Final Report for Grant Number: DE- FG02-10ER16146

This grant supported research in basic atomic, molecular and optical physics related to the interactions of atoms and molecules with photons and electrons. The duration of the grant was the 5 year period from 4/1/2010 – 10/31/2015. All of the support from the grant was used to pay salaries of the PI, graduate students, and undergraduates and travel to conferences and meetings. The results were in the form of publications in peer reviewed journals. There were 20 peer reviewed publications over these 5 years with 2 of the publications in *Physical Review Letters* and 1 in *Nature*; all of the other articles were in respected peer reviewed journals (Physical Review A, New Journal of Physics, Journal of Physics B ...).
I will discuss the results for the periods of time relevant for each year of the ~5 year grant period.

<u>Year 1</u>

Decay Dynamics following Core Photoionization of Methane

1. Overview:

Methane is one of the simplest polyatomic molecules, and as such provides a useful laboratory to study molecular dynamics. Its complexity is sufficient for exhibiting interesting decay behaviors and for challenging theory, but perhaps just at the boundary so there is great promise for expanding our fundamental understanding of how such complex systems can evolve over short time scales. Methane is a neon analogue with 10 electrons, two of which are deeply bound in the core of the carbon atom. Prompt photoionization of one of these C(1s) electrons leads to a highly excited molecular cation that has valence electronic structure nearly identical to the neutral ground state. We have just begun to explore how this state decays and to investigate the mechanisms that lead to the many dissociative pathways.

Our experiment uses the standard COLTRIMS approach, where we measure in coincidence up to four particles resulting from the photoionization and subsequent decay (one photoelectron and up to three ions). The initial photoionization reaction is given by

$$CH_4 + h\nu(>300\text{eV}) \rightarrow C(1s^{-1})H_4^{+*} + e^-_\gamma \text{(initial photoionization)}.$$

The dipole absorption of the photon promotes the essentially atomic C(1*s*) electron into a continuum *εp* state that can be strongly influenced by the molecular potential. Following this photoionization, the molecule can decay through multiple pathways, which we organize here into groups corresponding to three of the final fragment states (in bold) that we have measured in the experiment:

(1) $C(1s^{-1})H_4^{+*} + e^-_\gamma \rightarrow \mathbf{CH_3^+ + H^+} + e^-_\gamma + e^-_{Auger}$ (Auger, dication, $[H^+,CH_3^+]$)
(2) $C(1s^{-1})H_4^{+*} + e^-_\gamma \rightarrow \mathbf{CH_2^+ + H_2^+} + e^-_\gamma + e^-_{Auger}$ (Auger, dication, $[H_2^+,CH_2^+]$)
(3) $C(1s^{-1})H_4^{+*} + e^-_\gamma \rightarrow \mathbf{CH_2^+ + H^+ + H^+} + e^-_\gamma + 2\, e^-_A$ (*Double Auger* , trication, $[H^+, H^+,CH_2^+]$)

There are additional dissociative states, but we will limit the current presentation to cases (1) and (3).

2. Two-body breakup, $[H^+, CH_3^+]$:

The first step in understanding this decay mode is to see whether or not the axial recoil approximation holds, i.e. that measurement of the H^+ fragment momentum vector is equivalent to measurement of a bond axis. This will be the case for those events where the dissociation time is substantially faster than either the rotation period of the molecule or, more importantly, the timescales of conformation changes during the decay. We show data here for the case of 2.4 eV photoelectron energy.

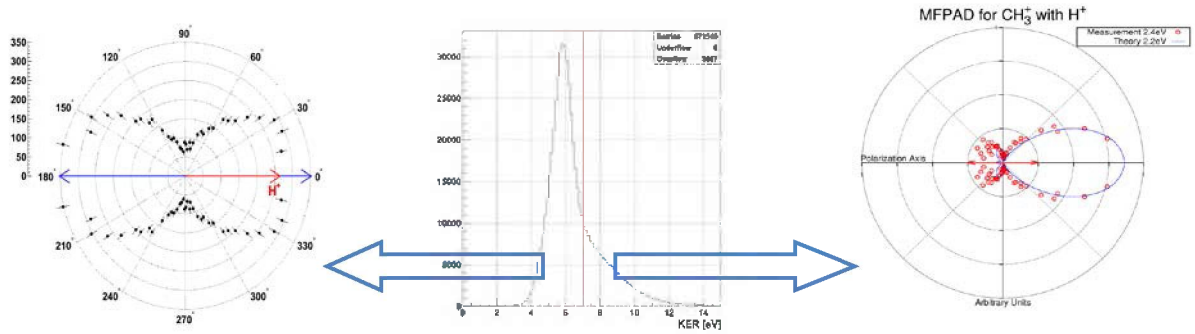

Figure1: Kinetic energy release for the $[H^+,CH_3^+]$ channel (center) corresponding to axial-recoil breakdown (left) or axial-recoil valid (right) molecular frame photoelectron angular distributions with theory.

