so it can be used most efficiently. However, subcycle field transients have been difficult to generate. We exploit the perturbative response of a sub-100 nm thick monocrystalline quartz plate irradiated by an intense few-cycle 1.8 μm pulse, which creates a phase-controlled supercontinuum spectrum. Within the quartz, the pulse becomes space–time coupled as it generates a parallel second harmonic. Vacuum propagation naturally leads to a subcycle electric-field transient whose envelope is sculpted by the carrier envelope phase of the incident radiation. We show that a second medium (either gas or solid) can generate isolated attosecond pulses in the extreme ultraviolet region. With no optical elements between the components, the process is scalable to very high energy pulses and allows

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Date Submitted: 6/27/18 12:00AM **Date Published:** 7/19/17 2:00PM
Publication Location:

Article Title: Producing and controlling half-cycle near-infrared electric-field transients

Authors: T. J. Hammond, D. M. Villeneuve, P. B. Corkum

Keywords: spectral phase, attosecond, pulse control

Abstract: In a few-cycle laser pulse, the peak field strength depends on the carrier envelope phase. Concurrently, coherent control requires the measurement and manipulation of the spectral phase of a light pulse to influence a dynamical process that has multiple interfering pathways. Here, we exploit the interference of second harmonic generation and self-phase modulation in an 80 μm thick quartz plate due to a two-cycle pulse centered at 1.8 μm with peak intensity $3 \times 10^{13} \text{ W/cm}^2$ to generate half-cycle electric field transients. In a monolithic step, we transform a measurement of the carrier envelope phase to the control over the pulse evolution with subcycle temporal accuracy. The high-intensity subcycle transient is scalable in pulse energy and will be useful for strong field physics and attosecond science: the ultrashort infrared pulse can generate isolated attosecond pulses from low bandgap semiconductor materials, and will be able to optically control currents on a subfemtosecond timescale

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Date Submitted: 6/27/18 12:00AM **Date Published:** 8/22/17 2:00PM
Publication Location:

Article Title: Molecular Frame Reconstruction Using Time-Domain Photoionization Interferometry

Authors: Claude Marceau, Varun Makhija, Dominique Platzer, A. Yu. Naumov, P. B. Corkum, Albert Stolow, D. M.

Keywords: photoionization, interfereometer, nitrogen

Abstract: Photoionization of molecular species is, essentially, a multipath interferometer with both experimentally controllable and intrinsic molecular characteristics. In this work, XUV photoionization of impulsively aligned molecular targets (N_2) is used to provide a time-domain route to “complete” photoionization experiments, in which the rotational wave packet controls the geometric part of the photoionization interferometer. The data obtained is sufficient to determine the magnitudes and phases of the ionizationmatrix elements for all observed channels, and to reconstruct molecular frame interferograms from lab frame measurements. In principle, this methodology provides a time-domain route to complete photoionization experiments and themolecular frame, which is generally applicable to any molecule (no prerequisites), for all energies and ionization channels.

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Date Submitted: 6/27/18 12:00AM **Date Published:** 11/7/17 4:00PM
Publication Location:

Article Title: Optical gain in rotationally excited nitrogen molecular ions

Authors: Ali Azarm, Paul Corkum, Pavel Polynkin

Keywords: laser gain, nitrogen ion, stimulated emission

Abstract: We pump low-pressure nitrogen gas with ionizing femtosecond laser pulses at 1.5 μ m wavelength. The resulting rotationally excited N₂⁺ molecular ions generate directional, forward-propagating stimulated and isotropic spontaneous emissions at 428 nm wavelength. Through high-resolution spectroscopy of these emissions, we quantify rotational population distributions in the upper and lower emission levels. We show that these distributions are shifted with respect to each other, which has a strong influence on the transient optical gain in this system. Although we find that electronic population inversion exists in our particular experiment, we show that sufficient dissimilarity of rotational distributions in the upper and lower emission levels could, in principle, lead to gain without net electronic population inversion.

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Volume: 120 **Issue:** 13 **First Page #:** 133208
Date Submitted: 6/27/18 12:00AM **Date Published:** 3/30/18 9:00PM
Publication Location:

Article Title: Testing the Role of Recollision in N₂⁺ Air Lasing

Authors: Mathew Britton, Patrick Laferrière, Dong Hyuk Ko, Zhengyan Li, Fanqi Kong, Graham Brown, Andrei Na

Keywords: nitrogen ion, filament, strong field, laser gain

Abstract: It has been known for many years that during filamentation of femtosecond light pulses in air, gain is observed on the B to X transition in N₂⁺. While the gain mechanism remains unclear, it has been proposed that recollision, a process that is fundamental to much of strong field science, is critical for establishing gain. We probe this hypothesis by directly comparing the influence of the ellipticity of the pump light on gain in air filaments. Then, we decouple filamentation from gain by measuring the gain in a thin gas jet that we also use for high harmonic generation. The latter allows us to compare the dependence of the gain on the ellipticity of the pump with the dependence of the high harmonic signal on the ellipticity of the fundamental. We find that gain and harmonic generation have very different behavior in both filaments and in the jet. In fact, in a jet we even measure gain with circular polarization. Thus, we establish that recollision does not play a significant role in c

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Article Title: Near-field imaging for single-shot waveform measurements

Authors: TJ Hammond, Aleksey Korobenko, A Yu Naumov, D M Villeneuve, Paul B Corkum, Dong Hyuk Ko

Keywords: few-cycle, strong-field, attosecond, pulse characterization

Abstract: The accurate measurement of ultrashort pulses is necessary to optimize pulse compression for nonlinear optics and strong-field physics. Additionally, field-sensitive time-domain techniques enable measuring femtosecond and sub-femtosecond dynamics. In this paper, we introduce a single-shot measurement to characterize few-cycle fields. This polarization-sensitive time domain measurement can retrieve the waveform of pulses shorter than two-cycles in duration with a temporal window of hundreds of femtoseconds. We measure how the field changes after propagation through dispersive optics, and we measure the sub-femtosecond phase shift imposed by the nonlinear intensity dependent response of a dielectric. Real-time field measurements will benefit low repetition rate systems used in strong-field physics and attosecond science, while the large temporal window complements Fourier transform spectroscopy.

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Publication Location:

Article Title: Generation of 1 kHz, 2.3 mJ, 88 fs, 2.5 μm pulses from a Cr²⁺:ZnSe chirped pulse amplifier

Authors: Xiaoming Ren, Lam H. Mach, Yanchun Yin, Yang Wang, Zenghu Chang

Keywords: mid infrared laser, optical parametric amplifier, attosecond

Abstract: We demonstrate the generation of 2.3 mJ, 88 fs, 2.5 μm laser pulses at 1 kHz repetition rate from a three-stage chirped pulse amplifier employing Cr²⁺:ZnSe crystals as the active gain media. 5 μJ seed of the amplifier is obtained via intrapulse difference frequency generation in a bismuth triborate (BIBO) crystal from spectrally broadened Ti:Sapphire amplifier output. A multi-pass amplifier followed by two single-pass amplifiers pumped by Q-switched Ho:YAG lasers boost the pulse energy to 6.5 mJ, yielding 2.3 mJ, 88 fs pulses upon pulse compression. Our results show the highest peak power at 2.5 μm with 1 kHz repetition rate. Such a laser will be a powerful source for studying strong-field physics and extending high-harmonic generation towards the keV region.

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Article Title: Enhanced high-order harmonic generation driven by a wavefront corrected high-energy laser

Authors: Yang Wang, Tianyi Guo, Jialin Li, Jian Zhao, Yanchun Yin, Xiaoming Ren, Jie Li, Yi Wu, Matthew Weidr

Keywords: high-order harmonics, XUV, wavefront, high flux attosecond

Abstract: We developed and implemented an experimental setup for the generation of high-energy high-order harmonics in argon gas for nonlinear experiments in the extreme ultraviolet (XUV) spectral range. High-order harmonics were generated by loosely focusing a high-energy laser pulse centered around 800 nm into a gas cell filled with argon in a phase-matching condition. The wavefront distortions of the driving pulse were corrected by a deformable mirror, and the XUV conversion efficiency was significantly improved due to the excellent beam profile at focus. A high-damage-threshold beam splitter was used to eliminate high-energy driving pulses copropagating with high-order harmonics. The setup has a potential to provide intense ultrashort XUV pulses reaching the intensity levels required for nonlinear experiments in the XUV spectral range.

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Article Title: Attosecond transient absorption spectrum of argon at the L_{2,3} edge

Authors: Andrew Chew, Nicolas Douguet, Coleman Cariker, Jie Li, Eva Lindroth, Xiaoming Ren, Yanchun Yin, Lu

Keywords: argon, attosecond, transient absorption, x-ray

Abstract: Progress in high-harmonic generation has led to high-energy attosecond pulses with cutoff above the carbon 1s edge (283.8 eV). These pulses are essential to extend time-resolved spectroscopies to the water window in order to control electron dynamics in solvated organic species. Here we report a step towards this goal: the measurement, with subcycle time resolution, of the attosecond transient absorption spectrum of argon at the 2p_{3/2} L_{2,3} edge (250 eV) in the presence of a short-wave infrared control pulse. The measurements, supported by theoretical simulations, demonstrate the concurrent role of Auger decay and tunnel ionization in the driven evolution of inner-valence holes of polyelectronic atoms.

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Article Title: Attosecond light sources in the water window

Authors: Xiaoming Ren, Jie Li, Yanchun Yin, Kun Zhao, Andrew Chew, Yang Wang, Shuyuan Hu, Yan Cheng, Ei

Keywords: high harmonic generation, attosecond laser, water window x-ray, attosecond laser characterization

Abstract: As a compact and burgeoning alternative to synchrotron radiation and free-electron lasers, high harmonic generation (HHG) has proven its superiority in static and time-resolved extreme ultraviolet spectroscopy for the past two decades and has recently gained many interests and successes in generating soft x-ray emissions covering the biologically important water window spectral region. Unlike synchrotron and free-electron sources, which suffer from relatively long pulse width or large time jitter, soft x-ray sources from HHG could offer attosecond time resolution and be synchronized with their driving field to investigate time-resolved near edge absorption spectroscopy, which could reveal rich structural and dynamical information of the interrogated samples. In this paper, we review recent progresses on generating and characterizing attosecond light sources in the water window region. We show our development of an energetic, two-cycle, carrier-envelope phase stable laser source at 1.7

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Publication Location:

Article Title: Generation of high-energy narrowband 2.05 μm pulses for seeding a Ho:YLF laser

Authors: Yanchun Yin, Xiaoming Ren, Yang Wang, Fengjiang Zhuang, Jie Li, Zenghu Chang

Keywords: Ultrafast lasers, Nonlinear optics, parametric processes

Abstract: We experimentally demonstrate efficient generation of high-energy (82 μJ) narrowband 2.05 μm pulses pumped with 1 mJ broadband Ti:sapphire laser pulses, utilizing dual-chirped optical parametric amplification (DC-OPA) in a BBO crystal. The narrowband 2.05 μm pulses will be primarily used for seeding an Ho:YLF laser, which solves the synchronization issue when Ti:sapphire and Ho:YLF lasers are needed for developing midinfrared lasers. The narrowband 2.05 μm pulse from the unique DC-OPA design can seed the Ho:YLF laser much more efficiently than the broadband 2.05 μm pulse from traditional OPA technology.

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Volume: 8

Issue: 1

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Date Submitted: 6/27/18 12:00AM

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Publication Location:

Article Title: High-harmonic generation in amorphous solids

Authors: Yong Sing You, Yanchun Yin, Yi Wu, Andrew Chew, Xiaoming Ren, Fengjiang Zhuang, Shima Gholam-

Keywords: high harmonic, solid

Abstract: High-harmonic generation in isolated atoms and molecules has been widely utilized in extreme ultraviolet photonics and attosecond pulse metrology. Recently, high-harmonic generation has been observed in solids, which could lead to important applications such as all-optical methods to image valence charge density and reconstruct electronic band structures, as well as compact extreme ultraviolet light sources. So far these studies are confined to crystalline solids; therefore, decoupling the respective roles of long-range periodicity and high density has been challenging. Here we report the observation of high-harmonic generation from amorphous fused silica. We decouple the role of long-range periodicity by comparing harmonics generated from fused silica and crystalline quartz, which contain the same atomic constituents but differ in long-range periodicity. Our results advance current understanding of the strong-field processes leading to high-harmonic generation in solids with implicati

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Publication Location:

Article Title: Direct Signatures of Light-Induced Conical Intersections on the Field-Dressed Spectrum of Na₂

Authors: Tamás Szidarovszky, Gábor J. Halász, Attila G. Császár, Lorenz S. Cederbaum, Ágnes Vibók

Keywords: light induced conical intersection, sodium dimer, LICI

Abstract: Rovibronic spectra of the field-dressed homonuclear diatomic Na₂ molecule are investigated to identify direct signatures of the light-induced conical intersection (LICI) on the spectrum. The theoretical framework formulated allows the computation of the (1) field-dressed rovibronic states induced by a medium-intensity continuous-wave laser light and the (2) transition amplitudes between these field-dressed states with respect to an additional weak probe pulse. The field-dressed spectrum features absorption peaks resembling the field-free spectrum as well as stimulated emission peaks corresponding to transitions not visible in the field-free case. By investigating the dependence of the field-dressed spectra on the dressing-field wavelength, in both full- and reduced-dimensional simulations, direct signatures of the LICI can be identified. These signatures include (1) the appearance of new peaks and the splitting of peaks for both absorption and stimulated emission and (2) the manifestat

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Journal: Journal of Physics B: Atomic, Molecular and Optical Physics

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Publication Location:

Article Title: Neon in ultrashort and intense x-rays from free electron lasers

Authors: Christian Buth, Randolph Beerwerth, Razib Obaid, Nora Berrah, Lorenz S Cederbaum, Stephan Fritzsche

Keywords: neon atom, ultrashort and intense x-rays, free-electron laser, ion yield, photon yield, rate equations, multiconfiguration Dirac–Hartree–Fock

Abstract: We theoretically examine neon atoms in ultrashort and intense x-rays from free electron lasers and compare our results with data from experiments conducted at the Linac Coherent Light Source. For this purpose, we treat in detail the electronic structure in all possible non-relativistic cationic configurations using a relativistic multiconfiguration approach. The interaction with the x-rays is described in rate-equation approximation. To understand the mechanisms of the interaction, a path analysis is devised which allows us to investigate what sequences of photoionization and decay processes lead to a specific configuration and with what probability. Thereby, we uncover a connection to the mathematics of graph theory and formal languages. In detail, we study the ion yields and find that plain rate equations do not provide a satisfactory description. We need to extend the rate equations for neon to incorporate double Auger decay of a K-shell vacancy and photoionization shake off for neon.

