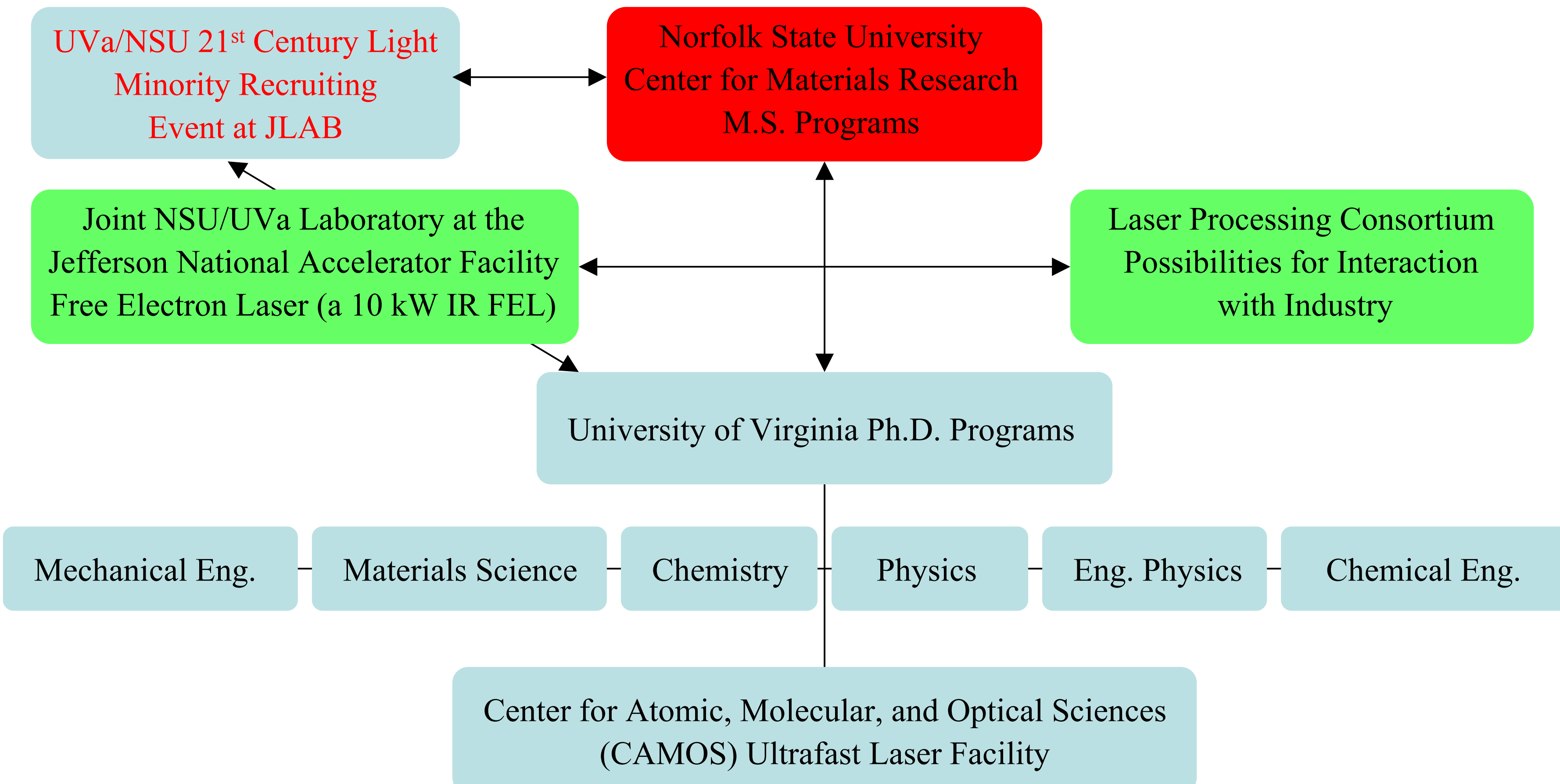
NSF: Integrative Graduate Education & Research Traineeship Program

Science and Engineering of Laser Interactions with Matter

- Partnering universities, a national lab, and industry to educate a graduate-level work[illegible]disciplinary challenges. multidisciplinary challenges.

- Emphasis on ultrafast laser interrogation and control of matter of all kinds.

