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Final Repor	rt: Bright Coh	erent Optical V	Waveforms from th	ne IR t	to W911	W911NF-12-1-0436		
the VUV				5b. GI	5b. GRANT NUMBER			
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Scientific Accomplishments: Four exciting advances resulted from this award 1. Revealing the Role of Electron-Electron Correlations during the Dissociation of Highly Excited D2 + using Attosecond XUV Pulses 2. Attosecond VUV Coherent Control of Molecular Dynamics 3. Attosecond Coherent Control of Single and Double Photoionization in Argon 4. Chemical dynamics relevant to ionospheric chemistry, interstellar processes and highenergy materials								
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	b. ABSTRACT	-	ABSTRACT		OF PAGES		Margaret Murnane	
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Report Title

Final Report: Bright Coherent Optical Waveforms from the IR to the VUV

ABSTRACT

Scientific Accomplishments: Four exciting advances resulted from this award

1. Revealing the Role of Electron-Electron Correlations during the Dissociation of Highly Excited D2 + using Attosecond XUV Pulses 2. Attosecond VUV Coherent Control of Molecular Dynamics 3. Attosecond Coherent Control of Single and Double Photoionization in Argon 4. Chemical dynamics relevant to ionospheric chemistry, interstellar processes and highenergy materials

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received	Paper
03/11/2016	4.00 C.W. Hogle, X.M. Tong, L. Martin, M.M. Murnane, H.C. Kapteyn, P. Ranitovic. Attosecond Coherent Control of Single and Double Photoionization in Argon, Physical Review Letters, (10 2015): 173004. doi: 10.1103/PhysRevLett.115.173004
TOTAL:	1

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

(c) Presentations

Invited and Keynote Presentations-

Plenary talk, Henry C. Kapteyn and Margaret M. Murnane, Frontiers in Optics 2013/ Laser Science XXIX, Orlando, FL, October 2013. Keynote talk, Margaret Murnane et al, "Coherent keV X-Rays from Tabletop Femtosecond Lasers and Applications in Nanometrology," 2013 International Workshop on EUV and Soft X-Ray Sources, Dublin, Ireland, November 2013. Presented by Margaret Murnane. Invited talk, Margaret Murnane et al, "Probing Electron Dynamics in Molecules, Quantum Dots and Materials at the Space-Time Limits Using Coherent Tabletop High Harmonic X-Rays," FEIS 2013 – Workshop on Femtosecond Electron Imaging and Spectroscopy, Key West, Florida, December 2013.

Invited talk, Wallenburg Foundation, Stockholm, Sweden, Jan 2014.

Invited talk, Uppsala University, Jan 2014.

Invited talk, AMOLF, Amsterdam, Jan 2014.

Invited talk, APS March Meeting, Denver, CO, March 2014.

Small Lecture, College of William and Mary, March 2014.

Irons Public Lecture, Rutgers University, March 2014.

Colloquium, Princeton University Mechanical Engineering, April 2014.

Colloquium, Cornell University Applied Physics, April 2014.

Colloquium, MIT Chemistry, May 2014.

Colloquium, CUNY Physics, May 2014.

Invited talk, CLEO-QELS Special Symposium for Howard Schlossberg at AFOSR, San Jose, CA, June 2014.

Invited talk, Conference on Light induced dynamics and control of correlated quantum systems, Hohwacht (Germany), June 2014. Ahmed Zewail Prize talk, Annual Meeting of the American Chemical Society, ""For the demonstration, development, and elucidation of the principles of femtosecond two-dimensional Fourier-transform spectroscopy, widely used for the investigation of diverse ultrafast phenomena.", New Orleans, LA, April 2013. (Presented by David Jonas)

Invited talk, Gordon Research Conference on Quantum Control, "Ultrafast Quantum Control in Atomic, Molecular and Materials Systems," South Hadley, MA, July 2013. (Presented by Margaret Murnane)

Invited talk, Ultrafast X-Ray Summer School, "Tabletop Coherent X-Rays: New Frontiers in X-Ray and Nano Science", Hamburg, Germany, June 2013. (Presented by Margaret Murnane)

Plenary talk, "Tabletop Coherent X-Rays: New Frontiers in X-Ray and Nano Science", Conference on Ultrafast Surface Dynamics, Estes Park, Colorado, May 2013. (Presented by Margaret Murnane)

Colloquium, Henry C. Kapteyn and Margaret Murnane, "Ultrafast coherent x-rays from tabletop lasers—a new tool for science and technology," Free University (VU) Amsterdam, January 2013. Presented by Henry Kapteyn.