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Journal: Chemical Physics

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Publication Identifier: 10.1016/j.chemphys.2017.10.004

Volume: 509

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Date Submitted: 6/27/18 12:00AM

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Publication Location:

Article Title: Dynamic interference in the resonance-enhanced multiphoton ionization of hydrogen atoms by short and intense laser pulses

Authors: Anne D. Müller, Eric Kutscher, Anton N. Artemyev, Lorenz S. Cederbaum, Philipp V. Demekhin

Keywords: Spectra induced by intense laser pulses, Multiphoton ionization and excitation, Photoinduced electron dynamics

Abstract: Photoionization of the hydrogen atom by intense and short coherent laser pulses is investigated from first principles by a numerical solution of the time-dependent Schrödinger equation in the dipole-velocity gauge. The considered photon energies are resonant to the $1s \rightarrow 2p$ excitation, and the pulse intensities are high enough to induce Rabi floppings. The computed resonance-enhanced two-photon ionization spectra as well as the three-photon above threshold ionization spectra exhibit pronounced multiple-peak patterns due to dynamic interference. Fingerprints of dynamic interference can also be seen directly in the radial density of the photoelectron. The impact of the variation of the pulse intensity and photon energy on the dynamic interference is investigated, and the angular distribution of the emitted electrons is analyzed in some details. The present precise numerical results confirm our previous theoretical predictions on the two-photon ionization spectra (Demekhin and Cederbaum, 2017).

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Journal: Physical Review A

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Date Submitted: 6/27/18 12:00AM

Date Published: 9/18/17 2:00PM

Publication Location:

Article Title: Impact of intense laser pulses on the autoionization dynamics of the 2s2p doubly excited state of He

Authors: Anton N. Artemyev, Lorenz S. Cederbaum, Philipp V. Demekhin

Keywords: photoionization, helium, Fano resonance, autoionization

Abstract: The photoionization of a helium atom by short intense laser pulses is studied theoretically in the vicinity of the 2s2p 1P doubly excited state with the intention to investigate the impact of the intensity and duration of the exciting pulse on the dynamics of the autoionization process. For that purpose, we solve numerically the corresponding time-dependent Schrödinger equation by applying the time-dependent restricted-active-space configuration-interaction method (TD-RASCI). The present numerical results clearly demonstrate that the Fano interferences can be controlled by a single high-frequency pulse. As long as the pulse duration is comparable to the autoionization lifetime, varying the peak intensity of the pulse enables manipulation of the underlying Fano interference. In particular, the asymmetric profile observed for the 2s2p 1P doubly excited state of He in the weak-field ionization can be smoothly transformed to a window-type interference profile.

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Date Submitted: 7/18/19 12:00AM

Date Published: 11/17/18 4:00PM

Publication Location:

Article Title: Charge migration in propiolic acid: A full quantum dynamical study

Authors: V. Despré, N. V. Golubev, and A. I. Kuleff

Keywords: charge migration, propionic acid, ultrafast

Abstract: Ionization of molecules very often populates several cationic states launching pure electron dynamics that appear as ultrafast migration of the hole charge throughout the system. A crucial question in the emerging field of attochemistry is whether these pure electronic coherences last long enough to allow for their efficient observation and eventual manipulation with ultrashort laser pulses. We report a full dimensional quantum calculation of concerted electron-nuclear dynamics initiated by outer-valence ionization of propiolic acid molecule, showing that the charge will oscillate between the carbon triple bond and the carbonyl oxygen for more than 10 fs before getting trapped by the nuclear motion. This time is enough for the charge migration to be observed and controlled. We argue that the molecule is very suitable for experimental studies.

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Publication Location:

Article Title: Communication: Substantial impact of the orientation of transition dipole moments on the dynamics of diatomics in laser fields

Authors: Péter Badankó, Gábor J. Halász, Lorenz S. Cederbaum, Ágnes Vibók, András Csehi

Keywords: light induced conical intersection, LICI, MgH ion

Abstract: The formation of light-induced conical intersections (LICIs) between electronic states of diatomic molecules has been thoroughly investigated over the past decade. In the case of running laserwaves, the rotational, vibrational, and electronic motions couple via the LICI giving rise to strong nonadiabatic phenomena. In contrast to natural conical intersections (CIs) which are given by nature and hard to manipulate, the characteristics of LICIs are easily modified by the parameters of the laser field. The internuclear position of the created LICI is determined by the laser energy, while the angular position is given by the orientation of the transition dipole moment (TDM) with respect to the molecular axis. In the present communication, using MgH⁺ as a showcase example, we exploit the strong impact of the orientation of the TDMs exerted on the light-induced nonadiabatic dynamics.

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Date Submitted: 11/9/20 12:00AM

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Publication Location:

Article Title: Size effects in charge migration in alkyne chains

Authors: Victor Despré, Alexander I. Kuleff

Keywords: Charge migration, Electronic correlation, Size effects, Attosecond science

Abstract: The charge migration dynamics initiated by an outer-valence ionization of alkyne chains containing between 4 and 12 carbon atoms is studied using ab initio methods only. It is shown that the removal of a HOMO –1 electron from each of the molecules in the series leads to the population of the same structure of a main state and correlation satellites in their ionization spectra. The resulting pure electron dynamics manifest as an ultrafast charge migration oscillations between the center of the molecule and its extremities. The charge migration follows the same pattern in all studied cases with a slight monotonic increase of the time scale with the system size. This shows that the correlation-driven charge migration can exhibit scaling properties and constitutes a proof of concept for the possibility of molecular design for charge migration. As alkynes are used as molecular wires, it is argued that the present results may open the door for ultrafast molecular electronics.

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Publication Identifier Type: DOI **Publication Identifier:** 10.1063/1.5085011
Volume: 6 **Issue:** 1 **First Page #:** 014101
Date Submitted: 6/14/19 12:00AM **Date Published:** 1/1/19 4:00PM
Publication Location:

Article Title: Ab initio investigation of Br-3d core-excited states in HBr and HBr ion toward XUV probing of photochemical dynamics

Authors: Yuki Kobayashi, Tao Zeng, Daniel M. Neumark, Stephen R. Leone

Keywords: HBr, theory, transition probability, attosecond

Abstract: Ultrafast X-ray/XUV transient absorption spectroscopy is a powerful tool for real-time probing of chemical dynamics. Interpretation of the transient absorption spectra requires knowledge of core-excited potentials, which necessitates assistance from high-level electronic-structure computations. In this study, we investigate Br-3d core-excited electronic structures of hydrogen bromide (HBr) using spin-orbit general multiconfigurational quasidegenerate perturbation theory (SO-GMC-QDPT). Potential energy curves and transition dipole moments are calculated from the Franck-Condon region to the asymptotic limit and used to construct core-to-valence absorption strengths for five electronic states of HBr (1R0p ; 3P1; 1P1; 3P0p ; 3R1) and two electronic states of HBrp (2P3=2, 2R1=2). The results illustrate the capabilities of Br-3d edge probing to capture transitions of the electronic-state symmetry as well as nonadiabatic dissociation processes that evolve across avoided crossings.

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Journal: Science
Publication Identifier Type: DOI **Publication Identifier:** 10.1126/science.aax0076
Volume: 365 **Issue:** 6448 **First Page #:** 79
Date Submitted: 8/24/19 12:00AM **Date Published:** 7/5/19 2:00PM
Publication Location:

Article Title: Direct mapping of curve-crossing dynamics in IBr by attosecond transient absorption spectroscopy

Authors: Y. Kobayashi, K. F. Chang, T. Zeng, D. M. Neumark, S. R. Leone

Keywords: iodine monobromide, attosecond, curve crossing

Abstract: The electronic character of photoexcited molecules can abruptly change at avoided crossings and conical intersections. Here, we report direct mapping of the coupled interplay between electrons and nuclei in a prototype molecule, iodine monobromide (IBr), using attosecond transient absorption spectroscopy. A few-femtosecond visible pulse resonantly excites the B(3|0+) state of IBr and the accompanying photodissociation dynamics are tracked by an attosecond extreme-ultraviolet pulse that simultaneously probes the I-4d and Br-3d corelevel absorption edges. Direct comparison with quantum mechanical simulations unambiguously identifies the core-level absorption features associated with adiabatic and diabatic channels at the B/Y avoided crossing and concurrent two-photon dissociation processes that involve the Y/Z avoided crossing. The results show clear evidence for rapid switching of valence molecular orbital occupations at the avoided crossing.

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Date Submitted: 7/18/19 12:00AM

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Publication Location:

Article Title: Disentangling conical intersection and coherent molecular dynamics in methyl bromide with attosecond transient absorption spectroscopy

Authors: H. Timmers, X. Zhu, Z. Li, Y. Kobayashi, M. Sabbar, M. Hollstein, M. Reduzzi, T. J. Martinez, D. M. Neu

Keywords: methyl bromide, conical intersection, coherence

Abstract: Attosecond probing of core-level electronic transitions provides a sensitive tool for studying valence molecular dynamics with atomic, state, and charge specificity. In this report, we employ attosecond transient absorption spectroscopy to follow the valence dynamics of strong-field initiated processes in methyl bromide. By probing the 3d core-to-valence transition, we resolve the strong field excitation and ensuing fragmentation of the neutral π^* excited states of methyl bromide. The results provide a clear signature of the non-adiabatic passage of the excited state wavepacket through a conical intersection. We additionally observe competing, strong field initiated processes arising in both the ground state and ionized molecule corresponding to vibrational and spin-orbit motion, respectively. The demonstrated ability to resolve simultaneous dynamics with few-femtosecond resolution presents a clear path forward in the implementation of attosecond XUV spectroscopy as a general tool for

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Volume: 377

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Date Submitted: 6/14/19 12:00AM

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Publication Location:

Article Title: Transient absorption spectroscopy using high harmonic generation: a review of ultrafast X-ray dynamics in molecules and solids

Authors: Romain Geneaux, Hugo J. B. Marroux, Alexander Guggenmos, Daniel M. Neumark, Stephen R. Leone

Keywords: ultrafast dynamics, transient absorption, X-rays

Abstract: Attosecond science opened the door to observing nuclear and electronic dynamics in real time and has begun to expand beyond its traditional grounds. Among several spectroscopic techniques, X-ray transient absorption spectroscopy has become key in understanding matter on ultrafast time scales. In this review, we illustrate the capabilities of this unique tool through a number of iconic experiments. We outline how coherent broadband X-ray radiation, emitted in high-harmonic generation, can be used to follow dynamics in increasingly complex systems. Experiments performed in both molecules and solids are discussed at length, on time scales ranging from attoseconds to picoseconds, and in perturbative or strong-field excitation regimes. This article is part of the theme issue 'Measurement of ultrafast electronic and structural dynamics with X-rays'.

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Date Submitted: 6/14/19 12:00AM

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Publication Location:

Article Title: Hot phonon and carrier relaxation in Si(100) determined by transient extreme ultraviolet spectroscopy

Authors: Scott K. Cushing, Michael Zürch, Peter M. Kraus, Lucas M. Carneiro, Angela Lee, Hung-Tzu Chang, Ch

Keywords: silicon, transient absorption, phonon

Abstract: The thermalization of hot carriers and phonons gives direct insight into the scattering processes that mediate electrical and thermal transport. Obtaining the scattering rates for both hot carriers and phonons currently requires multiple measurements with incommensurate timescales. Here, transient extreme-ultraviolet (XUV) spectroscopy on the silicon 2p core level at 100 eV is used to measure hot carrier and phonon thermalization in Si (100) from tens of femtoseconds to 200 ps, following photoexcitation of the indirect transition to the D valley at 800 nm. The ground state XUV spectrum is first theoretically predicted using a combination of a single plasmon pole model and the Bethe-Salpeter equation with density functional theory. The excited state spectrum is predicted by incorporating the electronic effects of photo-induced state-filling, broadening, and band-gap renormalization into the ground state XUV spectrum. A time-dependent lattice deformation and expansion is also required to

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Date Submitted: 11/9/20 12:00AM

Date Published: 7/15/19 7:00AM

Publication Location:

Article Title: Enhancing keV high harmonic signals generated by long-wave infrared lasers

Authors: ZENGHU CHANG

Keywords: mid infrared laser, attosecond, keV photons

Abstract: A multielectron wave-packet propagation method was used to calculate the electronic decay of oxygen and fluorine 2s vacancies for a group of trifluoroalkyl alcohols, $\text{HOCnH}(2n+1)\text{F}_3$, with n between 1 and 5. Whether ionizing O2s or F2s orbitals, it is shown that an electron can be emitted non-locally from the opposite terminus of the molecule. The decay of the $\text{O}(2s+1)$ state is found to be about 2–3 times faster than that of the $\text{F}(2s+1)$, but in both cases the process takes only a few femtoseconds, demonstrating a highly efficient energy transfer through the carbon bridge. A comparison to the previously reported non-local decay in linear difluorocumulenone systems shows that the non-linearity of the trifluoroalkyl alcohols does not appear to dramatically influence the decay efficiency. These results shed light onto the nature of the scaling of electron correlation and open the door to the potential design of molecules that take advantage of this mechanism.