SELIM Organization



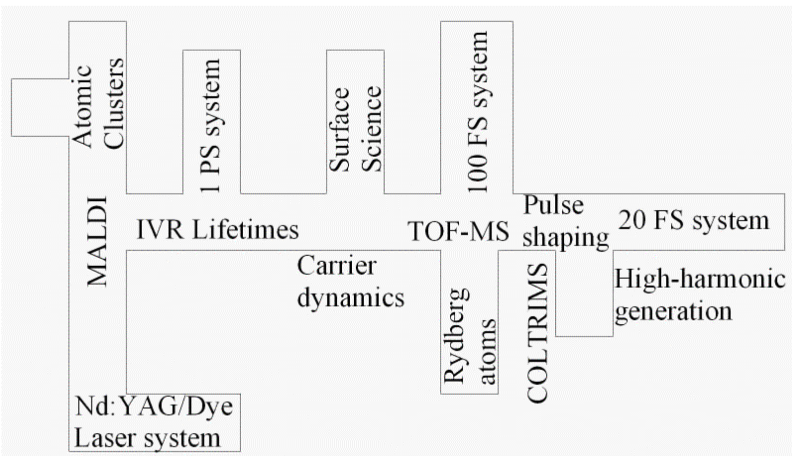
NSU Center for Materials Research

Optical Materials Development & Characterization

- Ultrafast & nanosecond lasers
- Organic molecular beam and Czochralski crystal growth facilities
- Extensive analytical facilities