The center panel of figure 1 shows the fragment kinetic energy release for this two body breakup channel, which has a fairly sharp peak at ~5.5eV and a long energetic tail. If we gate the data on the lower energy peak we get the molecular frame photoelectron angular distribution (MFPAD) shown to the left, whereas gating on the tail leads to the MFPAD shown to the right. In both cases we have confined the H^+ momentum vector to the positive horizontal axis along the photon polarization. The interesting result here is that on the left you get what is a nearly dipole distribution for the photoelectron, which is what you would expect for a randomly oriented molecule or breakdown of axial recoil. On the right, however, you see a strongly asymmetric MFPAD that is consistent with calculations by C. Trevisan and the group of C.W. McCurdy and T.N Rescigno.

The above result is quite fortunate, because we are able to identify at least two decay pathways: one prompt dissociation where axial recoil holds and a different pathway where axial recoil breaks down. This provides us a tool for exploring dynamics; by measuring the amount of axial-recoil breakdown when comparing the left and right spectrum and consulting with theory, we plan to determine the specific decay channels and associated timescales. This kind of information will be critical in interpreting future experiments with ultra-fast sources such the LCLS or the envisioned NGLS where event mode experiments might be possible.

This data set is enormously rich, because we can look at MFPADs associated with any of the many molecule orientations or tied to specific dissociative states. At the time of this writing, we have developed some preliminary theory-based interpretations about which channels are populating which dissociative states, and we anticipate new detailed results soon. In the meantime, we have also taken the next step in this type of measurement, where we have measured (as far as we know) the first fully three-dimensional molecular frame photoelectron angular distributions. These results are presented below.

2. Three-body breakup, [H^+, H^+, CH_2^+]:

As mentioned in the overview, methane is an electronic analogue to the neon atom. Our previous work on correlation effects in the core-photoionization of neon included an investigation of the double-Auger channel which leads to a triply charged Ne^{3+}. This channel is not only possible, but probable because there are no cascade pathways for a core-hole to decay to Ne^{3+}. We have found the exciting result that this appears to also be the case in methane. Furthermore, we have confirmed that when double Auger occurs in the molecule, the triply charged molecular ion is extremely unstable and dissociates rapidly along the bond axes. We show data here for the case of 4.5 eV photoelectron energy.

Ions: This axial-recoil result is illustrated in figure 2. The top left panel show the momentum distribution of the two protons in the plane defined by their momentum vectors and that of the CH_2^+ ion, which defines the positive horizontal axis. The upper half of the figure shows the first measured proton in the plane it forms with the CH_2^+ ion, and the lower half shows the second measured proton in its plane with the heavy fragment. The large exterior islands correspond to protons ejected roughly along the bond axes (shown in red) but distorted from the CM to lab frame change and by "post-collision interaction" with the doubly charged heavy fragment. We currently interpret that the interior islands arise from the coincidence of a light and heavy ion with a random hit corresponding to a neutral, which is demonstrated in the lower left panel where we've gated on the island for the first hit and get a roughly random distribution for the second. We believe these events correspond to the [H^0, H^+, CH_2^+] decay pathway, but have yet to purse analysis of this channel.

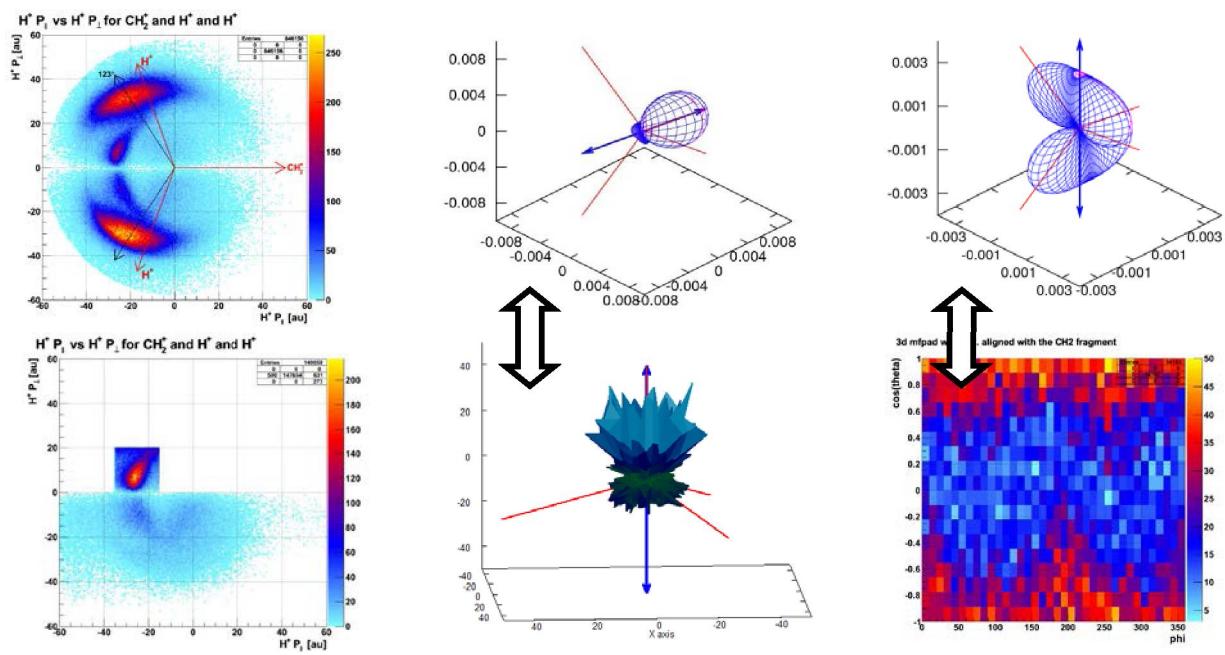

Figure2: Fragment ion momenta in for the three body breakup (left) and three-dimensional MFPADS (mid & right).