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Journal: Optical Society of America

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Date Submitted: 11/9/20 12:00AM

Date Published: 12/20/19 8:00AM

Publication Location:

Article Title: Real-time observation of electronic, vibrational, and rotational dynamics in nitric oxide with attosecond soft x-ray pulses at 400 eV

Authors: Nariyuki Saito, Hiroki Sannohe, Nobuhisa Ishii, Teruto Kanai, Nobuhiro Kosugi, Yi Wu, Andrew Chew, S

Keywords: Pump-probe spectroscopy using high harmonics, transient absorption spectroscopy, molecular dynamics

Abstract: Photoinduced quantum dynamics in molecules have hierarchical temporal structures with different energy scales that are associated with electron and nuclear motions. Femtosecond-to-attosecond transient absorption spectroscopy (TAS) using high-harmonic generation (HHG) with a photon energy below 300 eV has been a powerful tool to observe such electron and nuclear dynamics in a table-top manner. However, comprehensive measurements of the electronic, vibrational, and rotational molecular dynamics have not yet been achieved. Here we demonstrate HHG-based TAS at the nitrogen K-edge (400 eV) for the first time to our knowledge and observe all the electronic, vibrational, and rotational degrees of freedom in a nitric oxide molecule at attosecond to sub- picosecond time scales. This method of employing core-to-valence transitions offers an all-optical approach to reveal the complete molecular dynamics in photochemical reactions with element and electronic state specificity.

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Publication Location:

Article Title: Compensating chirp of attosecond X-ray pulses by a neutral hydrogen gas

Authors: Zenghu Chang

Keywords: chirp pulse compensation, hydrogen, attosecond

Abstract: It is demonstrated by numerical simulations that the attosecond chirp of high order harmonic pulses in the 530 to 1000 eV range can be partially compensated by the negative group delay dispersion of unionized molecular hydrogen gas. The transmission of X-rays through gas with the required pressure-length product for optimum chirp compensation is higher than 10% in the photon energy range that covers the K-edge of oxygen and L-edges of iron, cobalt, and nickel.

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Date Published: 2/2/19 12:00AM

Publication Location:

Article Title: Attosecond streaking phase retrieval with neural network

Authors: Jonathon White, Zenghu Chang

Keywords: neural network, attosecond, pulse measurement

Abstract: A new method for retrieving the spectral phase of isolated attosecond X-ray pulses from streaking traces is explored. The neural network method shows the potential for nearly instantaneous attosecond streaking phase retrieval, without use of the central momentum approximation. A neural network is trained with computer generated data that contain statistical noise and shown to correctly retrieve the phase of both computer generated and experimental attosecond streaking traces.

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First Page #: 33238

Date Submitted: 6/14/19 12:00AM

Date Published: 12/1/18 4:00PM

Publication Location:

Article Title: Attosecond chirp compensation in water window by plasma dispersion

Authors: Zenghu Chang

Keywords: chirp compensataion, water window, x-ray, plasma

Abstract: Compensating attosecond chirp (atto-chirp) of broadband high-order harmonic pulses in the water window region (282 to 533 eV) is a major challenge, due to the lack of natural materials that exhibit negative group velocity dispersion and low loss. Analysis shows that the amount of dispersion of fully ionized hydrogen plasma with suitable density-length product is sufficient to compensate the chirp of attosecond pulses with center photon energy above 300 eV. This is confirmed by numerical simulations based on the Strong Field Approximation.

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Publication Location:

Article Title: Ultrafast photodissociation dynamics and nonadiabatic coupling between excited electronic states of methanol probed by time-resolved photoelectron spectroscopy

Authors: Elio G. Champenois, Loren Greenman, Niranjana Shivaram, James P. Cryan, Kirk A. Larsen, Thomas N.

Keywords: attosecond, methanol, theory, photoelectron

Abstract: The electronic and nuclear dynamics in methanol, following 156 nm photoexcitation, are investigated by combining a detailed analysis of time-resolved photoelectron spectroscopy experiments with electronic structure calculations. The photoexcitation pump pulse is followed by a delayed 260 nm photoionization probe pulse to produce photoelectrons that are analyzed by velocity map imaging. The yields of mass-resolved ions, measured with similar experimental conditions, are found to exhibit the same time-dependence as specific photoelectron spectral features. Energy-resolved signal onset and decay times are extracted from the measured photoelectron spectra to achieve high temporal resolution, beyond the 20 fs pump and probe pulse durations. When combined with ab initio calculations of selected cuts through the excited state potential energy surfaces, this information allows the dynamics of the transient excited molecule, which exhibits multiple nuclear and electronic degrees of freedom, to

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Date Submitted: 7/18/19 12:00AM

Date Published: 6/6/19 7:00AM

Publication Location:

Article Title: Ultrafast Rydberg State Dissociation in Oxygen: Identifying the Role of Multielectron Excitations

Authors: Alexander Plunkett, Nathan Harkema, Robert R. Lucchese, C. William McCurdy and Arvinder Sandhu

Keywords: oxygen, attosecond, transient absorption

Abstract: We investigated the fragmentation dynamics of highly excited states of molecular oxygen using femtosecond transient photoelectron spectroscopy. An extreme ultraviolet (XUV) pulse populates the autoionizing Rydberg series converging to $O_2^+ c 4\pi^* u$, and a femtosecond near-infrared (IR) pulse was used to photoionize these states as they dissociate. Monitoring the differential photoelectron spectra as a function of time delay allowed us to obtain the relaxation lifetimes of these Rydberg states. We observed a photoelectron signal corresponding to the formation of a 4p excited atomic oxygen fragment, which is not an expected dissociation product of the $(O_2^+ c 4\pi^* u)nl^*g$ Rydberg series. Analysis of the time-dependent photoelectron spectra and photoionization calculations indicate that this fragment results from a previously unexplored $(O_2^+ 4f^1g)4p$ repulsive state and that, contrary to expectations, this multielectron excitation pathway presents a substantial cross section.

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Article Title: Non-Born-Oppenheimer electronic wave packet in molecular nitrogen at 14 eV probed by time-resolved photoelectron spectroscopy

Authors: Claude Marceau, Varun Makhija, Peng Peng, Marius Hervé, P. B. Corkum, A. Yu. Naumov, A. Stolow, E

Keywords: nitrogen, laser, attosecond, electronic wave packet

Abstract: We report the observation of a 4.1 ± 0.2 THz quantum beat in the photoelectron spectrum of molecular nitrogen following coherent optical excitation by 14 eV photons and probed by two photons at 800 nm. The intermediate states in the two pathway interference are the valence state $b_1\ 1\pi + u\ v = 13$ and Rydberg state $c_1\ 4\ 1\pi + u\ v = 4$. The amplitude of the coherent oscillations decays in about 3 ps and does not revive. Opposite phases in the photoelectron yield corresponding to different vibrational levels of the final cationic states $X2\pi + g\ v = 0$, $A\ 2\pi + u\ v = 1$, and $X2\pi + g\ v = 2, 3, 4$ are reported. Simulation results show that the interference originates from the strongly mixed character of the two simultaneously excited eigenstates, causing large population oscillations in the zeroth-order Born-Oppenheimer diabatic basis. No quantum beat was observed when probing with single photon at 400 nm, suggesting a resonance with a state just below the ionization potential which acts as a filter,

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Publication Type: Journal Article

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Journal: Phys. Rev. A.

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Volume: 100

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Date Submitted: 7/17/19 12:00AM

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Publication Location:

Article Title: Short- and long-term gain dynamics in N₂⁺ air lasing

Authors: M Britton, M. Lytova, P. Laferriere, P. Peng, F. Morales, D.H. Ko, M. Richter, P. Polynkin, D. M. Villeneuve

Keywords: air lasing, nitrogen, attosecond

Abstract: Air lasing in the nitrogen molecular ion is not well understood because the complex physics responsible for gain is interwoven with pulse propagation in an extreme environment. Here we use a short gas jet to limit the interaction length, thereby removing the propagation effects. We report on several mechanisms that contribute to the decay of gain in different conditions, and experimentally isolate two decay timescales: the decay of long-term gain due to collisional state mixing, and short-term gain that cannot be explained by population inversion. To test the former, we control the inelastic electron scattering rate by varying the gas concentration while keeping the propagation length fixed, and predict the change of the decay using a model of collisional state mixing. We show that the same mechanism causes the decay of rotational wave packets in the states of the ion. Finally, we simulate the complex modulations of gain due to rotational wave packets and the propagation of the probe pu

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Journal: Phys. Rev. A
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevA.99.063403
Volume: 99 **Issue:** **First Page #:** 063403
Date Submitted: 7/17/19 12:00AM **Date Published:** 6/6/19 7:00AM
Publication Location:

Article Title: Ultrafast Rydberg State Dissociation in Oxygen: Identifying the Role of Multielectron Excitations

Authors: Alex Plunkett, Nathan Harkema, Robert R. Lucchese, C. William McCurdy, and Arvinder Sandhu

Keywords: oxygen, attosecond, transient absorption

Abstract: We investigated the fragmentation dynamics of highly excited states of molecular oxygen using femtosecond transient photoelectron spectroscopy. An extreme ultraviolet (XUV) pulse populates the autoionizing Rydberg series converging to $O_2^+ c\ 4\sigma_u$, and a femtosecond near-infrared (IR) pulse was used to photoionize these states as they dissociate. Monitoring the differential photoelectron spectra as a function of time delay allowed us to obtain the relaxation lifetimes of these Rydberg states. We observed a photoelectron signal corresponding to the formation of a 4p excited atomic oxygen fragment, which is not an expected dissociation product of the $(O_2^+ c\ 4\sigma_u) n\pi_g$ Rydberg series. Analysis of the time-dependent photoelectron spectra and photoionization calculations indicate that this fragment results from a previously unexplored $(O_2^+ 4\sigma_g) 4p$ repulsive state and that, contrary to expectations, this multielectron excitation pathway presents a substantial cross section. Our study demonstrate

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Phys. Rev. Lett
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevLett.120.233201
Volume: 120 **Issue:** **First Page #:** 233201
Date Submitted: 11/9/20 12:00AM **Date Published:**
Publication Location:

Article Title: Selectivity of Electronic Coherence and Attosecond Ionization Delays in Strong-Field Double Ionization

Authors: Yuki Kobayashi, Maurizio Reduzzi, Kristina F. Chang, Henry Timmers, Daniel M. Neumark, and Stephen

Keywords: coherent electron dynamics, attosecond transient absorption spectroscopy

Abstract: Experiments are presented on real-time probing of coherent electron dynamics in xenon initiated by strong-field double ionization. Attosecond transient absorption measurements allow for characterization of electronic coherences as well as relative ionization timings in multiple electronic states of Xep and $Xe2p$. A high degree of coherence $g \approx 0.4$ is observed between $3P_{02}$ - $3P_{00}$ of $Xe2p$, whereas for other possible pairs of states the coherences are below the detection limits of the experiments. A comparison of the experimental results with numerical simulations based on an uncorrelated electron-emission model shows that the coherences produced by strong-field double ionization are more selective than predicted. Surprisingly short ionization time delays, 0.85 fs, 0.64 fs, and 0.75 fs relative to Xep formation, are also measured for the $3P_2$, $3P_0$, and $3P_1$ states of $Xe2p$, respectively. Both the unpredicted selectivity in the formation of coherence and the subfemtosecond time delays of specif

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Journal: Nature Communications

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Publication Identifier: 10.1038/s41467-020-17745-w

Volume: 11

Issue: 1

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Date Submitted: 11/8/20 12:00AM

Date Published: 8/1/20 7:00AM

Publication Location:

Article Title: Revealing electronic state-switching at conical intersections in alkyl iodides by ultrafast XUV transient absorption spectroscopy

Authors: Kristina F. Chang, Maurizio Reduzzi, Han Wang, Sonia M. Poullain, Yuki Kobayashi, Lou Barreau, David

Keywords: extreme ultraviolet, conical intersections, transient absorption, spectroscopy

Abstract: Conical intersections between electronic states often dictate the chemistry of photoexcited molecules. Recently developed sources of ultrashort extreme ultraviolet (XUV) pulses tuned to element-specific transitions in molecules allow for the unambiguous detection of electronic state-switching at a conical intersection. Here, the fragmentation of photoexcited iso-propyl iodide and tert-butyl iodide molecules (i-C₃H₇I and t-C₄H₉I) through a conical intersection between 3Q0/1Q1 spin-orbit states is revealed by ultrafast XUV transient absorption measuring iodine 4d core-to-valence transitions. The electronic state-sensitivity of the technique allows for a complete mapping of molecular dissociation from photoexcitation to photoproducts. In both molecules, the sub-100 fs transfer of a photoexcited wave packet from the 3Q0 state into the 1Q1 state at the conical intersection is captured. The results show how differences in the electronic state-switching of the wave packet in i-C₃H₇I and t-C₄H₉I

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Journal: Phys. Rev. A

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Volume: 102

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Date Submitted: 6/13/21 12:00AM

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Publication Location:

Article Title: Attosecond XUV probing of vibronic quantum superpositions in Br₂⁺

Authors: Yuki Kobayashi, Daniel M. Neumark, and Stephen R. Leone

Keywords: Ultrafast, Attosecond, XUV, Quantum, Superpositions

Abstract: Ultrafast laser excitation can create coherent superpositions of electronic states in molecules and trigger ultrafast flow of electron density on few-to sub-femtosecond time scales. While recent attosecond experiments have addressed real-time observation of these primary photochemical processes, the underlying roles of simultaneous nuclear motions and how they modify and disturb the valence electronic motion remain uncertain. Here, we investigate coherent electronic-vibrational dynamics induced among multiple vibronic levels of ionic bromine (Br₂⁺), including both spin-orbit and valence electronic superpositions, using attosecond transient absorption spectroscopy. Decay, revival, and apparent frequency shifts of electronic coherences are measured via characteristic quantum beats on the Br-3d core-level absorption signals. Quantum-mechanical simulations attribute the observed electronic decoherence to broadened phase distributions of nuclear wave packets on anharmonic potentials.