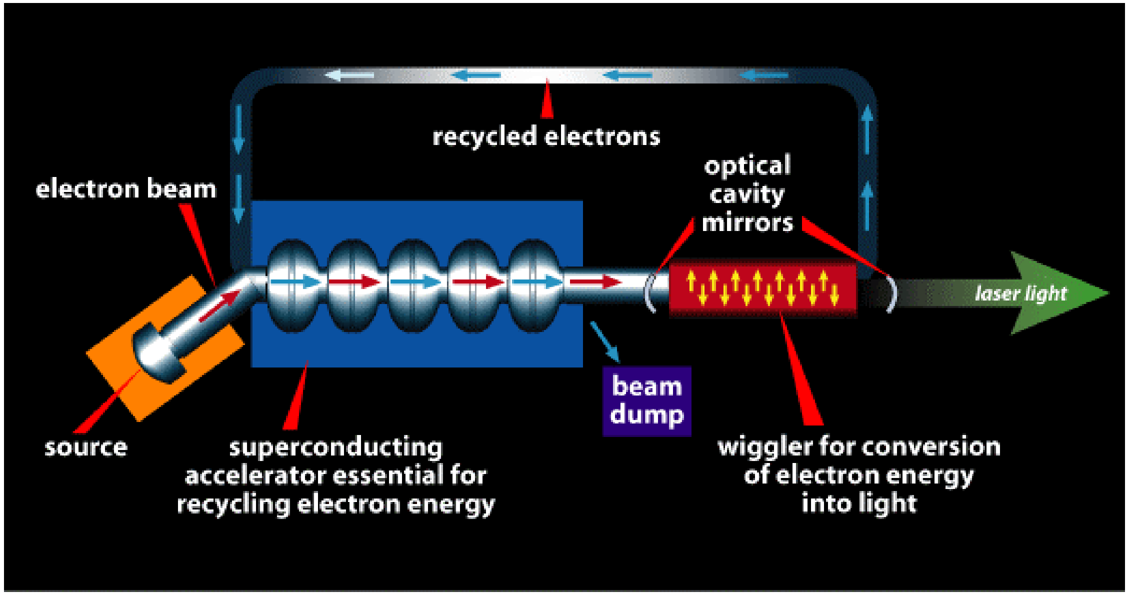
CAMOS Ultrafast Laser Facility

Overview of Laser Table Instrumentation

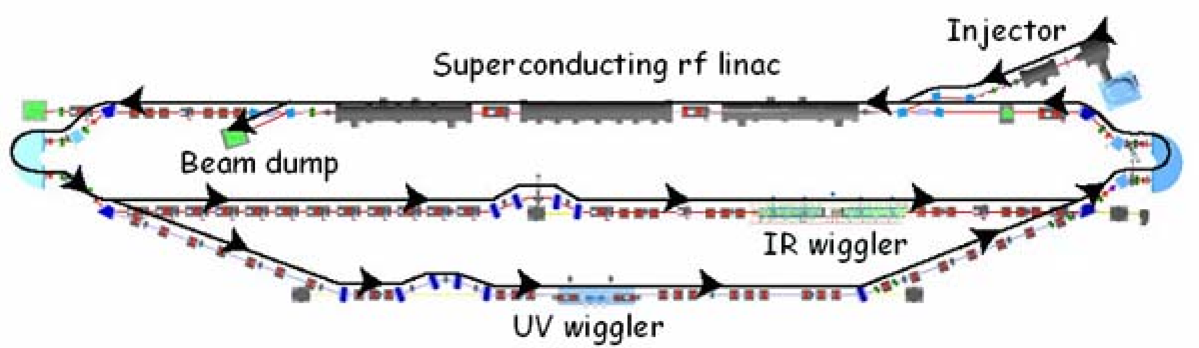


3000 ft^2 facility with 4 ultrafast amplified Ti:sapphire laser systems and a broad range of associated optical equipment and experimental stations.

JLAB Free Electron Laser



2 kW IR-FEL



<u>Upgrades</u>

10 kW IR-FEL

1 kW UV-FEL

Unique Ultrafast Laser Resources in Virginia

CAMOS Ultrafast Laser Facility

- Amplified Ti:Sapphire laser systems (800 nm)
- 2 mJ/pulse @ kHz; τ_p = 2 ps, 100 fs (2x), 30 fs (~.1 TW)
- 10 μm ≥ λ ≥ 200 nm; Soon, 30 fs harmonics to ~8 nm.

JLAB Free Electron Lasers (FEL)

- 10 kW IR FEL ('04); 1 kW UV FEL('06)
- 100 μJ/pulse @ 75 MHz; τ_p = 800 fs (~.1 GW)
- 14 μm ≥ λ_{ir} ≥ 1 μm, λ_{uv} ≥ 250 nm; Intense λ_{THz} ≥ 100 μm

Core SELIM Research

- Studies of ultrafast nuclear and electronic dynamics and energy flow in all states of matter.
- Coherence in quantum wavepackets, quantum entanglement, quantum information.
- High field laser-matter interactions.
- Spectroscopy development.
- Laser processing/deposition of materials.
- Optical materials development & characterization.

SELIM Education & Training

Fellows are supported for initial 2 years – subsequently by their advisors.

- All Fellows take "Introduction to SELIM" and "Research, Innovation, Entrepreneurship, and Ethics" courses.
- Most Fellows also take some of the following SELIM related courses:
  - **Optics (Phys 531), Photonics (Phys 532)**
  - **Lasers and Non-Linear Optics (Phys 822)**
  - **Ultrafast Laser Spectroscopy (Phys 826)**
  - **Surface Science (MSE 722)**
  - **Reaction Kinetics and Dynamics (Chem 722)**

SELIM Education & Training

- Monthly CAMOS Ultrafast Laser Facility science / scheduling / pizza meetings.
- Participation in Laser Processing Consortium Workshops & 21st Century Light Minority Recruiting Events at JLAB.
- Opportunities for development of multiple mentor research projects at UVa, NSU, and the JLAB FEL.
- Opportunities for “externships” working outside of academia.

Associates are not supported directly by the NSF IGERT grant but are encouraged to participate in all SELIM program elements.

Ph.Ds awarded by Departments <u>not</u> SELIM program.

SELIM Program

At UVa,

- 36 SELIM Fellows
- 18 SELIM Associates

At NSU,

- 9 SELIM Fellows

65 graduate students involved to date

Wide Range of SELIM Research:

Ultrafast Control of Electron Dynamics

→

Lab-on-a-Chip Medical Diagnostics

Bob Jones, Department of Physics

Research: Investigating the response of atoms and small molecules to intense short laser pulses and the use of coherent light to view and control quantum dynamics in atoms and molecules

Fellows:

Dan Pinkham, Joe Pole - Control of ultra-fast laser pulse shapes for manipulation of molecular orientation and alignment to investigate intense field fragmentation processes in small molecules and clusters.