Electrons: The mid-top and mid-bottom panels show theory (top) and experiment (bottom), where electrons have a propensity to emerge along a bond aligned with the polarization axis. The statistics are limited, but the strong asymmetry is evident. Even more interestingly, the right panels show

a comparison between theory (top) and experiment (bottom) with experiment plotted as a density plot in φ and cos(θ) to plainly emphasize the kidney shaped structure arising when the polarization axis bisects a bond. We are currently pursing clearer and more enlightening comparisons between these recent experimental results and theory. In brief, we believe there is a strong potential for 3D-MFPADS to become a powerful tool in furthering our understanding of molecular structure and dynamics.

Year 2

During the project year we participated in **two beamtimes** at the Advanced Light Source. The first took place September 14th – 25th 2011, and included three participants from Auburn: PI Landers and students Joshua Williams and Marcus Honig. The second took place March 14th – 26th, and included five participants from Auburn: PI Landers and students Joshua Williams, Averell Gatton, Dylan Reedy, and Cory Nook. This travel and travel by the PI to the 2011 contractors meeting were supported by the present grant. Travel to the 2012 DAMOP by the PI and one student (Williams) will also be supported during the current project year.

We highlight below the most exciting result from this year's work, which is the core photoionization of methane. This work is the dissertation project of Joshua Williams and has been published as two separate articles to Physical Review Letters and Journal of Physics B. Additional Auburn-led results during the current funding year include the recently measured ion-anion pair production in the photo-absorption of H_2, work for which a manuscript is in preparation.

(1): Decay Dynamics following Core Photoionization of Methane

Methane is one of the simplest polyatomic molecules, and as such provides a useful laboratory to study molecular dynamics. Its complexity is sufficient for exhibiting interesting decay behaviors and for challenging theory, but perhaps just at the boundary so there is great promise for expanding our fundamental understanding of how such complex systems can evolve over short time scales. Methane is a neon analogue with 10 electrons, two of which are deeply bound in the core of the carbon atom. Prompt photoionization of one of these C(1s) electrons leads to a highly excited molecular cation that has valence electronic structure nearly identical to the neutral ground state. We have just begun to explore how this state decays and to investigate the mechanisms that lead to the many dissociative pathways.

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$$C(1s^{-1})H_4^{+}{*} + e^{-}_{\gamma} \rightarrow \mathbf{CH_2^{+} + H^{+} + H^{+}} + e^{-}_{\gamma} + 2\,e^{-}_{A} \text{ (\textit{Double Auger}, trication, } [H^{+}, H^{+}, CH_2^{+}])$$
$$C(1s^{-1})H_4^{+}{*} + e^{-}_{\gamma} \rightarrow \mathbf{CH_3^{+} + H^{+}} + e^{-}_{\gamma} + e^{-}_{\text{Auger}} \text{ (Auger, dication, } [H^{+}, CH_3^{+}])$$

There are additional dissociative states, but we will limit the current presentation to the cases above.

Two-body breakup, [H^+, CH_3^+] (results published in JPB):

In addition to the imaging result discussed above, we find that we are able to gain some understanding into the decay dynamics. For example, in the [H^+, CH_3^+] decay channel of the dication, we find that the proton emission angle corresponds to the ground state bond angle only for certain values of the kinetic energy release. In a manuscript recently submitted to J. Phys. B, we interpret this result with the support of theoretical calculations to be due the channel arising through two different decay mechanisms. The lower energy decay mode distorts the molecule, and removes the correlation between bond angle and emission angle. The result is simple dipole distribution where the influence of the molecular potential on electron emission is lost in the measurement.

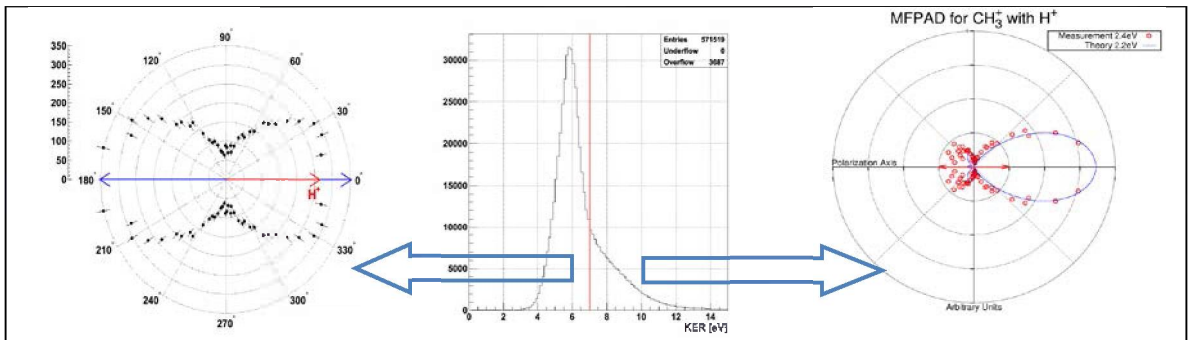


Figure 2: Kinetic energy release for the [H^+,CH_3^+] channel (center) corresponding to axial-recoil breakdown (left) or axial-recoil valid (right) MFPADs. The right panel shows comparison with theory.