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Journal: Physical Review A

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Volume: 101

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Date Submitted: 11/8/20 12:00AM

Date Published: 6/1/20 7:00AM

Publication Location:

Article Title: Coherent electronic-vibrational dynamics in deuterium bromide probed via attosecond transient-absorption spectroscopy

Authors: Yuki Kobayashi, Kristina F. Chang, Sonia Marggi Poullain, Valeriu Scutelnic, Tao Zeng, Daniel M. Neumark

Keywords: Ultrafast, Attosecond, Coherent Superpositions, Spectroscopy

Abstract: Ultrafast laser excitation can create coherent superpositions of electronic states in molecules and trigger ultrafast flow of electron density on few-to sub-femtosecond time scales. While recent attosecond experiments have addressed real-time observation of these primary photochemical processes, the underlying roles of simultaneous nuclear motions and how they modify and disturb the valence electronic motion remain uncertain. Here, we investigate coherent electronic-vibrational dynamics induced among multiple vibronic levels of ionic bromine (Br^+), including both spin-orbit and valence electronic superpositions, using attosecond transient absorption spectroscopy. Decay, revival, and apparent frequency shifts of electronic coherences are measured via characteristic quantum beats on the Br-3d core-level absorption signals. Quantum-mechanical simulations attribute the observed electronic decoherence to broadened phase distributions of nuclear wave packets on anharmonic potentials.

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Journal: The Journal of Chemical Physics

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Volume: 151

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Date Submitted: 11/8/20 12:00AM

Date Published: 11/1/19 7:00AM

Publication Location:

Article Title: NaI revisited: Theoretical investigation of predissociation via ultrafast XUV transient absorption spectroscopy

Authors: Yuki Kobayashi, Tao Zeng, Daniel M. Neumark, Stephen R. Leone

Keywords: Ultrafast XUV, spectroscopy, predissociation dynamics

Abstract: Avoided crossings can trigger abrupt changes of electronic character and redirect the outcomes of photochemical reactions. We report a theoretical investigation into core-level spectroscopic probing of predissociation dynamics of sodium iodide (NaI), a prototype system for studies of avoided crossing dynamics. The elegant femtochemistry work of Zewail and co-workers pioneered the real-time dynamics of NaI, detecting the Na atoms bursting forth from the avoided crossing and the residual NaI molecules oscillating inside the quasibound potential. The simulated results show that core-level spectroscopy not only observes these integrated outcomes but also provides a direct measure of the abrupt switching of electronic character at the avoided crossing. The valence and core-excited electronic structures of NaI are computed by spin-orbit general multiconfigurational quasidegenerate perturbation theory, from which core-level absorption spectra of the predissociation dynamics are constructed.

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Volume: 14

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Date Submitted: 6/13/21 12:00AM

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Publication Location:

Article Title: Simultaneous Observation of Carrier-Specific Redistribution and Coherent Lattice Dynamics in 2H-MoTe

Authors: Andrew R. Attar, Hung-Tzu Chang, Alexander Britz, Xiang Zhang, Ming-Fu Lin, Aravind Krishnamoorthy

Keywords: femtosecond, XUV, transient absorption, structural dynamics

Abstract: We employ few-femtosecond extreme ultraviolet (XUV) transient absorption spectroscopy to reveal simultaneously the intra- and interband carrier relaxation and the light-induced structural dynamics in nanoscale thin films of layered 2H-MoTe₂ semiconductor. By interrogating the valence electronic structure via localized Te 4d (39-46 eV) and Mo 4p (35-38 eV) core levels, the relaxation of the photoexcited hole distribution is directly observed in real time. We obtain hole thermalization and cooling times of 15 ± 5 fs and 380 ± 90 fs, respectively, and an electron-hole recombination time of 1.5 ± 0.1 ps. Furthermore, excitations of coherent out-of-plane A_{1g} (5.1 THz) and in-plane E_{1g} (3.7 THz) lattice vibrations are visualized through oscillations in the XUV absorption spectra. By comparison to Bethe-Salpeter equation simulations, the spectral changes are mapped to real-space excited-state displacements of the lattice along the dominant A_{1g} coordinate.

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Journal: Condensed Matter Materials Science

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Volume: 103

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Publication Location:

Article Title: Electron Thermalization and Relaxation in Laser-Heated Nickel by Few-Femtosecond Core-Level Transient Absorption Spectroscopy

Authors: Hung-Tzu Chang,^{1, ?} Alexander Guggenmos,^{1, †} Scott K. Cushing,^{1, 2, ‡} Yang Cui,^{3, 4, §} Naseem Ud

Keywords: carrier dynamics, few-femtosecond, XUV

Abstract: Direct measurements of photoexcited carrier dynamics in nickel are made using few-femtosecond extreme ultraviolet (XUV) transient absorption spectroscopy at the nickel M_{2,3} edge. It is observed that the core-level absorption lineshape of photoexcited nickel can be described by a Gaussian broadening ($\leftarrow$) and a red shift ($\rightarrow$) of the ground state absorption spectrum. Theory predicts, and the experimental results verify that, after initial rapid carrier thermalization, the electron temperature increase ($\uparrow T$) is linearly proportional to the Gaussian broadening factor $\leftarrow$, providing quantitative real-time tracking of the relaxation of the electron temperature. Measurements reveal an electron cooling time for 50 nm thick polycrystalline nickel films of 640 ± 80 fs. With hot thermalized carriers, the spectral red shift exhibits a power-law relationship with the change in electron temperature of $\rightarrow \propto \uparrow T^{1.5}$. Rapid electron thermalization via carrier-carrier scattering accompanies and follows the non

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Journal: Physical Review Letters

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Publication Identifier: 10.1103/PhysRevLett.124.207401

Volume: 124 Issue: 20

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Date Submitted: 11/8/20 12:00AM

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Publication Location:

Article Title: Attosecond Time-Domain Measurement of Core-Level-Exciton Decay in Magnesium Oxide

Authors: Romain Geneaux, Christopher J. Kaplan, Lun Yue, Andrew D. Ross, Jens E. Bækhoj, Peter M. Kraus, F

Keywords: Attosecond, magnesium oxide, ultrafast dynamic, spectroscopy

Abstract: Excitation of ionic solids with extreme ultraviolet pulses creates localized core-level excitons, which in some cases couple strongly to the lattice. Here, core-level-exciton states of magnesium oxide are studied in the time domain at the Mg L_{2,3} edge with attosecond transient reflectivity spectroscopy. Attosecond pulses trigger the excitation of these short-lived quasiparticles, whose decay is perturbed by time-delayed near-infrared pulses. Combined with a few-state theoretical model, this reveals that the infrared pulse shifts the energy of bright (dipole-allowed) core-level-exciton states as well as induces features arising from dark core-level excitons. We report coherence lifetimes for the two lowest core-level excitons of 2.3 ± 0.2 and 1.6 ± 0.5 fs and show that these are primarily a consequence of strong exciton-phonon coupling, disclosing the drastic influence of structural effects in this ultrafast relaxation process.

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Journal: Applied Physics Letters

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Date Submitted: 11/9/20 12:00AM

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Publication Location:

Article Title: Ho:YLF amplifier with Ti:Sapphire frontend for pumping mid-infrared optical parametric amplifier

Authors: Krishna Murari, Fangjie Zhou, Yanchun Yin, Yi Wu, Bruce Weaver, Timur Avni, Esben Larsen, Zenghu C

Keywords: Amplification, high harmonic, Ti:Sapphire, infrared

Abstract: We present a Ho:YLF Chirped-Pulse Amplification laser for pumping a longwave infrared Optical Parametric Chirped Pulse Amplifier at a 1 kHz repetition rate. By utilizing a Ti:Sapphire laser as a frontend, 5-lJ seed pulses at 2051nm laser pulse are generated in a Dual-Chirp Optical Parametric Amplifier, which are amplified to 28 mJ pulses with a pulse duration of 5.6 ps. The scheme offers a potential driver for two-color (800nm and 8 lm) high harmonic generation with an increased keV x-ray photon flux.

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Journal: Advances in Atomic, Molecular, and Optical Physics

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Volume: 69

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Date Submitted: 11/9/20 12:00AM

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Publication Location:

Article Title: Tabletop attosecond X-rays in the water window

Authors: Seunghwoi Han, Jie Li, Zheyuan Zhu, Andrew Chew, Esben W. Larsen, Yi Wu, Shuo Sean Pang, and Z

Keywords: attosecond, electron dynamics, infrared, spectroscopy

Abstract: In recent years, a new generation of attosecond driving lasers centered at $\sim 1.7 \mu\text{m}$ based on carrier-envelope phase stabilized Optical Parametric Amplification have enabled the generation of X-ray pulses reaching the water window (282–533 eV), which is of great interest given their applications in the study of electron dynamics in atoms, molecules and condensed matter containing carbon, nitrogen, oxygen and other important elements. Various gating methods emanated isolated attosecond pulses for pump-probe experiments. Atto-chirp compensation has made the X-ray pulses at the carbon K-edge as short as 53 attoseconds. Novel spectral phase retrieval schemes such as neural network have been implemented to attosecond streaking techniques for faster and more reliable characterization of X-ray pulses. The water window X-ray sources have been applied to ultrafast measurements, such as attosecond transient absorption spectroscopy and photoelectron spectroscopy with element specificity and sub-op

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Date Submitted: 11/9/20 12:00AM

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Publication Location:

Article Title: Attosecond science based on high harmonic generation from gases and solids

Authors: Jie Li, Jian Lu, Andrew Chew, Seunghwoi Han, Jialin Li, Yi Wu, He Wang, Shambhu Ghimire, Zenghu C

Keywords: ultrafast, infrared, high harmonic, attosecond

Abstract: Recent progress in high power ultrafast short-wave and mid-wave infrared lasers has enabled gas-phase high harmonic generation (HHG) in the water window and beyond, as well as the demonstration of HHG in condensed matter. In this Perspective, we discuss the recent advancements and future trends in generating and characterizing soft X-ray pulses from gasphase HHG and extreme ultraviolet (XUV) pulses from solid-state HHG. Then, we discuss their current and potential usage in time-resolved study of electron and nuclear dynamics in atomic, molecular and condensed matters.

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Journal: Scientific Reports
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Volume: 10 **Issue:** 1 **First Page #:**
Date Submitted: 11/9/20 12:00AM **Date Published:** 5/1/20 7:00AM
Publication Location:

Article Title: Generation of few-cycle multimillijoule 2.5 μ m pulses from a single-stage Cr²⁺:ZnSe amplifier

Authors: Yi Wu, Fangjie Zhou, Esben W. Larsen, Fengjiang Zhuang, Yanchun Yin, Zenghu Chang

Keywords: high-order harmonic, ultrafast, extreme ultraviolet (XUV), attosecond

Abstract: Recent progress in high power ultrafast short-wave and mid-wave infrared lasers has enabled gas-phase high harmonic generation (HHG) in the water window and beyond, as well as the demonstration of HHG in condensed matter. In this Perspective, we discuss the recent advancements and future trends in generating and characterizing soft X-ray pulses from gasphase HHG and extreme ultraviolet (XUV) pulses from solid-state HHG. Then, we discuss their current and potential usage in time-resolved study of electron and nuclear dynamics in atomic, molecular and condensed matters.

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Journal of Physics B: Atomic, Molecular and Optical Physics
Publication Identifier Type: DOI **Publication Identifier:** 10.1088/1361-6455/ab8e57
Volume: 53 **Issue:** 14 **First Page #:** 145602
Date Submitted: 11/9/20 12:00AM **Date Published:** 7/1/20 7:00AM
Publication Location:

Article Title: Beam optimization in a 25 TW femtosecond laser system for high harmonic generation

Authors: Jialin Li, Yang Wang, Tianyi Guo, Jonathon White, Matthew Weidman, Yi Wu, Kai Hu, Marieke F Jager,

Keywords: wavefront correction, high harmonic generation, XUV

Abstract: It has been demonstrated in the past that high fluxes of extreme ultraviolet (XUV) light could be obtained by driving high harmonic generation (HHG) with high energy lasers. However, the peak intensity at the focal point of a femtosecond laser with more than 100 mJ can be too high for phase-matched HHG in gases. We propose a method to optimize the spatial profile at off-focus locations of the high energy driving laser to avoid fully ionizing the target atoms. The beam profile before or after the focal point depends on the wavefront quality of the laser beam and the near field intensity distribution. The beam spot diameter reaches three times that at the focus by adding customized wavefront terms. An XUV pulse energy of 5.6 μ J was obtained from HHG when a larger off-focus spot of a 500 mJ Ti: sapphire laser beam was applied in an argon gas cell.