Brett Sickmiller - Creation of sub-20 femtosecond VUV light pulses from intense near infrared light via high harmonic generation.

Mary Kutteruf – Spectroscopy of doubly-excited atoms in strong electric fields

Associates:

Jeremy Murray-Krezan, Lung Ko, Xiangdong Zhang, Peter Gee, Santosh Pisharody*, Jason Zeibel* - Manipulation of electronic wavepackets for probing and controlling coherent time-dependent processes in atoms

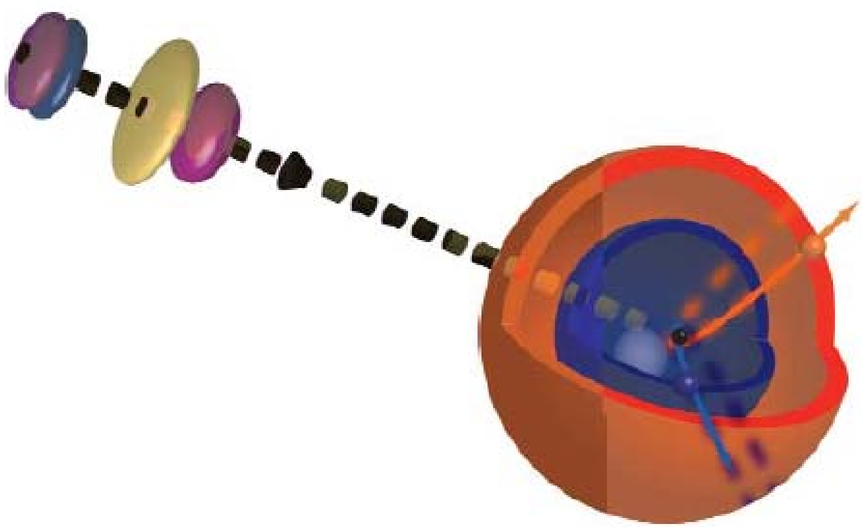
Merrick DeWitt*, Eric Wells* - Investigation of intense laser ionization and fragmentation in molecules and small clusters

*Past Associates and Fellows

PHYSICS

Pas de Deux for Atomic Electrons

C. R. Stroud Jr.



Dance of the electrons. In the experiment by Pisharody and Jones (1), a sequence of four laser pulses excites the two valence electrons into radial wave packets whose corresponding classical orbits are indicated. The outer electron (orange) is shown both in terms of one of its classical orbits and in terms of its radial wave packet. The electron excited into the lower energy orbit (blue) is shown in one of its classical orbits. Varying the timing of the pulses allows control of the collision dynamics of the two electrons.

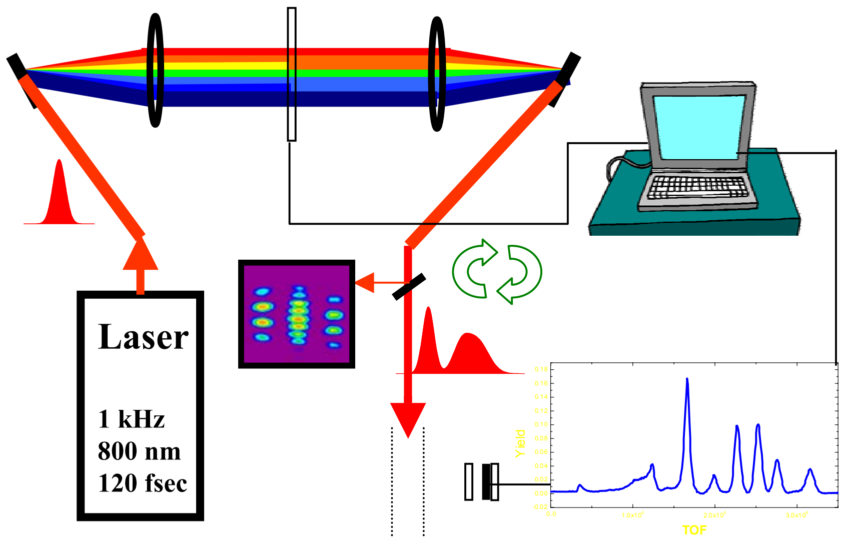
SCIENCE **303** 813 (2004) - FEBRUARY 6, '04