Invited talk, Henry C. Kapteyn, Predrag Ranitovic, Craig W. Hogle, Xibin Zhou, Leigh Martin, William Peters, Austin P. Spencer, David Jonas, Xiao-Min Tong, Margaret Murnane, "Molecular Dynamics with Ultrafast X-rays," Frontiers in Optics 2012/ Laser Science XXVIII, Rochester, NY, October 2012. Talk LTu1H.1. Presented by Henry Kapteyn

Number of Presentations: 21.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received		Paper		
08/21/2014	3.00	D. Popmintchev, M.C. Chen, C. Hernández-García, J.A. Pérez-Hernández, J. Sequeira, S. Brown, F. Dollar, B. Walker, Luis Plaja, M. Murnane, H.C. Kapteyn, T. Popmintchev. Ultrahigh-Efficiency High Harmonic Generation Driven by UV Lasers, CLEO: QELS_Fundamental Science. 09-JUN-13, . : ,		
08/21/2014	2.00	D. Popmintchev, C. Hernández-García, F. Dollar, C. Mancuso, J. A. Pérez-Hernández, M-C. Chen, A. Hankla, X. Gao, B. Shim, A. Gaeta, M. Tarazkar, D. Romanov, R. Levis, J. Gaffney, M. Foord, S. Libby, A. Jaron-Becker, A. Becker, L. Plaja, M. M. Murnane, H. C. Kapteyn, T. Popmintchev. BrightHighOrder Harmonic Generationin aMultiplyIonizedPlasmaupto the Water Window, OSA Conference on Lasers and Electro-Optics. 12-JUN-14, . : ,		
TOTAL:		2		
Number of P	eer-R	eviewed Conference Proceeding publications (other than abstracts):		
(d) Manuscripts				
Received		Paper		
08/21/2014	21/2014 1.00 F. Martin, M. M. Murnane, P. Ranitovic, C. W. Hogle, P. Riviere, A. Palacios, XM. Tong, N. Toshima, A. Gonzalez-Castrillo, L. Martin, H. Kapteyn. Attosecond vacuum UV coherent control of molecular dynamics, Proceedings of the National Academy of Sciences (01 2014)			
TOTAL:		1		
Number of Manuscripts:				
Books				
Received		Book		

TOTAL:

TOTAL:

Patents Submitted

Method for phase-matched generation of coherent VUV, EUV, and x-ray light using VUV-UV-VIS lasers, US Provisional Patent 61873794

(2013)

Patents Awarded

Awards

2015 Honorary Degree of Doctor of Science, National University of Ireland

2015 Elected to Member, American Philosophical Society

2015 Honorary Degree of Doctor of Science, University College Dublin

2015 Honorary Degree of Doctor of Science, Trinity College Dublin

2014 NSF Graduate Fellowship (David Couch, graduate student)

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2013 ACS Ahmed Zewail Award in Ultrafast Science and Technology (David Jonas)

2013 Honorary Member, Royal Irish Academy (Murnane)

2012 Willis Lamb Award for Laser Science and Quantum Optics (shared, Margaret Murnane and Henry Kapteyn)

2012 Chair, President's Committee for the US National Medal of Science (2012-2014) (Murnane)

Graduate Students				
NAME	PERCENT_SUPPORTED	Discipline		
Craig Hogle	1.00			
Austin Spencer	0.30			
Daniel Hickstein	1.00			
FTE Equivalent:	2.30			
Total Number:	3			

Names of Post Doctorates

NAME	PERCENT_SUPPORTED	
William Peters	1.00	
FTE Equivalent:	1.00	
Total Number:	1	