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Journal: Scientific Reports

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Date Submitted: 11/9/20 12:00AM

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Publication Location:

Article Title: Attosecond pulse retrieval from noisy streaking traces with conditional variational generative network

Authors: Zheyuan Zhu, Jonathon White, Zenghu Chang, Shuo Pang

Keywords: ultrafast electron dynamics, noisy streaking traces, variational generative network

Abstract: Accurate characterization of an attosecond pulse from streaking trace is an indispensable step in studying the ultrafast electron dynamics on the attosecond scale. Conventional attosecond pulse retrieval methods face two major challenges: the ability to incorporate a complete physics model of the streaking process, and the ability to model the uncertainty of pulse reconstruction in the presence of noise. Here we propose a pulse retrieval method based on conditional variational generative network (CVGN) that can address both demands. Instead of learning the inverse mapping from a streaking trace to a pulse profile, the CVGN models the distribution of the pulse profile conditioned on a given streaking trace measurement, and is thus capable of assessing the uncertainty of the retrieved pulses. This capability is highly desirable for low-photon level measurement, which is typical in attosecond streaking experiments in the water window X-ray range.

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Date Submitted: 11/9/20 12:00AM

Date Published: 3/4/20 8:00AM

Publication Location:

Article Title: Signal Retrieval With Measurement System Knowledge Using Variational Generative Model

Authors: Zheyuan Zhu, Yangyang Sun, Johnathon White, Zenghu Chang and Shuo Pang

Keywords: Variational generative model, computational imaging, neural networks, inverse problem

Abstract: Signal retrieval from a series of indirect measurements is a common task in many imaging, metrology, and characterization platforms in science and engineering. Because most of the indirect measurement processes are well-described by physical models, signal retrieval can be solved with an iterative optimization that enforces measurement consistency and prior knowledge on the signal. These iterative algorithms are time-consuming and only accommodate a linear measurement process and convex signal constraints. Recently, neural networks have been widely adopted to supersede iterative methods by directly approximating the inverse mapping of the measurement process. However, such vanilla network with a deterministic multi-layer structure is unable to distinguish signal ambiguities in ill-posed measurement systems, and the retrieved signals often lack consistency with the measurement. In this work, we incorporate the known measurement process into a customized variational generative model to c

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Journal: Optics Express
Publication Identifier Type: DOI **Publication Identifier:** 10.1364/OE.27.030280
Volume: 27 **Issue:** 21 **First Page #:** 30280
Date Submitted: 11/9/20 12:00AM **Date Published:** 10/1/19 7:00AM
Publication Location:

Article Title: Double optical gating for generating high flux isolated attosecond pulses in the soft X-ray regime

Authors: Jie Li, Andrew Chew, Shuyuan Hu, Jonathon White, Xiaoming Ren, Seunghwoi Han, Yanchun Yin, Yan

Keywords: Double optical gating, polarization based gating , high flux isolated attosecond pulses

Abstract: Double optical gating (DOG) technique was implemented with a two-cycle, 1.7 μm driving field to generate isolated attosecond pulses in the 100–250 eV spectrum range. The strong ellipticity dependency of the high harmonics from the 1.7 μm driving field makes polarization based gating method very efficient. When a second harmonic (SH) field is introduced, complete gating can be achieved with less ionization from the leading edge of the driving field, which yields supercontinua with a pulse energy of 0.3 nJ. We perform an attosecond streaking measurement to confirm the generation of isolated attosecond pulses.

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Nature Communications
Publication Identifier Type: DOI **Publication Identifier:** 10.1038/s41467-018-08131-8
Volume: 10 **Issue:** 1 **First Page #:**
Date Submitted: 11/9/20 12:00AM **Date Published:** 1/1/19 8:00AM
Publication Location:

Article Title: Electron correlation driven non-adiabatic relaxation in molecules excited by an ultrashort extreme ultraviolet pulse

Authors: A. Marciniak, V. Despré, V. Lorient, G. Karras, M. Hervé, L. Quintard, F. Catoire, C. Joblin

Keywords: time-dependent two-color TR-PES, ultrashort extreme ultraviolet pulses, Vibronic-coupling Hamiltonian

Abstract: The many-body quantum nature of molecules determines their static and dynamic properties, but remains the main obstacle in their accurate description. Ultrashort extreme ultraviolet pulses offer a means to reveal molecular dynamics at ultrashort timescales. Here, we report the use of time-resolved electron-momentum imaging combined with extreme ultraviolet attosecond pulses to study highly excited organic molecules. We measure relaxation timescales that increase with the state energy. High-level quantum calculations show these dynamics are intrinsic to the time-dependent many-body molecular wavefunction, in which multi-electronic and non-Born-Oppenheimer effects are fully entangled. Hints of coherent vibronic dynamics, which persist despite the molecular complexity and high-energy excitation, are also observed. These results offer opportunities to understand the molecular dynamics of highly excited species involved in radiation damage and astrochemistry, and the role of quantum mechanics.

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Journal: Journal of Physics B: Atomic, Molecular and Optical Physics

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Publication Identifier: 10.1088/1361-6455/aba1ca

Volume: 53

Issue: 18

First Page #: 184006

Date Submitted: 11/9/20 12:00AM

Date Published: 9/1/20 7:00AM

Publication Location:

Article Title: Electronic decay through non-linear carbon chains

Authors: John B Mullenix, Victor Despré, Alexander I Kuleff

Keywords: non-local decay, ultrafast electron dynamics, electron correlation, carbon chains

Abstract: A multielectron wave-packet propagation method was used to calculate the electronic decay of oxygen and fluorine 2s vacancies for a group of trifluoroalkyl alcohols, $\text{HOCnH}(2n+1)\text{F}_3$, with n between 1 and 5. Whether ionizing O2s or F2s orbitals, it is shown that an electron can be emitted non-locally from the opposite terminus of the molecule. The decay of the $\text{O}(2s+1)$ state is found to be about 2–3 times faster than that of the $\text{F}(2s+1)$, but in both cases the process takes only a few femtoseconds, demonstrating a highly efficient energy transfer through the carbon bridge. A comparison to the previously reported non-local decay in linear difluorocumulenone systems shows that the non-linearity of the trifluoroalkyl alcohols does not appear to dramatically influence the decay efficiency. These results shed light onto the nature of the scaling of electron correlation and open the door to the potential design of molecules that take advantage of this mechanism.

Distribution Statement: 1-Approved for public release; distribution is unlimited.

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nature Phys.

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41567-020-01073-3

Volume: 17

Issue:

First Page #: 327

Date Submitted: 6/13/21 12:00AM

Date Published: 11/17/20 12:00AM

Publication Location:

Article Title: Ultrafast dynamics of correlation bands following XUV molecular photoionization

Authors: M. Hervé, V. Despré, P. Castellanos Nash, V. Loriot, A. Boyer, A. Scognamiglio, G. Karras, R. Brédy, E.

Keywords: Attosecond, Dynamics, Molecules, Nonadiabatic, XUV

Abstract: Extreme ultraviolet (XUV) transient absorption spectroscopy has emerged as a sensitive tool for mapping the real-time structural and electronic evolution of molecules. Here, attosecond XUV transient absorption is used to track dynamics in the A-band of methyl iodide (CH_3I). Gaseous CH_3I molecules are excited to the A-band by a UV pump (277 nm, 20 fs) and probed by attosecond XUV pulses targeting iodine $\text{I}(4d)$ core-to-valence transitions. Owing to the excellent temporal resolution of the technique, passage through a conical intersection is mapped through spectral signatures of nonadiabatic wave packet bifurcation observed 15 ± 4 fs following UV photoexcitation. The observed XUV signatures and time dynamics are in agreement with previous simulations [H. Wang, et al. J. Chem. Phys. 151, 124106 (2019)]. Due to the short duration of the UV pump pulse, coherent vibrational motion in the CH_3I ground state along the C-I stretch mode (538 ± 7 cm^{-1}) launched by resonant impulsive stimulated Raman

Distribution Statement: 1-Approved for public release; distribution is unlimited.

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Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nature Commun

Publication Identifier Type: DOI

Publication Identifier: <https://doi.org/10.1038/s41467-020-19496-0>

Volume: 11

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First Page #: 1

Date Submitted: 6/13/21 12:00AM

Date Published: 11/17/20 12:00AM

Publication Location:

Article Title: Attosecond spectroscopy reveals alignment dependent core-hole dynamics in the ICI molecule

Authors: Hugo J. B. Marroux, Ashley P. Fidler, Aryya Ghosh, Yuki Kobayashi, Kirill Gokhberg, Alexander I. Kuleff

Keywords: Attosecond, XUV, Molecules, Spectroscopy, Nonadiabatic

Abstract: Extreme ultraviolet (XUV) transient absorption spectroscopy has emerged as a sensitive tool for mapping the real-time structural and electronic evolution of molecules. Here, attosecond XUV transient absorption is used to track dynamics in the A-band of methyl iodide (CH₃I). Gaseous CH₃I molecules are excited to the A-band by a UV pump (277 nm, ~20 fs) and probed by attosecond XUV pulses targeting iodine I(4d) core-to-valence transitions. Owing to the excellent temporal resolution of the technique, passage through a conical intersection is mapped through spectral signatures of nonadiabatic wave packet bifurcation observed 15 ± 4 fs following UV photoexcitation. The observed XUV signatures and time dynamics are in agreement with previous simulations [H. Wang, et al. J. Chem. Phys. 151, 124106 (2019)]. Due to the short duration of the UV pump pulse, coherent vibrational motion in the CH₃I ground state along the C-I stretch mode (538 ± 7 cm⁻¹) launched by resonant impulsive stimulated Raman

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Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 5-Submitted

Journal: Phys. Rev. Lett.

Publication Identifier Type:

Publication Identifier:

Volume:

Issue:

First Page #:

Date Submitted:

Date Published:

Publication Location:

Article Title: Core-valence attosecond transient absorption spectroscopy of polyatomic molecules

Authors: N. V. Golubev, J. Vaníček, and A. I. Kuleff

Keywords: electronic coherence, attosecond

Abstract: Tracing ultrafast processes induced by interaction of light with matter is often very challenging. In molecular systems, the initially created electronic coherence becomes damped by the slow nuclear rearrangement on a femtosecond timescale which makes real-time observations of electron dynamics in molecules particularly difficult. In this work, we demonstrate theoretically that the attosecond transient absorption spectroscopy (ATAS) can be a very useful technique to trace such ultrafast processes in molecules. We report an extension of the theory underlying ATAS for the case of molecules, including a full account for the coupled electron-nuclear dynamics in the initially created wave packet, and apply it to probe the oscillations of the positive charge created after outer-valence ionization of the propiolic acid molecule. By taking advantage of element-specific core-to-valence transitions induced by X-ray radiation, we show that the resolution of ATAS makes it possible to trace the dyn

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: The Journal of Physical Chemistry C

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acs.jpcc.1c02101

Volume: 125

Issue: 17

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Date Submitted: 6/13/21 12:00AM

Date Published: 4/1/21 9:00PM

Publication Location:

Article Title: Characterization of Carrier Cooling Bottleneck in Silicon Nanoparticles by Extreme Ultraviolet (XUV) Transient Absorption Spectroscopy

Authors: Ilana J. Porter, Angela Lee, Scott K. Cushing, Hung-Tzu Chang, Justin C. Ondry, A. Paul Alivisatos, Ste

Keywords: Nonoparticles, Silicon, Hot Carriers, Ultraviolet, Phonon Bottleneck

Abstract: Silicon nanoparticles have the promise to surpass the theoretical efficiency limit of single-junction silicon photovoltaics by the creation of a "phonon bottleneck," a theorized slowing of the cooling rate of hot optical phonons that in turn reduces the cooling rate of hot carriers in the material. Verifying the presence of a phonon bottleneck in silicon nanoparticles requires simultaneous resolution of electronic and structural changes at short timescales. Here, extreme ultraviolet transient absorption spectroscopy is used to observe the excited-state electronic and lattice dynamics in polycrystalline silicon nanoparticles following 800 nm photoexcitation, which excites carriers with 0.35–0.03 eV excess energy above the Γ 1 conduction band minimum. The nanoparticles have nominal 100 nm diameters with crystalline grain sizes of about $\sim$ 16 nm. The extracted carrier-phonon and phonon-phonon relaxation times of the nanoparticles are compared to those for a silicon (100) single-crystal thin

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Structural Dynamics

Publication Identifier Type: DOI

Publication Identifier: 10.1063/4.0000048

Volume: 8

Issue: 1

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Date Submitted: 6/13/21 12:00AM

Date Published: 1/2/21 12:00AM

Publication Location:

Article Title: Carrier-specific dynamics in 2H-MoTe

Authors: Alexander Britz, Andrew R. Attar, Xiang Zhang, Hung-Tzu Chang, Clara Nyby, Aravind Krishnamoorthy,