Probing Two-Electron Dynamics of an Atom

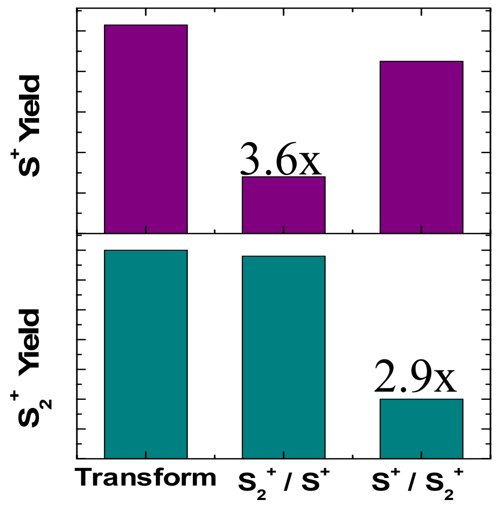
S. N. Pisharody and R. R. Jones*

Coherent short-pulse laser excitation has been used to control the approximate energy and relative proximity of two valence electrons within the same alkaline-earth atom, thereby providing insight into the dynamical evolution of a three-body Coulomb system. Our time-domain experiments enable direct experimental study of the electron dynamics at the classical limit of a two-electron atom. As an example, we look at the mechanism of autoionization for one two-electron configuration class and find that the doubly excited atom decays through a single violent electron-electron collision rather than a gradual exchange of energy between the electrons.

Research on Closed Loop Control of Intense Laser Fragmentation of Clusters



Schematic of a closed loop laser control apparatus. A genetic algorithm searches for the "best" laser pulse to optimize a specified laser fragmentation pattern. The algorithm controls a liquid crystal based laser pulse shaper based on feedback from a fragmentation experiment .



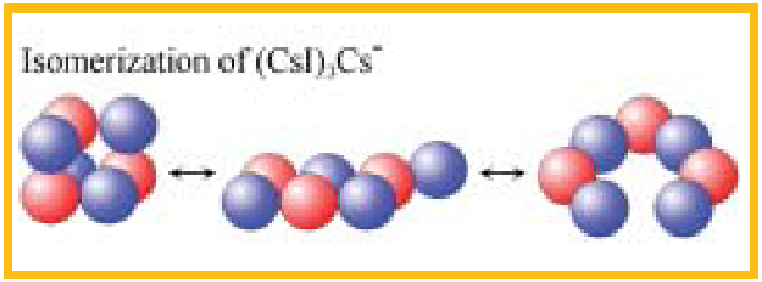
Typical results from a control experiment using S_8 as a target. The left most columns show the S^+ and S^{2+} product yields using an unshaped 100 fsec laser pulse. The middle and right hand columns show the same product yields when the algorithm is told to optimize the ratios S^{2+}:S^+ and S^+:S^{2+}, respectively.

Physics News in 2003

A Supplement to APS News

Edited by Phillip F. Schewe, Ben Stein, and James Riordon
Media & Government Relations Division, American Institute of Physics (AIP)

SALT: THE MOVIE. Solid, liquid, melting, and freezing are concepts that refer to bulk matter, not to individual atoms. But what about a small cluster of atoms or molecules? Louis Bloomfield and Andrew Dally (University of Virginia) looked at a pulsed beam of clusters of a salt; each cluster contained dozens of molecules that each had four cesium atoms and three iodine atoms. An ordinary salt grain has more than a million atoms along each side of its cubical structure. The Cs_4I_3 molecule can take on three different shapes or "isomers": a cube, ladder, or ring (shown in the figure). The researchers sent the salt clusters through a laser interaction region, where the cubic isomer was depleted. Using probing lasers downstream, the researchers watched at a cinematic 30 "frames" per second as the population of the cubic form was restored at the expense of ladders and rings. The interconversion, known as isomerization, happened more quickly with higher temperature. In fact, at about 500 K, the molecules spent only enough time in any one shape to convert into another, the signature of a phase transition from solid to liquid in a bulk system. Interestingly, the melting temperature of bulk cesium iodide is about 900 K. (A. J. Dally, L. A. Bloomfield, *Phys. Rev. Lett.* **90**, 063401, 2003.)