Names of Faculty Supported				
NAME	PERCENT SUPPORTED	National Academy Member		
Margaret Murnane	0.00	Yes		
Henry Kapteyn	0.00	Yes		
David Jonas	0.00	Yes		
FTE Equivalent:	0.00			
Total Number:	3			
Names of Under Graduate students supported				
NAME	PERCENT_SUPPORTED			
FTE Equivalent: Total Number:				
Student Metrics This section only applies to graduating undergraduates supported by this agreement in this reporting period				
The number of undergraduates funded by this agreement who graduated during this period: 3.00				
The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields: 0.00				
The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields: 0.00				
Number of gra	aduating undergraduates who ach	ieved a 3.5 GPA to 4.0 (4.0 max scale): 0.00		
Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for				
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Education, Research and Engineering:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: 0.00

Names of Personnel receiving masters degrees

<u>NAME</u>

Total Number:

Names of personnel receiving PHDs

<u>NAME</u>

Total Number:

Names of other research staff

NAME

PERCENT_SUPPORTED

FTE Equivalent: Total Number:

Sub Contractors (DD882)

Inventions (DD882)

See Attachment

Scientific Progress

Technology Transfer

FINAL REPORT for W911NF-12-1-0436 (ARO proposal number: 60607-PH)

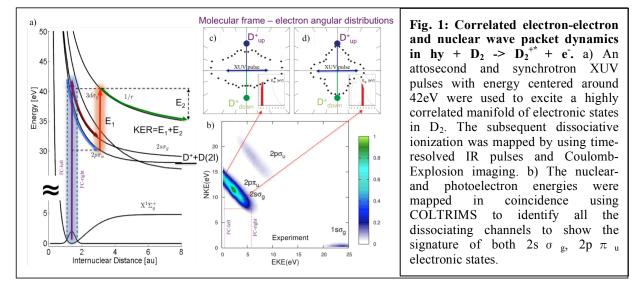
Bright Coherent Optical Waveforms from the Infrared to the VUV for Manipulation and Detection of Molecules Margaret Murnane, David Jonas and Henry Kapteyn JILA, Department of Physics, Department of Chemistry, University of Colorado at Boulder

ARO Program Manager: DOD Army ARO, Richard.Hammond@us.army.mil

Scientific Accomplishments: Four exciting advances resulted from this award -

<u>1. Revealing the Role of Electron-Electron Correlations during the Dissociation of Highly</u> Excited D_2^+ using Attosecond XUV Pulses [1]

Understanding electron-electron correlation in matter ranging from the atoms to solids represents a grand challenge for both experiment and theory. For experiment, the challenge is that in real molecular and materials systems, electron-electron interactions occur on attosecond time scales on up. Fortunately, advances in generating attosecond pulses of ionizing radiation are providing new capabilities for uncovering these electron-electron correlations. In our work we used attosecond pulse trains of extreme ultraviolet radiation to coherently populate and probe highly excited states of D_2^+ that undergo rapid Coulomb-explosion. By using electron-ion coincidence 3D momentum imaging coupled with advanced ab-initio simulations, we find that contrary to past understanding, the presence of correlations between the two electrons in D_2 can populate throughout the entire dissociation process, demonstrating that the entire Coulomb explosion occurs in a strong non-Born Oppenheimer regime. The good agreement between theory and experiment demonstrates that electron-electron correlations can play an important role even in the simplest of molecular systems.