Keywords: Ultraviolet, Attosecond, XUV, Transient Absorption, Spectroscopy

Abstract: Extreme ultraviolet (XUV) transient absorption spectroscopy has emerged as a sensitive tool for mapping the real-time structural and electronic evolution of molecules. Here, attosecond XUV transient absorption is used to track dynamics in the A-band of methyl iodide (CH₃I). Gaseous CH₃I molecules are excited to the A-band by a UV pump (277 nm, $\sim$ 20 fs) and probed by attosecond XUV pulses targeting iodine I(4d) core-to-valence transitions. Owing to the excellent temporal resolution of the technique, passage through a conical intersection is mapped through spectral signatures of nonadiabatic wave packet bifurcation observed 15–4 fs following UV photoexcitation. The observed XUV signatures and time dynamics are in agreement with previous simulations [H. Wang, et al. J. Chem. Phys. 151, 124106 (2019)]. Due to the short duration of the UV pump pulse, coherent vibrational motion in the CH₃I ground state along the C-I stretch mode (538 $\pm$ 7 cm⁻¹) launched by resonant impulsive stimulated Raman

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review Research
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevResearch.3.013228
Volume: 3 **Issue:** 1 **First Page #:** 013228
Date Submitted: 6/13/21 12:00AM **Date Published:** 3/10/21 4:00PM
Publication Location:

Article Title: Correlated variational treatment of ionization coupled to nuclear motion: Ultrafast pump and ionizing probe of electronic and nuclear dynamics in LiH

Authors: Roger Y. Bello, Robert R. Lucchese, Thomas N. Rescigno, C. William McCurdy

Keywords: Attosecond, XUV, Dynamics, Spectroscopy, Dynamics

Abstract: Extreme ultraviolet (XUV) transient absorption spectroscopy has emerged as a sensitive tool for mapping the real-time structural and electronic evolution of molecules. Here, attosecond XUV transient absorption is used to track dynamics in the A-band of methyl iodide (CH₃I). Gaseous CH₃I molecules are excited to the A-band by a UV pump (277 nm, ~20 fs) and probed by attosecond XUV pulses targeting iodine I(4d) core-to-valence transitions. Owing to the excellent temporal resolution of the technique, passage through a conical intersection is mapped through spectral signatures of nonadiabatic wave packet bifurcation observed 15 ± 4 fs following UV photoexcitation. The observed XUV signatures and time dynamics are in agreement with previous simulations [H. Wang, et al. J. Chem. Phys. 151, 124106 (2019)]. Due to the short duration of the UV pump pulse, coherent vibrational motion in the CH₃I ground state along the C-I stretch mode (538 ± 7 cm⁻¹) launched by resonant impulsive stimulated Raman

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Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Structural Dynamics
Publication Identifier Type: DOI **Publication Identifier:** <https://doi.org/10.1063/4.0000048>
Volume: 8 **Issue:** **First Page #:** 014501
Date Submitted: 6/13/21 12:00AM **Date Published:** 1/13/21 4:00PM
Publication Location:

Article Title: Carrier-specific dynamics in 2H-MoTe₂ observed by femtosecond soft x-ray absorption spectroscopy using an x-ray freeelectron laser

Authors: A. Britz, A. R. Attar, X. Zhang, H. T. Chang, C. Nyby, A. Krishnamoorthy, S. H. Park, S. Kwon, M. Kim, I.

Keywords: XFEL, transient absorption, electrons, spectroscopy, femtosecond

Abstract: Femtosecond carrier dynamics in layered 2H-MoTe₂ semiconductor crystals have been investigated using soft x-ray transient absorption spectroscopy at the x-ray free-electron laser (XFEL) of the Pohang Accelerator Laboratory. Following above-bandgap optical excitation of 2H-MoTe₂, the photoexcited hole distribution is directly probed via short-lived transitions from the Te 3d_{5/2} core level (M₅-edge, 572–577 eV) to transiently unoccupied states in the valence band. The optically excited electrons are separately probed via the reduced absorption probability at the Te M₅-edge involving partially occupied states of the conduction band. A 400±110 fs delay is observed between this transient electron signal near the conduction band minimum compared to higher-lying states within the conduction band, which we assign to hot electron relaxation. Additionally, the transient absorption signals below and above the TeM₅ edge, assigned to photoexcited holes and electrons, respectively, are observed to d

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Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nat. Phys.

Publication Identifier Type: DOI

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Volume: 17

Issue:

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Date Submitted: 6/13/21 12:00AM

Date Published: 11/16/20 4:00PM

Publication Location:

Article Title: Ultrafast dynamics of correlation bands following XUV molecular photoionization

Authors: M. Hervé, V. Despré, P. Castellanos Nash, V. Lorient, A. Boyer, A. Scognamiglio, G. Karras, R. Brédy, E.

Keywords: Ultrashort, X-ray, XUV, electron, matter

Abstract: Modern ultrashort X-ray/XUV (extreme ultraviolet) sources provide unique opportunities to investigate the primary reactions of matter upon energetic excitation. Understanding these processes in molecules on ultrafast timescales is required to improve bespoke high-energy radiation detectors, nanomedicine schemes or to study the molecular composition of interstellar media. However, current experiments struggle to provide a general framework because of the uniqueness and complexity of each system. Here we show the universal role of correlation bands—features created by electron correlation. This is done by studying ultrafast energy relaxation of size-scalable two-dimensional molecules following ionization by an ultrashort XUV pulse. We observed long lifetimes that nonlinearly increase with the number of valence electrons. A general law based on solid-like electron–phonon scattering is proposed, which explains both our results and previously reported measurements. This offers new opportuni

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Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review Research

Publication Identifier Type: DOI

Publication Identifier: <https://doi.org/10.1103/PhysRevResearch.3.013228>

Volume: 3

Issue:

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Date Submitted: 6/13/21 12:00AM

Date Published: 3/10/21 4:00PM

Publication Location:

Article Title: A Correlated Variational Treatment of Ionization Coupled to Nuclear Motion: Ultrafast Pump and Ionizing Probe of Electronic and Nuclear Dynamics in LiH

Authors: Roger Y. Bello, Robert R. Lucchese, Thomas N. Rescigno, and C. William McCurdy

Keywords: wave packet, photoelectron, ionization, LiH molecule

Abstract: Tracing ultrafast processes induced by interaction of light with matter is often very challenging. In molecular systems, the initially created electronic coherence becomes damped by the slow nuclear rearrangement on a femtosecond timescale which makes real-time observations of electron dynamics in molecules particularly difficult. In this work, we demonstrate theoretically that the attosecond transient absorption spectroscopy (ATAS) can be a very useful technique to trace such ultrafast processes in molecules. We report an extension of the theory underlying ATAS for the case of molecules, including a full account for the coupled electron-nuclear dynamics in the initially created wave packet, and apply it to probe the oscillations of the positive charge created after outer-valence ionization of the propiolic acid molecule. By taking advantage of element-specific core-to-valence transitions induced by X-ray radiation, we show that the resolution of ATAS makes it possible to trace the dyn

Distribution Statement: 1-Approved for public release; distribution is unlimited.

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CONFERENCE PAPERS:

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Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: High Intensity Lasers and High Field Phenomena
Date Received: Conference Date: 21-Mar-2016 Date Published: 21-Mar-2016
Conference Location:
Paper Title: Attosecond XUV Transient Absorption in Dense Systems: Interplay between Strong-field Effects and Resonant Pulse Propagation
Authors: Chen-Ting Liao, Seth Camp, Ken Schafer, Mette Gaarde, Arvinder S Sandhu
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: Nonlinear Optics
Date Received: Conference Date: 27-Jul-2015 Date Published: 27-Jul-2015
Conference Location:
Paper Title: Investigation of Non-adiabatic Molecular Dynamics with Attosecond Transient Absorption
Authors: Chen-Ting Liao, Xuan Li, Daniel Haxton, William McCurdy, Arvinder S Sandhu
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Bulletin of the American Physical Society - DAMOP 2017
Date Received: Conference Date: 26-May-2016 Date Published: 26-May-2016
Conference Location: Providence, Rhode Island
Paper Title: Observing Superexcited State Dynamics in Atomic and Molecular Systems by Attosecond Transient Absorption
Authors: Chen-Ting Liao, Xuan Li, Daniel Haxton, Robert Lucchese, C William McCurdy, Arvinder Sandhu
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Bulletin of the American Physical Society
Date Received: Conference Date: 16-Oct-2015 Date Published: 16-Oct-2015
Conference Location:
Paper Title: Attosecond spectroscopy: Application in study of conical intersections
Authors: Arvinder Sandhu
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: Conference on Lasers and Electro-Optics
Date Received: Conference Date: 15-May-2017 Date Published: 15-May-2017
Conference Location: San Jose, CA
Paper Title: Investigating Impulsive Strong Field Perturbation of Molecular Rydberg States with XUV Transient Absorption
Authors: Conference on Lasers and Electro-Optics
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: DAMOP 2017
Date Received: Conference Date: 05-Jun-2017 Date Published: 06-Jun-2017
Conference Location: Sacramento, CA
Paper Title: Controlling electronic couplings with tunable long wavelength pulses: Study of Autler-Townes splitting and XUV emission spectra
Authors: Nathan Harkema, Chen-Ting Liao, Arvinder Sandhu
Acknowledged Federal Support: **Y**

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Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: Conference on Lasers and Electro-Optics
Date Received: Conference Date: 13-May-2017 Date Published: 14-May-2017
Conference Location: San Jose, CA
Paper Title: Control of Laser Induced Couplings in Autoionizing States by XUV Transient Absorption
Authors: Chen-Ting Liao, Nathan Harkema, Arvinder Sandhu
Acknowledged Federal Support: Y

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: American Physical Society Annual National Convention
Date Received: Conference Date: 17-Mar-2017 Date Published:
Conference Location: New Orleans, LA
Paper Title: Attosecond electronic band gap dynamics
Authors: Stephen R. Leone
Acknowledged Federal Support: Y

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: CLEO: QELS_Fundamental Science
Date Received: 25-Jun-2018 Conference Date: 18-May-2018 Date Published: 18-May-2018
Conference Location: San Jose, California
Paper Title: Electron dynamics in transition metal dichalcogenides utilizing attosecond transient absorption spectroscopy
Authors: Alexander Guggenmos, Hung-Tzu Chang, Michael Zurek, Diana Y. Qiu, Romain Geneaux, Yen-Chang
Acknowledged Federal Support: Y

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: CLEO 2018
Date Received: 27-Jun-2018 Conference Date: 13-May-2018 Date Published:
Conference Location: San Jose Convention Center, San Jose, CA
Paper Title: Attosecond Transient Absorption and Four-Wave Mixing with Tunable IR Pulses
Authors: Nathan Harkema, Jens E. Bækhoj, Kenneth J. Schafer, Mette B. Gaarde, Chen-Ting Liao, Arvinder Sandhu
Acknowledged Federal Support: Y

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: ACS National Meeting
Date Received: Conference Date: 19-Aug-2018 Date Published:
Conference Location: Boston, MA
Paper Title:
Authors: Lorenz Cederbaum
Acknowledged Federal Support: Y

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: 256th ACS National Meeting
Date Received: Conference Date: 19-Aug-2018 Date Published:
Conference Location: Boston, Massachusetts
Paper Title: Non-adiabatic coherent electron dynamics in iodine monobromide probed by XUV attosecond transient-absorption spectroscopy
Authors: Y. Kobayashi, K. F. Chang, T. Zeng, D. M. Neumark, S. R. Leone
Acknowledged Federal Support: Y

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Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: International Conference on Ultrafast Phenomena
Date Received: Conference Date: 15-Jul-2018 Date Published:
Conference Location: Hamburg, Germany
Paper Title: Ultrafast dissociation of vinyl bromide after passage through a conical intersection: an attosecond transient absorption spectroscopy study
Authors: M. Reduzzi, Y. Kobayashi, F. Rott, T. Schnappinger, H. Timmers, K. F. Chang, S. Oesterling, D. M. Neu
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Physical Chemistry Colloquium
Date Received: Conference Date: 01-Oct-2018 Date Published:
Conference Location:
Paper Title: Attosecond science and applications to chemical dynamics
Authors: Daniel M. Neumark
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: MURI Program Review
Date Received: Conference Date: 15-Nov-2018 Date Published:
Conference Location: Arlington, VA
Paper Title: Attosecond XUV probing of coherence, in atoms and molecules
Authors: Daniel M. Neumark
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Special Lecture(s)
Date Received: Conference Date: 01-Mar-2019 Date Published:
Conference Location: China
Paper Title: Unimolecular dynamics studied by negative ion photodetachment and attosecond transient absorption (three lectures)
Authors: Daniel M. Neumark
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: ACS National Meeting (Debye Award Lecture)
Date Received: Conference Date: 01-Apr-2019 Date Published:
Conference Location: Orlando, FL
Paper Title: Adventures in spectroscopy and dynamics
Authors: Daniel M. Neumark
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Dreyfus Foundation Lecture
Date Received: Conference Date: 11-Mar-2019 Date Published:
Conference Location: The State University of New York, Geneseo, NY
Paper Title: Attosecond Electron Dynamics: From Atoms to Semiconductor Solids
Authors: Stephen R. Leone
Acknowledged Federal Support: **Y**