Lou Bloomfield, Dept. of Physics

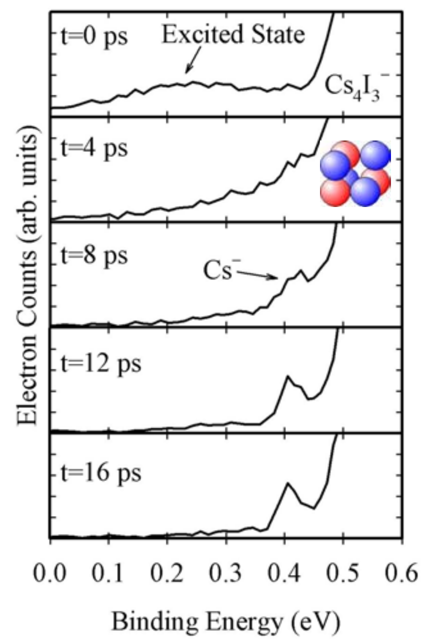
Research: Dynamics of Cluster Structure, Isomerization, and Photodissociation.

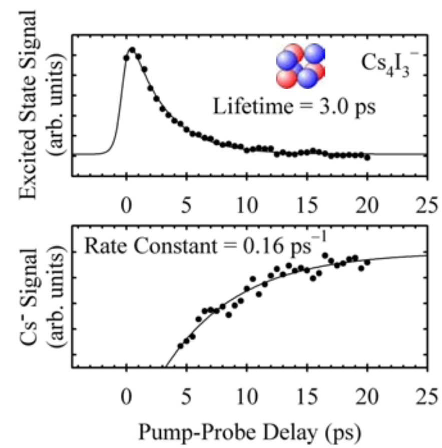
Associates: Andy Dally & Songbai Ye

Photodesorption of Alkali Negative Ions from Alkali-Halide Cluster Anions

Using picosecond laser pulses, we have examined the photodesorption of negatively charged alkali ions from alkali-halide clusters. These fragile atomic ions, with extra electrons that are only barely held in place, have not been observed previously among the fragments leaving alkali-halide (salt) surfaces following exposure to light.

We find this unusual desorption in a broad class of negatively charge alkali-halide clusters—those containing two or more electrons that are not involved in the salt's ionic bonding. The desorption starts via electronic excitation, with the excitation decaying quickly to eject the outgoing negative ion.





Thermal Isomerization Dynamics and Melting in Alkali-Halide Clusters

We have produced *(a)* ensembles of cluster ions with enough thermal energy to undergo rapid changes in geometric structure, a process known as thermal isomerization. Because they switch quickly from one isomeric form to another, these clusters are effectively molten.

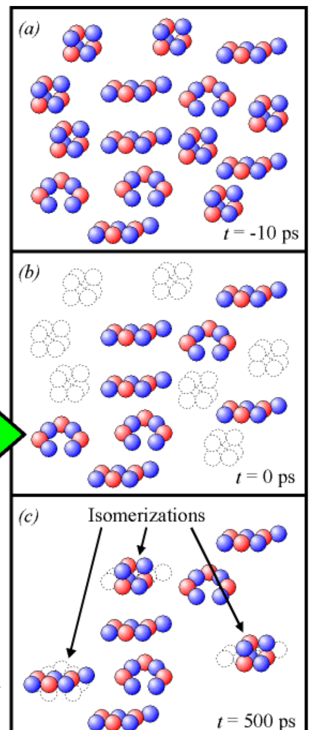
To study the dynamics of these clusters, we use an ultrashort laser pulse *(b)* to selectively destroy most of the clusters in one isomeric form. Thermal isomerization immediately begins to repopulate the missing form. We use a second laser pulse to measure the isomer populations at later times *(c)*, and thus learn about the structure, energetics, and thermal properties of these tiny systems.