Three-body breakup, [H^+, H^+, CH_2^+] (results published in PRL):

We have confirmed that when double Auger occurs in the molecule, the triply charged molecular ion (trication) is extremely unstable and dissociates rapidly along the bond axes, allowing us to fully orient the molecule in three dimensions. We are then able to observe the electron emission in the molecule frame, in the form of molecular frame photoelectron angular distributions (MFPADs). We show data here for the case of 4.5 eV photoelectron energy.

The most exciting outcome of these measurements is that when we integrate over the polarization axis in the molecule frame, we can fully isolate the influence of the molecular potential on the photoelectron emission. When we do this, we find the striking result that the photoelectrons tend to emerge along the bond axes, effectively imaging the molecule (Fig 1 top left). Although this effect is predicted by complex Kohn variational calculations (McCurdy, Fig 1 bottom left), we still lack a simple model explanation. Furthermore, we find that even in the highly differential case of the molecule oriented relative to a specific polarization axis, we are able to see the complicated emission patterns predicted by the calculation (Fig 1 right).

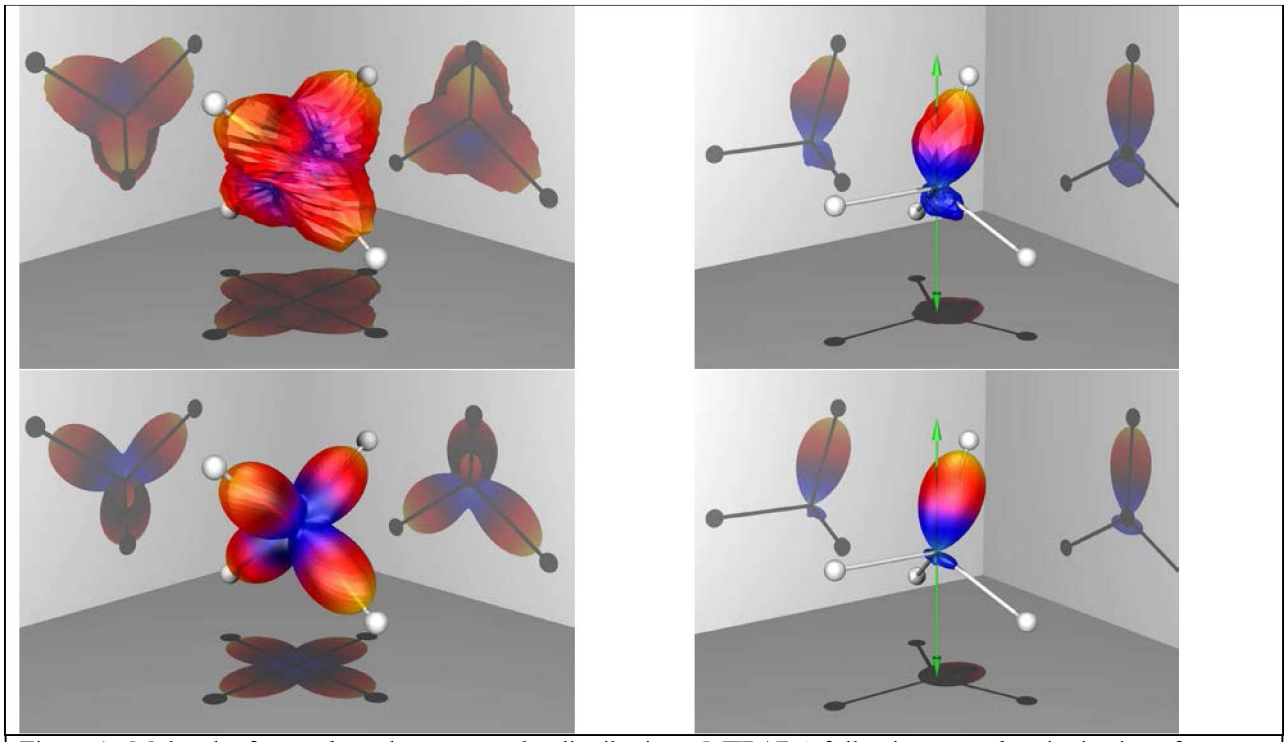
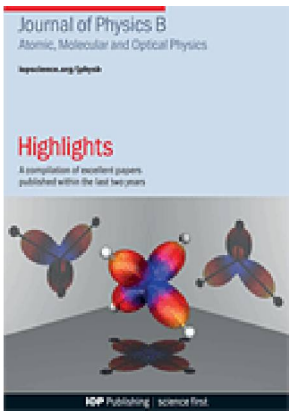


Figure 1: Molecular frame photoelectron angular distributions (MFPADs) following core photoionization of methane. *Top row*: ALS COLTRIMS experiment, *Bottom Row*: Complex Kohn variational calculations. *Left*: MFPAD integrated over polarization axis isolating influence of molecular potential. *Right*: MFPAD for a specific geometry demonstrating level of detailed agreement between experiment and theory.