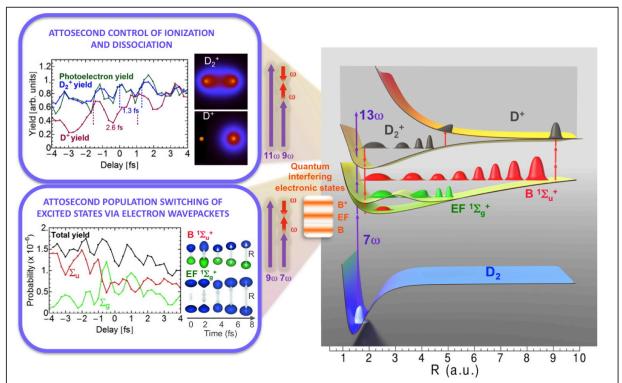


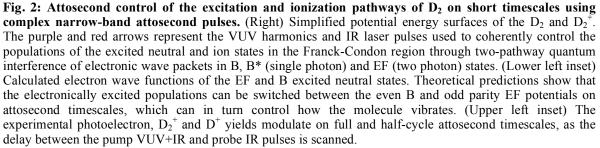
Specifically, we probed the Coulomb explosion of deuterium (D₂) following single-photon ionization by a 42 eV extreme ultraviolet (XUV) pulse, to make a remarkable and unexpected observation. Photoionization of D₂ leaves most molecules in the D₂⁺ ion ground state. However, some fraction of the molecules undergo a shake-up excitation, where a second electron is excited simultaneously during photoionization. These excited state D₂^{+*} molecules dissociate, with characteristic energies and timescales that are accessible in experiment, and that are only now fully theoretically tractable. By using a time-delayed infrared probe pulse combined with electron-ion coincidence imaging techniques, we can map the energy and angular distributions of the molecular fragments. When combined with advanced ab-initio calculations that include the coupled nuclear and electron-electron motions, this allows us to make two surprising findings.

First, the shake-up dynamics is not dominated by excitation to the $2p\pi_u$ state—as has been identified previously and as might be expected from a dipole s \rightarrow p transition. Rather, a hybrid state dominated by $2s\sigma_g$ is populated as a result of strong electron-electron correlations in the photoionization process. Second, throughout the Coulomb explosion, the characters of the $2p\pi_u$ and $2s\sigma_g$ states are mixed due to strong electron-electron correlations, demonstrating that the entire Coulomb explosion occurs in a non-Born Oppenheimer regime. This imprints a unique signature on the energy and angular distribution of the molecular fragments.

2. Attosecond VUV Coherent Control of Molecular Dynamics [2]

A breakthrough showing that that we can precisely control molecular dynamics on both nuclear (i.e. femtosecond) and electronic (i.e. attosecond) timescales to control the electronic state and outcome of a reaction was published in PNAS. By using VUV light pulses that are tunable in wavelength with adjustable attosecond time structure, we show that it is possible to switch population between electronic excited states, and use this ability to select specific pathways for ionization or dissociation of a molecule. This results represents a milestone advance because almost two decades after attosecond physics was demonstrated, attosecond pulses did not match the (VUV) wavelengths and (~<eV) spectral features involved in molecular quantum state control.





Specifically, we used attosecond pulse trains from a comb of VUV harmonics to steer the excited states and dissociation pathways in the simplest molecule - deuterium. We took advantage of interfering electronic wave packets in the excited neutral and singly-ionized molecule to switch the excited electronic state on attosecond timescales, and coherently guide the nuclear wave packets to dictate the way a neutral molecule vibrates. This also allowed us to steer and manipulate the ionization channels. The observed richness and complexity of the dynamics, *even in this very simplest of molecules*, is both remarkable and daunting, and presents intriguing new possibilities for state-selective chemistry. Finally, through advanced theory, we succeeded in observing and rigorously modeling multi-scale quantum control in a molecule for the first time.

We note that our results represent the first successful scheme for manipulating a chemical reaction using attosecond capabilities. A large component of the attosecond community is increasingly doctrinaire in focusing on isolated attosecond pulses as a requirement to capture the fastest dynamics. Much of this work has evolved into studies that place priority on measuring the shortest possible timescales without a clear physical question, or indeed without any particular connection to new or consequential physical phenomena. Our paper shows where these fast timescales can be relevant to the *outcome* of a physical process, but to do this requires electronic excitation in a molecule with precise (i.e. attosecond) timing and multi-state selectivity. Our work shows this requires using more complex narrow-band attosecond pulse trains, not broadband attosecond pulses.

<u>3. Attosecond Coherent Control of Single</u> and Double Photoionization in Argon [3]

This work is the first to apply quantum control techniques to double photoionization, which is a fundamental process where a single, high-energy photon ionizes two electrons simultaneously from an atom. To extend coherent control concepts to higher photon energies and shorter time scales, and access core-hole, inner-valance or doubly-excited systems autoionization. where Auger decay. interatomic Coulomb decay, and other ultrafast processes play a role, ultrafast pulses at high photon energies are required. Tabletop high harmonics represent a unique light source that is perfectly synchronized to the driving laser, which can be used to manipulate the fastest electron and molecular dynamics in matter.