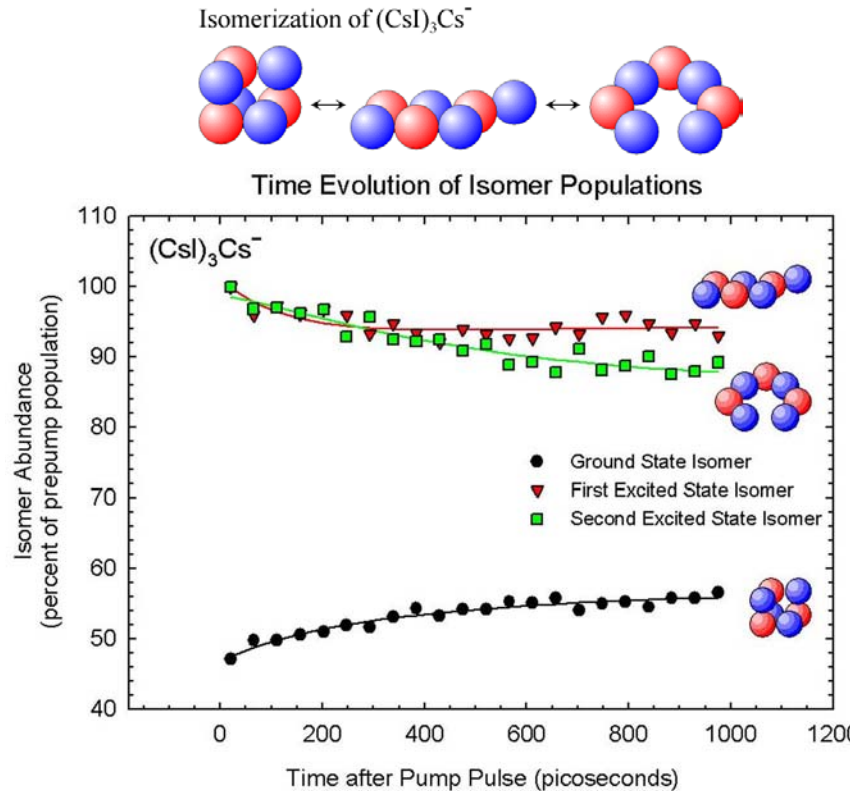
Original population of $(CsI)_3Cs^-$ isomers, produced with a temperature near room temperature

Ground state isomers are destroyed by light

Laser Pulse

New population of $(CsI)_3Cs^-$ isomers develops as the damaged ensemble reequilibrates through thermal isomerization





Brooks H. Pate, Dept. of Chemistry

Research: unimolecular isomerization kinetics, solvent effects on intramolecular vibrational dynamics, dynamic rotational spectroscopy

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Fellows:

Pam Crum (3rd Year) - Reaction dynamics in gas and solution by selective-excitation, broadband probe ultrafast IR spectroscopy (ps pump - fs probe)

Kevin Douglass (3rd Year) - Time-domain 2D-Microwave spectroscopy of
Gordon Brown (2nd Year) high-energy isomerization reactions (using Fourier transform signal acquisition)

Associates:

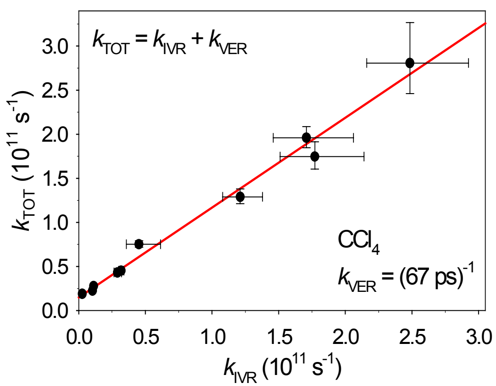
John Keske (Ph.D. 2001) - Rotational spectroscopy of isomerizing molecules

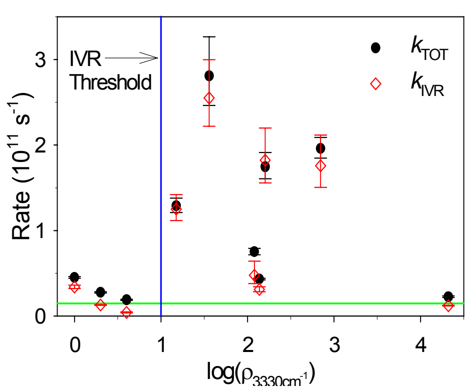
Hyun Yoo (Ph.D. 2002) - Vibrational dynamics in gas and solution

Yehudi Self-Medlin (Ph.D. 2004) - Vibrational dynamics and isomerization in gas and solution