Year 3

During the current project year we will have participated in **two beamtimes** at the Advanced Light Source. The first took place August 13th – 22nd 2012, and included three participants from Auburn: PI Landers and students Ali Moradmand and Marco Keling. The second will take place May 22nd – June 1st 2013, and will included three participants from Auburn: PI Landers and students Averell Gatton and James Sartor. This travel and travel by the PI to the 2012 contractors meeting were supported by the present grant. Travel to the 2013 DAMOP by the PI and two students (Moradmand and Sartor) will also be supported during the current project year.

Publication Highlights

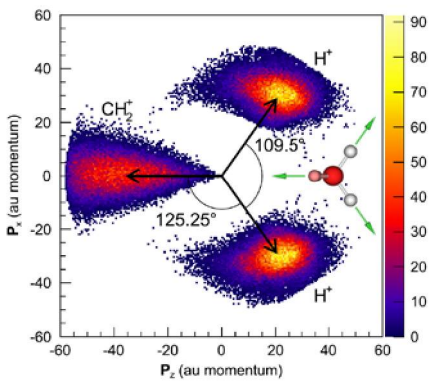


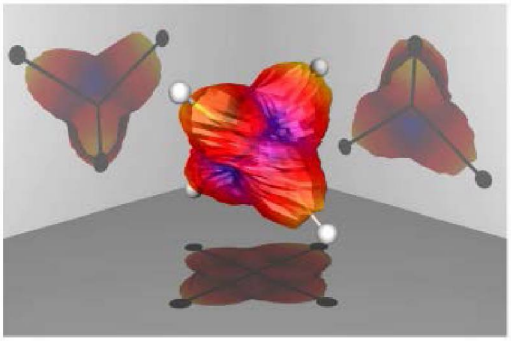
We had a strong year for high impact publications resulting from work initiated by the Auburn part of the international ALS-COLTRIMS collaboration. We published in Physical Review Letters our measurement that used three-dimensional coincident imaging to demonstrate that low energy photoelectrons ejected from the core of a methane molecule emerge along the bond axes.

In addition, two of our papers were featured in the "highlights 2012" for the Journal of Physics B (one featured on the cover at left). The first was an extension of the work published in PRL, where we showed that these imaging experiments allowed us to trace decay dynamics at an unprecedented level of detail. The second was a collaborative paper with Francis Robicheaux, where his calculation coupled with our new measurements explained a previously not-understood phenomenon in the angular correlation of electrons ejected from the same atom with very different energies. We summarize these three articles here.

(1) Imaging Polyatomic Molecules in Three Dimensions Using Molecular Frame Photoelectron Angular Distributions, *Phys. Rev. Lett.* **108,** 233002 (2012)

Here we demonstrated a method for determining the full three-dimensional molecular-frame photoelectron angular distribution in polyatomic molecules using methane as a prototype. Simultaneous double Auger decay and subsequent dissociation allowed measurement of the initial momentum vectors of the ionic fragments and the photoelectron in coincidence, yielding full orientation by observing a three-ion decay pathway, (H^+, H^+, CH_2^+). We found the striking result that at low photoelectron energies the molecule is effectively imaged by the focusing of photoelectrons along bond directions. The figures below show the momentum spectra of the ions (left) and the photoelectrons (right), both in the molecule frame.





(2) Probing the dynamics of dissociation of methane following core ionization using three-dimensional molecular-frame photoelectron angular distributions, *J. Phys. B: At. Mol. Opt. Phys.* **45** 194003 (cover, and highlight 2012)

Here we presented experimental measurements and theoretical calculations for the photoionization of CH_4 at the carbon K-edge. Measurements performed using cold target recoil ion momentum spectroscopy (COLTRIMS) combined with complex Kohn variational calculations of the photoelectron in the molecular frame verified the surprising result that the low energy photoelectrons effectively image the molecule by emerging along the bond axes. Furthermore, we observed a dynamic breakdown of axial recoil behavior in one of the dissociation pathways of the intermediate dication, which were interpreted using electronic structure calculations. This investigation was driven in large part by the observation of the strong influence of the ion fragment kinetic energies on the photoelectron angular distributions found in our initial analysis.

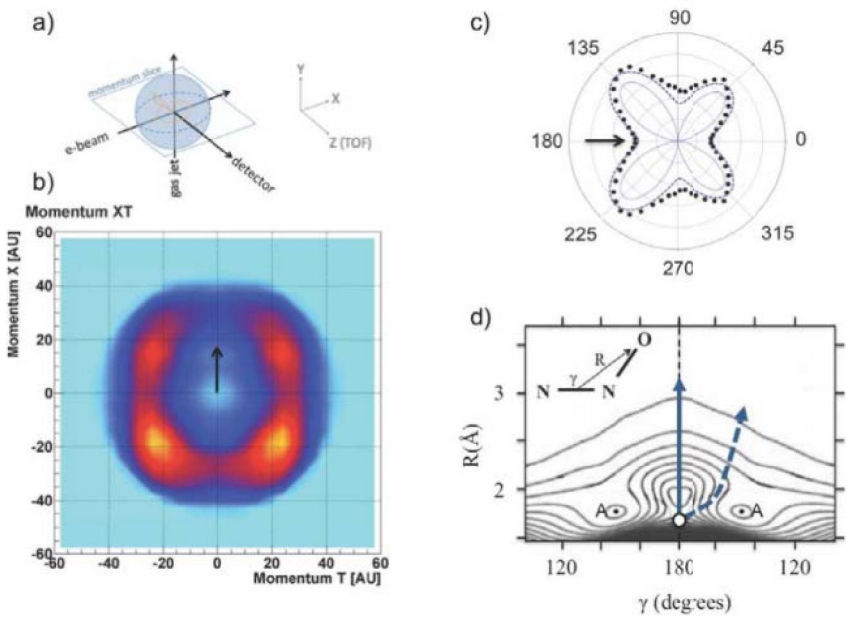
(3) Calculated and measured angular correlation between photoelectrons and Auger electrons from K-shell ionization, *J. Phys. B: At. Mol. Opt. Phys.* **45** 175001 (highlight 2012)