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Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Quantum Effects In Small Molecular Systems: Faraday Discussion
Date Received: Conference Date: 11-Sep-2018 Date Published:
Conference Location: John McIntyre Conference Centre, Edinburgh, UK
Paper Title: Multiple pulse coherent dynamics and wave packet control of the N2 a? 1?g+ dark state by attosecond four wave mixing
Authors: Stephen R. Leone
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Dreyfus Foundation Special Public Lecture
Date Received: Conference Date: 12-Mar-2019 Date Published:
Conference Location: The State University of New York Geneseo, NY
Paper Title: X-ray Eyes
Authors: Stephen R. Leone
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Applied Physics Seminar
Date Received: Conference Date: 18-Jan-2019 Date Published:
Conference Location: The University of Georgia, Athens, GA
Paper Title: Ultrafast X-Ray Molecular Dynamics
Authors: Stephen R. Leone
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Chemical Sciences & Engineering Colloquium
Date Received: Conference Date: 09-Apr-2019 Date Published:
Conference Location: Argonne National Laboratories, Argonne, IL
Paper Title: Attosecond Electron Dynamics: From Atoms to Semiconductor Solids
Authors: Stephen R. Leone
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 1-Published
Conference Name: APS Division of Atomic, Molecular and Optical Physics
Date Received: Conference Date: 30-May-2019 Date Published:
Conference Location: Milwaukee, Wisconsin
Paper Title: Photoionization calculations in molecules using an overset grid implementation
Authors: Carlos Marante , Loren Greenman, Robert L. Lucchese, C. William McCurdy, Thomas N. Rescigno
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: ACS National Meeting
Date Received: Conference Date: 19-Aug-2018 Date Published:
Conference Location: Boston, MA
Paper Title: ICD and its Exploration by Short, Intense and Coherent Light Pulses
Authors: Lorenz Cederbaum
Acknowledged Federal Support: **Y**

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as of 28-Sep-2021

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: QSCP XXIII
Date Received: Conference Date: 23-Sep-2018 Date Published:
Conference Location: Kruger Park, South Africa
Paper Title: How good is the Gross Pitaevskii wave function of BECs made of many bosons?
Authors: Lorenz Cederbaum
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: QSCP XXIII
Date Received: Conference Date: 23-Sep-2018 Date Published:
Conference Location: Kruger Park, South Africa
Paper Title: Ultrafast electron dynamics as a route to explore chemical processes
Authors: Alexander Kuleff
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Attosecond MURI annual meeting
Date Received: Conference Date: 15-Nov-2018 Date Published:
Conference Location: Arlington, VA
Paper Title: Ultrafast electron dynamics as a route to explore chemical processes
Authors: Alexander Kuleff
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Quantum Dynamics in Tailored Intense Fields (QUTIF annual meeting)
Date Received: Conference Date: 25-Feb-2019 Date Published:
Conference Location: Oldenburg, Germany
Paper Title: Charge-directed reactivity and its control by short laser pulses
Authors: Alexander Kuleff
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: ELI-ALPS 6th Science Day
Date Received: Conference Date: 19-Mar-2019 Date Published:
Conference Location: Szeged, Hungary
Paper Title: Correlation driven ultrafast electron dynamics
Authors: Alexander Kuleff
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: ACS National Meeting
Date Received: Conference Date: 31-Mar-2019 Date Published:
Conference Location: Orlando, FL
Paper Title: Is an efficient intermolecular energy transfer from vibrations to electronic motion possible?
Authors: Lorenz Cederbaum
Acknowledged Federal Support: **Y**

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Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Annual Meeting of the APS Division of Atomic, Molecular and Optical Physics
Date Received: Conference Date: 27-May-2019 Date Published:
Conference Location: Milwaukee, WI
Paper Title: Is an efficient intermolecular energy transfer from vibrations to electronic motion possible?
Authors: Lorenz Cederbaum
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Annual Meeting of the APS Division of Atomic, Molecular and Optical Physics
Date Received: Conference Date: 27-May-2019 Date Published:
Conference Location: Milwaukee, WI
Paper Title: Ultrafast coupled electron-nuclear dynamics following ionization of propiolic acid
Authors: Alexander Kuleff
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Chemistry Colloquium
Date Received: Conference Date: 01-Oct-2018 Date Published:
Conference Location: Caltech
Paper Title: Attosecond science and applications to chemical dynamics
Authors: Daniel M. Neumark
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: attosecond MURI meeting
Date Received: Conference Date: 01-Nov-2018 Date Published:
Conference Location: Arlington, VA
Paper Title: Attosecond XUV probing of coherence, in atoms and molecules
Authors: Daniel M. Neumark
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name:
Date Received: Conference Date: 02-Mar-2019 Date Published:
Conference Location: China
Paper Title: Unimolecular dynamics studied by negative ion photodetachment and attosecond transient absorption (three lectures)
Authors: Daniel M. Neumark
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: ACS National Meeting (Debye Award Lecture)
Date Received: Conference Date: 01-Apr-2019 Date Published:
Conference Location: Orlando, FL
Paper Title: Adventures in spectroscopy & dynamics
Authors: Daniel M. Neumark
Acknowledged Federal Support: **Y**

RPPR Final Report
as of 28-Sep-2021

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: 21st Photonics North Conference
Date Received: Conference Date: 21-May-2019 Date Published:
Conference Location: Quebec city, Canada
Paper Title: Novel mid-infrared lasers for pushing the attosecond science frontier
Authors: Zenghu Chang
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Frontiers in Optics/Laser Science
Date Received: Conference Date: Date Published:
Conference Location: Washington DC
Paper Title: Microjoule-level 20 fs UV pulses for molecular dynamics via attosecond transient absorption spectroscopy
Authors: K. F. Chang, M. Reduzzi, S. M. Poullain, Y. Kobayashi, D. M. Neumark, S. R. Leone
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: 14th Femtochemistry Conference
Date Received: Conference Date: 28-Jul-2020 Date Published:
Conference Location: Shanghai, China
Paper Title: Direct mapping of photochemical dynamics across conical intersections by XUV transient absorption spectroscopy.
Authors: K. F. Chang, M. Reduzzi, Y. Kobayashi, D. M. Neumark, S. R. Leone
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Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: Nonlinear Optics
Date Received: Conference Date: 14-Jul-2019 Date Published:
Conference Location: Waikaloa Village, HI
Paper Title: Studying Multielectron Excitation and Fragmentation with Ultrafast XUV-IR Spectroscopy
Authors: A. Plunkett, N. Harkema, Robert Lucchese, C. W. McCurdy, A. S. Sandhu
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: APS March Meeting 2021
Date Received: Conference Date: 15-Mar-2021 Date Published:
Conference Location: Virtual
Paper Title: High harmonic generation driven by mid-infrared lasers
Authors: Zenghu Chang
Acknowledged Federal Support: **Y**

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: CECS Virtual Seminar Series
Date Received: Conference Date: 19-Feb-2021 Date Published:
Conference Location: University of Central Florida
Paper Title: Attosecond X-rays in the Water Window
Authors: Zenghu Chang
Acknowledged Federal Support: **Y**

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Conference Name: High Intensity Lasers and High Field Phenomena

Date Received:

Conference Date: 16-Nov-2020

Date Published:

Conference Location: Virtual

Paper Title: Attosecond Transient Absorption Spectroscopy of Diatomic and Triatomic Molecules at 400 eV,"
High Intensity Lasers and High Field Phenomena,

Authors: Nariyuki Saito, Hiroki Sannohe, Nobuhisa Ishii, Nicolas Douguet, Teruto Kanai, Nobuhiro Kosugi, Yi Wu.

Acknowledged Federal Support: Y

DISSERTATIONS:

Publication Type: Thesis or Dissertation

Institution: University of Arizona

Date Received:

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Title: Exploring ultrafast quantum dynamics of electrons by attosecond transient absorption

Authors: Chen-Ting Liao

Acknowledged Federal Support: Y

PATENTS:

Intellectual Property Type: Patent

Date Received:

Patent Title: High energy broadband laser system, methods, and applications

Patent Abstract:

Patent Number: 10,790,631

Patent Country: USA

Application Date:

Application Status: 3

Date Issued: 29-Sep-2020

Partners

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Dr. Franck Lépine, ILM, CNRS, Lyon, France (collaboration on relaxation dynamics in PAHs). Collaboration on crea

I certify that the information in the report is complete and accurate:

Signature: Stephen R Leone

Signature Date: 9/27/21 2:31PM

Final Report
"Post-Born-Oppenheimer Dynamics Using Isolated Attosecond Pulses"
ARO Grant W911NF1410383

Stephen R. Leone (University of California, Berkeley)

Principal Investigator

(Daniel Neumark, Arvinder Sandhu, C. William McCurdy, Zenghu Chang, Lorenz Cederbaum, and Paul Corkum Participants)

June 2021

Laser Development and Attosecond Pulses: Prior to this MURI program, Ti:Sapphire lasers centered at 800 nm were the work horse for attosecond transient absorption spectroscopy (ATAS). The spectral range of isolated attosecond pulses with sufficient photon flux for ATAS was limited to extreme ultraviolet (10 to 150 eV). It was demonstrated in 2001 that the cutoff photon energy of the high harmonic spectrum can be extended by increasing the center wavelength of driving lasers. An Optical Parametric Chirped Pulse Amplification centered at 1.7 micron was developed by Chang in this program, which delivers 3 mJ, 12 fs, carrier-envelope stable pulses at 1 kHz repetition rate. By driving high harmonic generation and implementing polarization gating, isolated X-ray pulses reaching the Carbon K-edge with 53-as duration have been characterized by attosecond streaking measurements for the first time.

The attosecond X-ray pulses covering the “water window” region (284-543 eV) has enabled tracing fundamental dynamics in atoms or molecules consisting of light elements with an ultimate temporal resolution in an electronic-state-specific and element-specific manner. Chang’s group and their collaborator, Jiro Itatani’s team at the University of Tokyo, observed sub-optical-cycle electron dynamics in NO molecules by attosecond transient absorption spectroscopy. It is the first time that ATAS was demonstrated for molecules at the N K-edge (400 eV). Very recently, they applied the same technique to N₂O molecules.

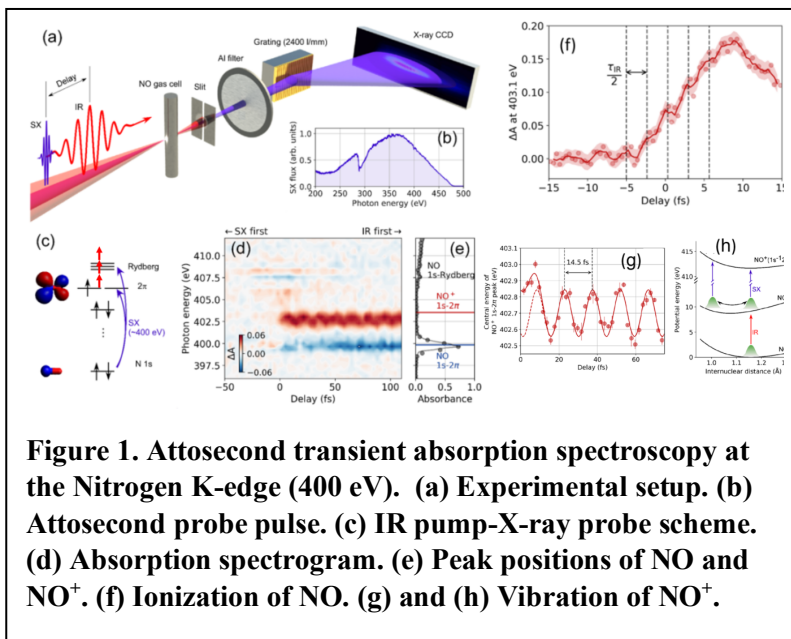
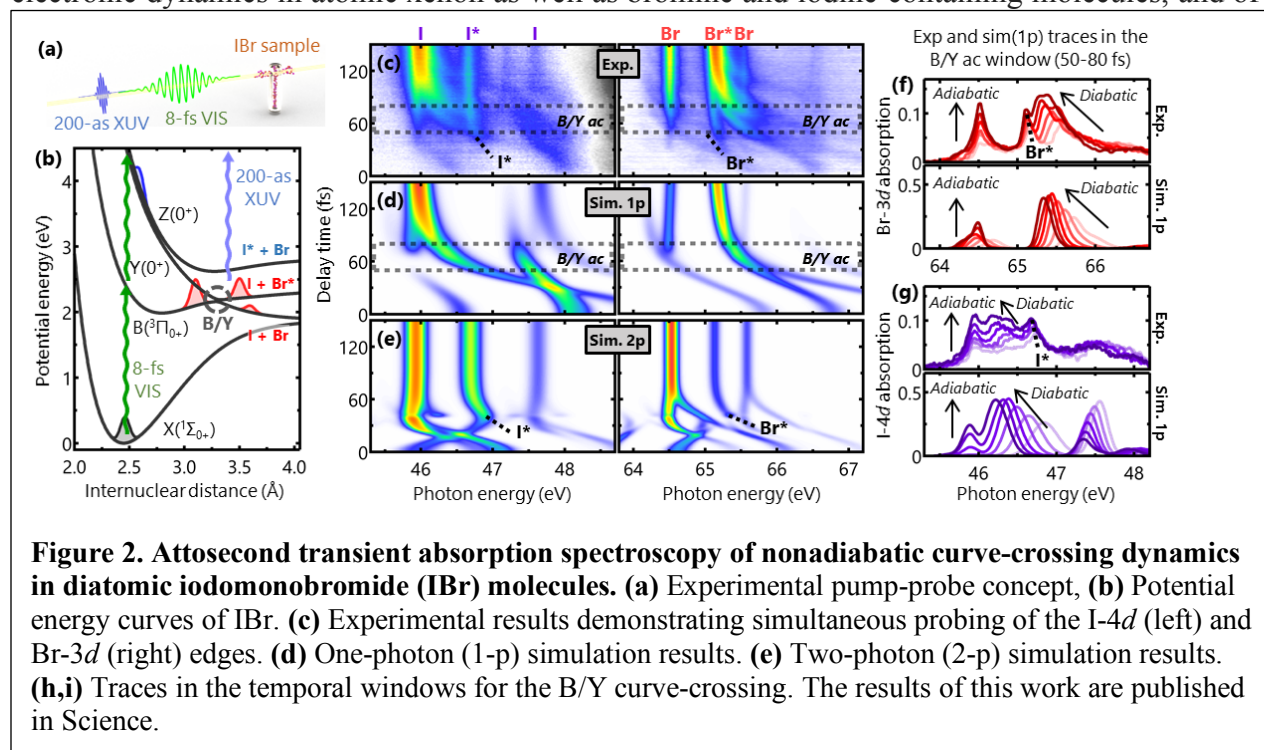


Figure 1 shows the measured transient absorption spectra of NO. The pump intensity for NO is set to $\sim 1 \times 10^{14}$ W/cm². It is observed that the absorption peaks of the neutral species decrease and that of the cation species

(NO⁺) increases. This fact clearly indicates that the strong-field ionization process takes place. Furthermore, on top of the monotonic increase or decrease of the absorbance, oscillatory structures with a period of a half cycle of the pump IR pulse are observed. The oscillation phase is opposite for the absorption peaks of the neutral and the cation species, implying that the origin of the oscillation is the change of the hole population in their HOMO orbitals.