In our work we showed that by spectrally separating a comb of high harmonics into VUV and XUV components, we can coherently populate Rydberg states of Ar and Ar^+ , close to the single and double ionization thresholds. Then, in the presence of two IR fields, one co-propagating and phase-locked with the VUV/XUV pump pulse and another that is time-delayed relative to the combined VUV/XUV/IR pump pulse, we show that we can coherently control both Ar^+ and Ar^{++} total yields using

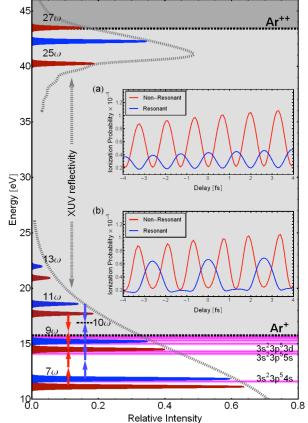


Fig. 3: Schematic showing the relevant energy levels for coherently controlling both single and double ionizion pathways in Ar. The tuning range of the harmonics is also shown. The inset shows the ion yield difference when controlling single ionization using quantum interferences alone in (a), or a combination of quantum and optical interferences in (b).

two-quantum-pathway electron wave packet interference processes. Furthermore, by tuning the energy of the VUV/XUV photons around different resonant Ar and Ar^{+*} states, we demonstrate an ability to fine-tune the phases of the optical and quantum interferences produced by the combined action of the IR laser and VUV/XUV harmonic fields. This work demonstrates for the first time that attosecond coherent control methods can manipulate double ionization processes on attosecond time scales, where electron-electron interactions play an important role. Double photoionization is a fundamental mechanism where a single photon can ionize two electrons simultaneously from an atom, providing insight into electron/electron dynamical correlation processes. Understanding and coherently controlling these correlations in simple atoms and molecules, where theoretical models are possible, will help develop the more advanced concepts necessary to control ultrafast dynamics in complex molecular systems or novel, strongly-correlated materials.

4. Chemical dynamics relevant to ionospheric chemistry, interstellar processes and highenergy materials [4, 5]

Molecular studies of internal conversion of electronic energy into nuclear motion have, until recently, largely focused on the first one or two excited states. Higher excited states, near 7-10 eV, have been little-explored due to the lack of good experimental approaches - but are critical to ionospheric chemistry, interstellar processes and high-energy materials. Dynamics in this energy range may be expected to exhibit very fast internal conversion, depositing enough vibrational energy to cause molecular dissociation on timescales faster than the validity regime of statistical theories. To probe these processes for the first time on the few-femtosecond timescales on which they occur, we performed time-resolved photoelectron-photoion coincidence (PEPICO) experiments on acetone (which is a prototype for organic molecules in the atmosphere) and methyl azide (a high energy density molecule). In these experiments an 8 eV pump pulse creates a highly excited state in the molecule, and a time-delayed 3.1 eV probe pulse ionizes the excited electron. To untangle the dissociation pathways, we detect the full 3D momentum vectors of the departing electron and of any ionic fragments produced from excess nuclear motion in the cation.

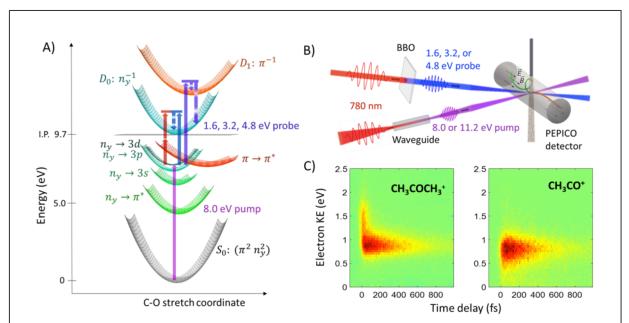


Fig. 4: a) An energy level diagram illustrating highly excited states of acetone and the two lowest cation states, with arrows indicating various pump-probe pathways investigated. b) An illustration of HHG upconversion techniques and PEPICO detection. c) Photoelectron energy vs. time plots in coincidence with acetone molecular ion (left) and the acetyl fragment ion (right).