Here we repeated and improved measurements to provide experimental support to calculations performed by Francis Robicheaux, who applied a recently developed computational method to an experimental puzzle that involves a slow outgoing electron that is scattered by a high-energy Auger electron. Although the experiment seemed to be in a regime accurately described by classical mechanics, such classical calculations could not accurately model the angular distribution of the electron pair. Using the wavefunction from his calculations to generate the energy and angular distributions of the two electrons, we compared his results to our measurements performed at the Advanced Light Source. We obtained good agreement between the experiment and our quantum results, attributing the poor classical result to the small number of angular momenta in the wavefunction.

Year 4

Electron driven Molecular Dynamics:

We have performed a number of dissociative electron attachment measurements on small molecules in collaboration with Prof. Mike Fogle at Auburn, where we have used momentum imaging of the resulting anion in order to explore the dissociation dynamics. Specifically, we have employed a momentum imaging technique to measure the dissociative electron attachment to a number of small molecules including O_2, CO_2, and N_2O. By measuring the momentum distribution of the anion produced we are able to determine the kinetic energy release in the reaction as a function of emission angle over the full 4π solid angle of acceptance. This allows, for example, a comparison with theory that calculates the entrance amplitudes leading to the dissociative resonance being studied. We have found that the high angular resolution of our supersonic crossed-beam experiment has provided new insights into previous measurements of some systems, and has opened the door to fruitful collaboration with the LBNL theoretical (listed above) and experimental group (Belkacem and Slaughter) also funded within the DOE-AMOS program.

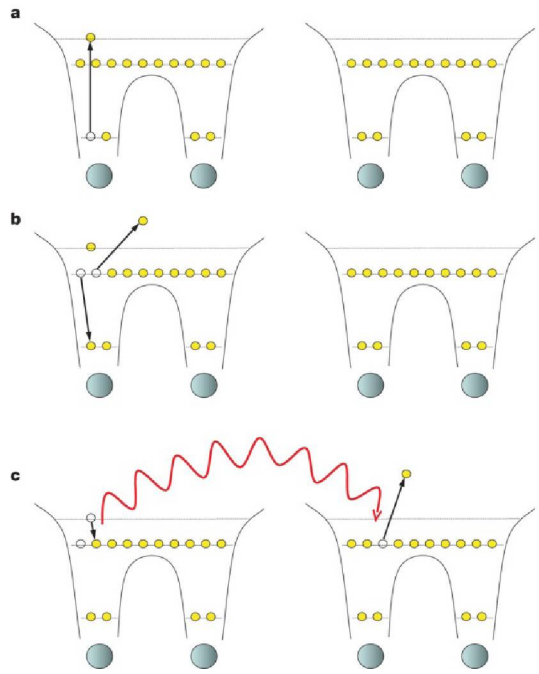


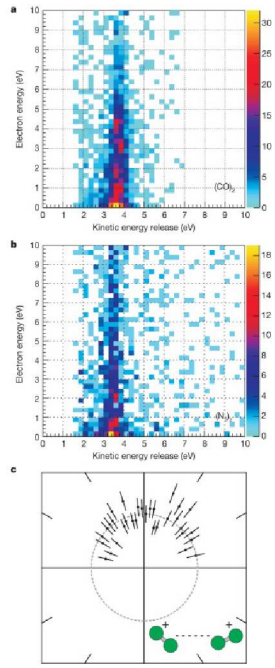
One such example is shown in the figure on the previous page from our recently published results for dissociative electron attachment (DEA) to N_2O at the 2.3eV shape resonance resulting in O^- fragments (Ref. 6). (a) the coordinate system for orienting dissociation events. (b) a weighted momentum space O− fragment distribution. From this data we can extract the angular distribution of fragments with respect to the incoming electrons, seen (c) and kinetic energy distribution (not shown here). (d) the potential energy landscape of the N2O− temporary negative ion state along with possible dissociation dynamics. The solid curve is a direct dissociation across a Renner-Teller reef with no significant bending dynamic, indicative of Π character. The dashed curve represents a significant bending upon dissociation, indicative of a Σ character.