Interestingly, in the case of N₂O, it is observed that the oscillation amplitude is smaller in the Nc1s-3 π peak than that in the Nt1s-3 π peak, even though they both probe the same 3 π orbital. Here, Nc and Nt mean the “central” and “terminal” nitrogen atoms, respectively. Because the 1s core orbital of each nitrogen atom is localized, the Nc1s-3 π and Nt1s-3 π peaks are expected to probe different locations of the 3 π orbital. Therefore, the observed difference of the oscillation amplitude in these peaks might reflect the intramolecular position-specific electronic dynamics.

Nonadiabatic and coherent dynamics in gas phase atomic and molecular systems: The observation of ultrafast nonadiabatic and coherent dynamics in gaseous systems is a central topic in attosecond science. In the Leone and Neumark groups at Berkeley, attosecond transient absorption spectroscopy based on a table-top source of attosecond XUV pulses was performed on a series of atomic, diatomic, and polyatomic molecular systems. With attosecond XUV probe pulses spanning the 40-70 eV photon energy region, atomic and molecular dynamics in the gas phase were probed through element-specific Xe-4*d*, Br-3*d*, and I-4*d* core-to-valence transitions. Combined with developed sources of strong-field NIR pulses lasting a few optical cycles in duration, femtosecond visible pump pulses, and femtosecond UV pump pulses, the attosecond transient absorption methodology allows for the preparation and detection of strong-field driven electronic dynamics in atomic xenon as well as bromine and iodine-containing molecules, and of

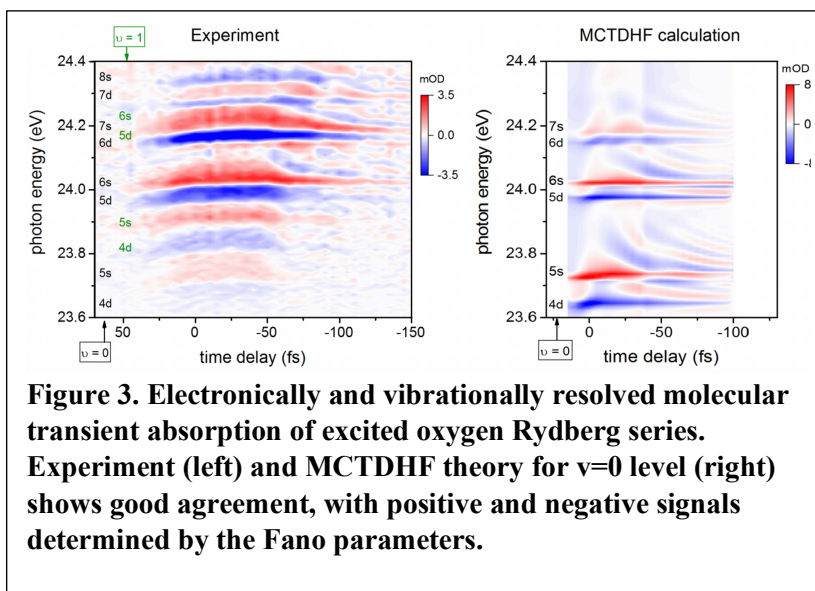


nonadiabatic electronic-nuclear dynamics at curve-crossings (Figure 2) and conical intersections

in brominated and iodinated molecular species. This work has led to publications in *Optica*, *Optics Letters*, *Physical Review A*, *Physical Review Letters*, *Nature Physics*, *Nature Communications*, *The Journal of Chemical Physics*, *Structural Dynamics*, and *Science*. Finally, the extension of attosecond transient absorption to the direct probing of carbon atoms comprising polyatomic organic molecules, was enabled by a newly-developed attosecond source of ~ 100 -400 eV pulses targeting carbon K-edge transitions in the soft X-ray regime. A manuscript summarizing the development of this new source is published in *Scientific Reports*.

Condensed-phase dynamics in solid-state materials and aqueous biological systems: The development of spectroscopic techniques based on table-top attosecond and femtosecond XUV sources for the investigation of condensed-phase dynamics in solids and in aqueous environments have expanded the applications of ultrafast XUV spectroscopy to materials science and photobiology. In the Leone and Neumark groups at Berkeley, attosecond transient absorption spectroscopy was used to resolved fundamental carrier dynamics in key semiconductor and metallic systems including Silicon, Germanium, Magnesium Oxide, and Nickel, and in layered semiconductors such as Molybdenum Ditelluride and Tungsten Disulfide. The results of these works are published in *Nature Communications*, *Physical Review Letters*, *Physical Review B*, *ACS Nano*, and *Structural Dynamics*. In the Neumark group at Berkeley, the design of a new femtosecond XUV photoelectron spectroscopy setup paves the way to performing ultrafast spectroscopy on liquid-phase samples. With a new XUV light source at 21.7 eV, a laser-assisted photoelectric effect in liquid water using 800 nm and 400 nm dressing fields and ground and excited-state dynamics in aqueous organic dyes excited by 400 nm pump have been observed.

Light-Induced Dynamics: Sandhu employed attosecond extreme-ultraviolet and near-infrared laser pulses, to advance the scientific understanding of non-adiabatic and light-induced dynamics. They explored the interplay between resonant attosecond pulse propagation and laser induced phase in an optically thick gas to quantify the nature of collective effects and establish the criterion for consideration of such effects in attosecond transient absorption studies. In a significant step forward, they extended the strong field attosecond transient absorption to molecules. Superexcited oxygen was investigated and it was found that the sign and magnitude of the laser modified autoionizing lineshapes depends on the underlying Fano parameters.



Electronically and vibrationally resolved results were supported by MCTDH calculations conducted by the McCurdy group. The Sandhu group also performed molecular polarization

experiments in polyatomic molecules (SF_6) and observed the suppression of XUV photoabsorption due to inter-valence transitions in the presence of a strong near-infrared field.

A second arm of the Sandhu research program employed differential photoelectron imaging to investigate the fragmentation of autoionizing Rydberg states of oxygen. They identified a new channel leading to the formation of a p-shell excited atomic fragment, which is not an expected dissociation product. Analysis of the time-dependent photoelectron spectra conducted in collaboration with McCurdy group showed that this fragment results from a previously unexplored repulsive state associated with the multielectron excitations. They obtained data on the non-adiabatic dynamics in large polyatomics (Iodobenzene), indicating correlations between photofragmentation pathways. These results will be used to interpret the non-adiabatic coupling mechanisms. The MURI program has provided opportunities to train several graduate students in a frontier area of research, contributing to the development of the next generation of nation's scientific workforce.

Attosecond Pulse Characterization and Generation: In 2013, as the ARO MURI was about to begin, the Corkum group, working with a former postdoctoral fellow, Dr. Quéré, demonstrated the Light House method of generating isolated attosecond pulses in ionizing gases. Within the context of the MURI, the Light House provides multiple pulses of an attosecond pulse train for different applications (such as pump and probe) and the method became a focus of early experiments, including measuring the duration of individual pulses. The Light House was the first attosecond pulse generation method that did not depend on controlling the single atom response. Instead, the phased output of many emitters was controlled by the fundamental beam itself. The Light House method is closely related to *in situ* methods for attosecond pulse characterization, previously developed by the Corkum group and since shown to measure an attosecond pulse created in any atom, including multi-electron atoms (with Fano Resonances or the Giant Plasmon Resonance) or near Cooper minima, as accurately as by the streaking method (also first proposed by Corkum).

In 2015, the Corkum group showed the deep connection between solid and gas harmonic generation, and they began integrating solid and gas-phase high harmonic and attosecond pulse generation. Integration provided a second means of controlling the phased emitters and therefore, creating cleanly isolated attosecond pulses. In 2015 Corkum also became aware of the air (N_2^+ ion) laser, created by ultrashort pulse strong-field ionization. The laser showed that the N_2^+ in the air was inverted by the end of an ultrashort pulse with B-state population sufficient to allow lasing on the $\text{B} \rightarrow \text{X}$ transition. The question was, how was this possible when atto-measurements showed that strong field ionization created mainly X-state population by tunnelling? Thus began a series of molecular experiments on the dynamics of “air” lasing, with the first papers published in 2018 and the last in 2020.

The Heidelberg team of Kuleff and Cederbaum developed a computational

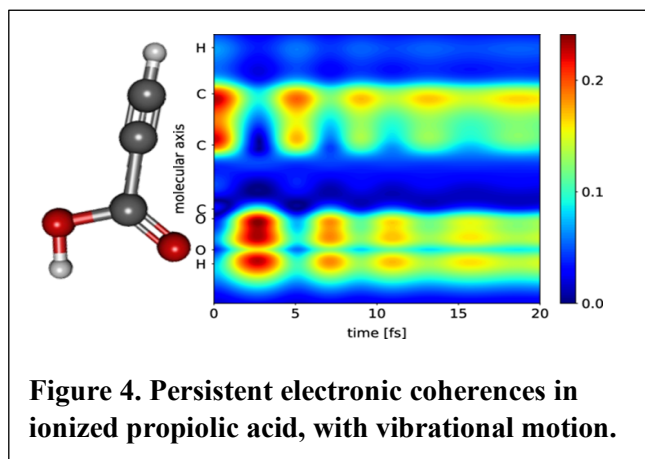


Figure 4. Persistent electronic coherences in ionized propionic acid, with vibrational motion.

scheme that allowed to accurately simulate ultrafast dynamics initiated by the ionization of a medium-size molecule, taking into account the correlated motion of all electrons and nuclei, as well as the influence of laser pulses on the evolution of the system. With the help of this methodology, various electron-correlation phenomena were studied, ranging from non-local electronic decay processes involving energy transfer through carbon chains to ultrafast charge migration phenomena triggered by core and outer-valence ionization. The first fully quantum coupled electron-nuclear dynamics simulations of charge migration in a polyatomic molecule were performed by the Heidelberg group, demonstrating that longer-lived electronic coherences can exist even in complex systems. The charge created upon outer-valence ionization of the propiolic acid molecule (Figure 4) can undergo several oscillations before getting trapped and delocalized by the nuclear motion. The figure shows the evolution of the hole-charge density. They also investigated the XUV-induced ultrafast non-adiabatic dynamics in polycyclic aromatic hydrocarbons and showed that the so-called correlation bands are responsible for characteristic relaxation dynamics observed in series of experiments. This allowed the derivation of simple electron-phonon scattering models that can accurately predict the relaxation of complex molecules after high-energy photoionization and provide important information about the XUV-induced chemistry in the interstellar media.

The second research line followed by the Heidelberg group was the study of light-induced conical intersections (LICI) in diatomic molecules. In a series of works, they were able to demonstrate the impact of LICI on the dissociation dynamics of homo- and heteronuclear diatomic molecules, and to show how LICI can be used to control the photofragmentation.

Theory of Attosecond Processes in Atoms and Molecules: The McCurdy group developed and exploited new theoretical capabilities in two areas: (1) Direct time-dependent simulation of photoionization ionization processes by one or more ultrashort pulses, and (2) New computational capabilities for the description of the electronic continuum states that are the final quantum states in photoionization. The large bandwidths of attosecond pulses almost always lead to attosecond experiments that – whether transient absorption, pump/probe or wave-mixing – access and probe the photoionization continuum. The simulation and interpretation of attosecond experiments on complex polyatomic molecules thus requires computational methods that can accurately describe the electron-ion scattering continuum and the coupled channels leading to different molecular ion states. The correlated description of the ionization continuum of polyatomic molecules is the signature of the McCurdy group's contribution to this MURI (Figure 5). The extension of those computational theoretical capabilities to higher energies and larger molecules, while including the effects of electron correlation to accurately describe multiple ionization channels and autoionization, were major goals of this part of the effort.

At the beginning of the grant period the McCurdy group investigated the coherent superposition that is formed when multiple ion states are created by a single attosecond pulse, including the role of electron correlation and coupling between ionization. Later, the control of the angular dependence of two-photon double ionization of atoms by varying the pulse length was investigated. In a collaboration with the Sandhu group in this MURI, the Schwinger multichannel method was used to interpret attosecond transient absorption experiments on superexcited states

of O₂. The group's activities in the time-dependent simulation of short pulse ionization culminated in a simulation of ultrafast pump and ionizing probe experiments with ionization fully coupled to nuclear motion in a diatomic molecule (Figure 5). In a major code and method development project, the group has produced a completely new computational tool for describing the ionization continuum, called the Overset Grid Complex Kohn Variational method. That code is now complete in its single channel version. Its multichannel version is under development and other publications based on it are being prepared. It will be made publicly available and will be a lasting legacy of this project.