This new capability allows us to uncover the correct dissociation pathways for highly-excited molecules. In acetone we re-assign the 8 eV state as a Rydberg-valence mixed state between $(n_y \rightarrow 3p)$ and $(\pi \rightarrow \pi^*)$, which internally converts to lower Rydberg states with a time constant of 340 fs. This mixing produces a non-separable electronic wavefunction, which can frustrate typical photoelectron imaging experiments and produce incorrect state assignments. The flexibility of our waveguide-based low-order harmonic generation allows us to measure 1-photon ionization with the 7th and 9th harmonics under identical experimental conditions as our pump-probe experiments, providing reliable identification by a comparison between 1 and 2-photon processes and access to multiple cation states.

In methyl azide we find Rydberg-Rydberg mixing between series converging to different ion states: $(HOMO \rightarrow 3p)$ and $(HOMO - 1 \rightarrow 3s)$. This mixing opens up relaxation pathways which are otherwise symmetry-forbidden, and represents an unanticipated phenomenon in the ultrafast dynamics of low Rydberg molecules. This mixed Rydberg state undergoes fast (~20 fs) wavepacket motion before undergoing a surprisingly quick (~25 fs) internal conversion. Again, the unambiguous assignment of the mixed electronic state required access to both relevant cation states as well as comparison to 1-photon ionization data provided by high harmonic generation.

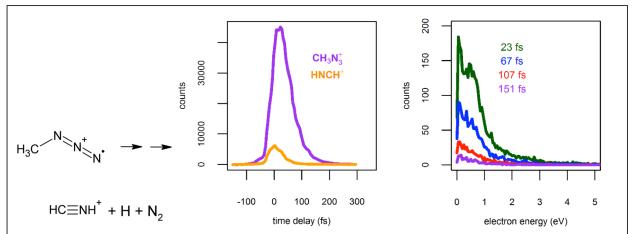


Fig. 5: After ionization, the methyl azide ion may undergo a sequential 3-body breakup. In our 8.0 eV pump - 1.6 eV probe experiment, we measure the time-dependent branching ratios, revealing how the excited state dynamics influence the post-ionization breakup. Time-dependent photoelectron spectra taken in coincidence with the fragment reveal the mixed character of the excited electronic state.

Publications from ARO Support

- P. Ranitovic, L. Martin, R. Y. Bello, C. W. Hogle, A. Palacios, X. M. Tong, J. L. Sanz-Vicario, T. Jahnke, M. Schöffler, R. Dörner, Th. Weber, F. Martín, H. C. Kapteyn, M. M. Murnane, "Revealing the Role of Electron-Electron Correlations during the Dissociation of Highly Excited D₂⁺ using Attosecond XUV Pulses," in preparation (2016).
- P. Ranitovic, C. W. Hogle, P. Rivière, A Palacios, Xiao-Min Tong, N. Toshima, A. González-Castrillo, L. Martin, F. Martín, M.M. Murnane, H.C. Kapteyn, "Attosecond VUV Coherent Control of Molecular Dynamics", PNAS 111 (3), 912-917 (2014).
- 3. C. W. Hogle, X. M. Tong, L. Martin, M. M. Murnane, and H. C. Kapteyn, P. Ranitovic. "Attosecond Coherent Control Processes in Single and Double Photoionization of Argon", Physical Review Letters **115**, 173004 (2015).
- 4. D. Couch, W. Peters, M.M. Murnane and H.C. Kapteyn, "Deep-UV excited states of acetone probed using angleresolved electron-ion coincidence detection," in preparation (2016).
- 5. W. Peters, D. Couch, M.M. Murnane and H.C. Kapteyn, "Angle-resolved PEPICO imaging of dissociative ionization of methyl azide and methylenimine using a tabletop high harmonic light source," in preparation (2016).

Honors during grant

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