Photoionization experiments at ALS:

Auburn has continued participating in the collaboration with the University of Frankfurt and LBNL groups in COLTRIMS experiments at the Advanced Light Source at LBNL. The most prominent example resulting from this joint effort is a work led by F. Trinter and T. Jahnke (Frankfurt) that was

published in *Nature*. It was recently suggested that inter-coulombic decay (ICD) can be triggered efficiently and site-selectively by resonantly core-exciting a target atom, which then transforms through Auger decay into an ionic species with sufficiently high excitation energy to permit ICD to occur. Here we demonstrated experimentally that resonant Auger decay can indeed trigger ICD in dimers of both molecular nitrogen and carbon monoxide. Our experimental confirmation of this process and its efficiency may trigger renewed efforts to develop resonant X-ray excitation schemes for more localized and targeted cancer radiation therapy.



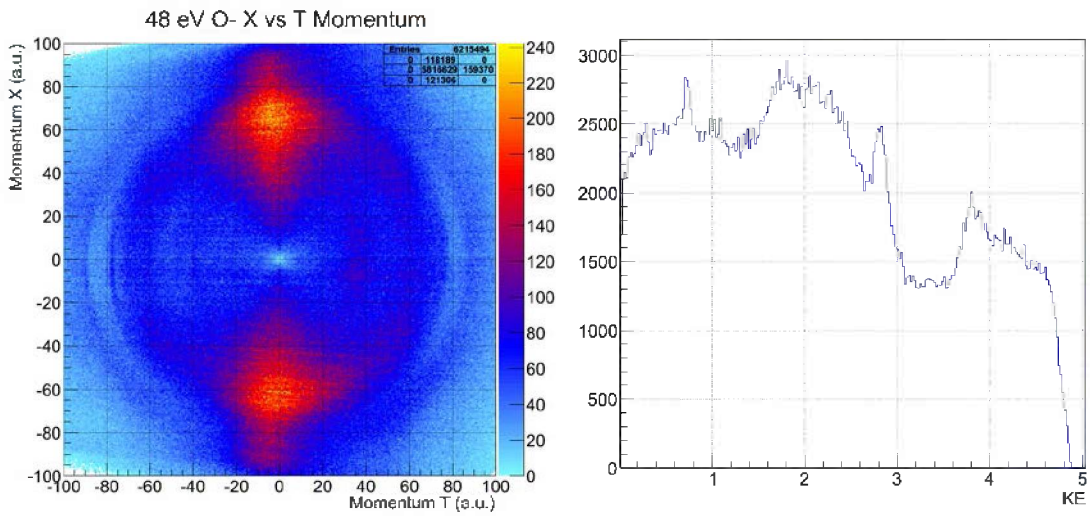


Previous Page Left: Shown is the series of events involved in resonant-Auger-driven ICD. a, One molecule (left) of the molecular dimer is core-excited. b, The core-excited state decays by a spectator Auger decay to a highly excited state of the molecular ion. c, ICD transfers the excitation energy to the molecular neighbour (right), where a low-energy ICD electron is emitted.

Previous Page Right: **a**, Kinetic energy release of $(CO)_2$ versus energy of one of the two electrons created by ICD after resonant excitation and subsequent Auger decay at a photon energy of 287.4 eV (Π* excitation of CO). The colour scale shows the intensity in counts. **b**, Same plot for $(N_2)_2$ recorded at a photon energy of 401.9 eV (Π* excitation of N_2). **c**, Emission direction of the Auger electron with respect to the molecular axis of the N_2 dimer (with statistical error bars). The dimer is oriented horizontally, as depicted by the green icon. The grey circle is a line to guide the eye, corresponding to isotropic emission.

Year 5

Electron driven Molecular Dynamics:

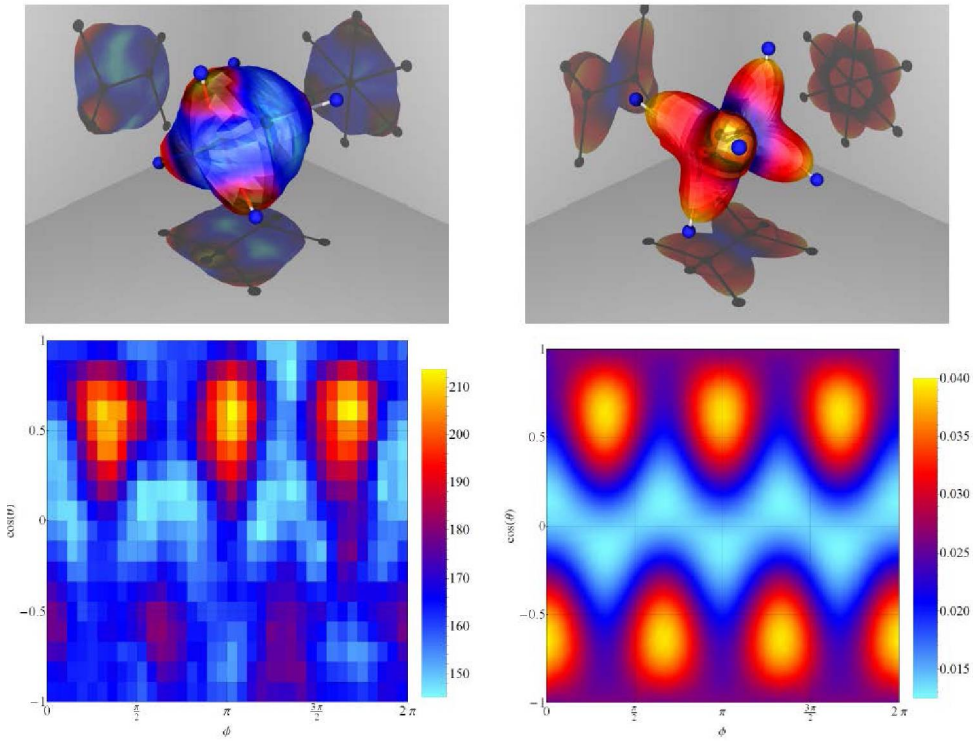


Left: O^- momentum distribution following interaction with a 48 eV electron (electron beam direction is up in the figure). Right: Associated Kinetic Energy distribution for the momentum distribution at left. Note the multiple sharp peaks, corresponding to excitations that decay through ion-anion pair production.

We have performed a number of dissociative electron attachment measurements on small molecules in collaboration with Prof. Mike Fogle at Auburn University, where we have used momentum imaging of the resulting anion in order to explore the dissociation dynamics. Recently, we have employed a momentum imaging technique to measure the electron impact anion production far above the dissociative ionization threshold for number of small molecules including O_2. By measuring the momentum distribution of the anion produced we are able to determine the kinetic energy release in the reaction as a function of emission angle over the full 4π solid angle of acceptance. We have found that the high angular resolution of our supersonic crossed-beam experiment has provided new insights into previous measurements of some systems, and we anticipate additional fruitful collaboration with the LBNL theoretical (listed above) and experimental group (Belkacem and Slaughter) also funded within the DOE-AMOS program.

One such example is shown in the figure on the previous page showing recent results for dissociative electron excitation at 48eV electron energy resulting in O^- fragments. Although well above the dissociative attachment energies, we find a substantial number of anions formed through excitations that decay through ion-anion pair production. Currently we are working to identify individual states populated, determine their relative production fractions, and individual angular distributions. Theory support for these experiments is greatly needed.

Photoionization experiments at ALS:



MFPAD for C_2H_6 at 4.5 eV above the C 1s threshold from analysis of the experimental data assuming the three protons in the breakup channel $C_2H_6 + h\nu \rightarrow H^+ + H^+ + H^+ +$ *neutrals* $+ 3e^-$ originated from the same carbon atom (left column) and complex Kohn calculations at 4.35 eV (right column).

Auburn has continued participating in the collaboration with the University of Frankfurt and LBNL groups in COLTRIMS experiments at the Advanced Light Source at LBNL. The most recent example resulting from this joint effort is a work led by Joshua Williams and the McCurdy group recently submitted to *Journal of Physics B*.

We measured molecular frame photoelectron angular distributions (MFPADs) in electron-ion momentum imaging experiments and compared these with complex Kohn variational calculations (LBNL, McCurdy group) for carbon K-shell ionization of CF_4, C_2H_6 (shown above) and $C_2H_2F_2$. While in ethane the polarization averaged MFPADs show a tendency at low energies for the photoelectron to be emitted in the directions of the bonds, the opposite effect is seen in CF_4. A combination of these behaviors is seen in difluoroethylene where ionization from the two carbons can be distinguished experimentally because of their different K-shell ionization potentials. Excellent agreement is found between experiment and theoretical calculations performed by Cynthia Trevisan and the LBNL theory group.

DOE BES Supported Publications 4/1/2010 – 10/31/2015

1. **Molecular frame photoelectron angular distributions for core ionization of ethane, carbon tetrafluoride and 1,1-difluoroethylene** A. Menssen, C.S. Trevisan, M.S. Schöffler, T. Jahnke, I. Bocharova, F. Sturm, N. Gehrken, B. Gaire, H. Gassert, S. Zeller, J. Voigtsberger, A. Kuhlins, A. Gatton, J. Sartor, D. Reedy, C. Nook, B. Berry, M. Zohrabi, A. Kalinin, A. Belkacem, R. Dörner, Th. Weber, A.L. Landers, T.N. Rescigno, C.W. McCurdy, J.B. Williams, (submitted to *J Phys B*)
2. **Hydrogen and fluorine migration in photo-double-ionization of 1,1-difluoroethylene (1,1-$C_2H_2F_2$) near and above threshold** B. Gaire, I. Bocharova, F. P. Sturm, N. Gehrken, J. Rist, H. Sann, M. Kunitski, J. Williams, M. S. Schöffler, T. Jahnke, B. Berry, M. Zohrabi, M. Keiling, A. Moradmand, A. L. Landers, A. Belkacem, R. Dörner, I. Ben-Itzhak, and Th. Weber, *Phys Rev A* **89** 043423 (2014).
3. **Photo-double-ionization of ethylene and acetylene near threshold** B Gaire, SY Lee, DJ Haxton, PM Pelz, I Bocharova, FP Sturm, N Gehrken, M Honig, M Pitzer, D Metz, H-K Kim, M Schöffler, R Dörner, H Gassert, S Zeller, J Voigtsberger, W Cao, M Zohrabi, J Williams, A. Gatton, D Reedy, C Nook, Thomas Müller, AL Landers, CL Cocke, I Ben-Itzhak, T Jahnke, A Belkacem, Th Weber, *Phys Rev A* **89** 013403 (2014).
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