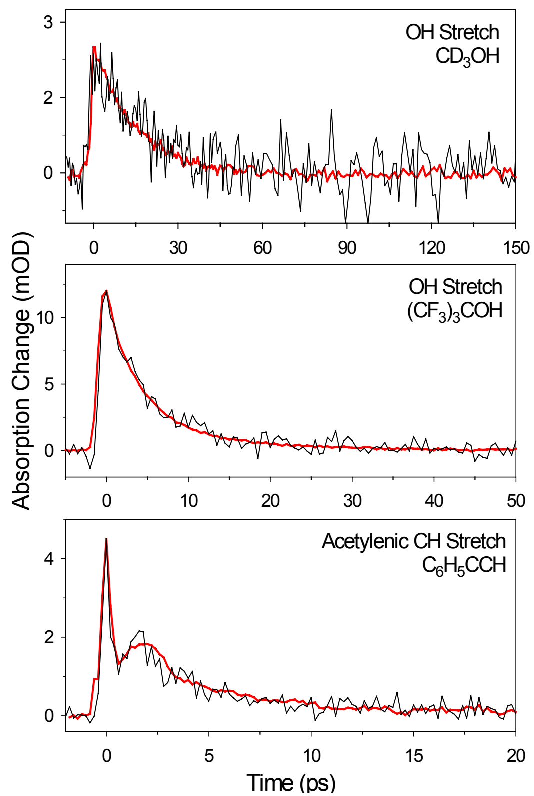
Vibrational Dynamics by Ultrafast Infrared Spectroscopy and Dynamic Rotational Spectroscopy

Competition Between Intramolecular and Solution Dynamics



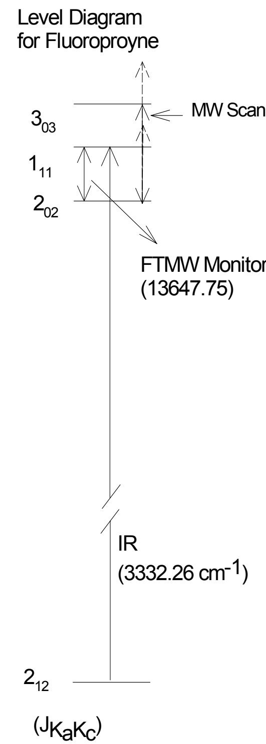


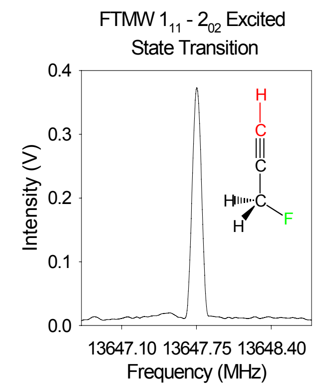
Direct Comparison of Gas- and Solution-Phase Dynamics

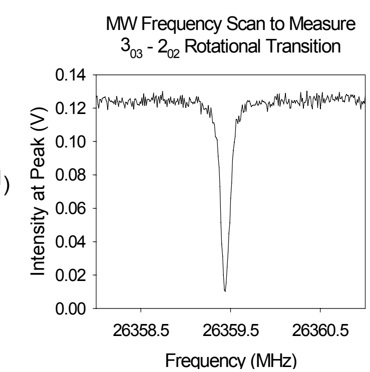


Using the SELIM Ultrafast Laser Facility we have performed the first direct comparison of the isolated molecule and solution phase vibrational energy relaxation rates of polyatomic molecules. We find that solvent effects are minor and that the total relaxation rate in solution is dominated by the purely intramolecular dynamics. (Second plot: Gas (black), 0.05 M CCl_4 solution (red))

Rotational Spectroscopy of Excited States by IR - FTMW - MW Triple Resonance Spectroscopy







The first measurements of the rotational spectrum of a laser-prepared vibrational excited state by Fourier transform microwave (FTMW) spectroscopy is shown. This technique has also been extended to include IR-MW-MW triple-resonance measurements.

Ian Harrison, Dept. of Chemistry

Research: Laser induced photochemistry and spectroscopy at surfaces, reaction dynamics of catalysis.

Fellows:

Alex Bukoski, Heather Abbott - Microcanonical rate theory at surfaces: Application to non-equilibrium laser, electron, and collisionally induced processes at surfaces.

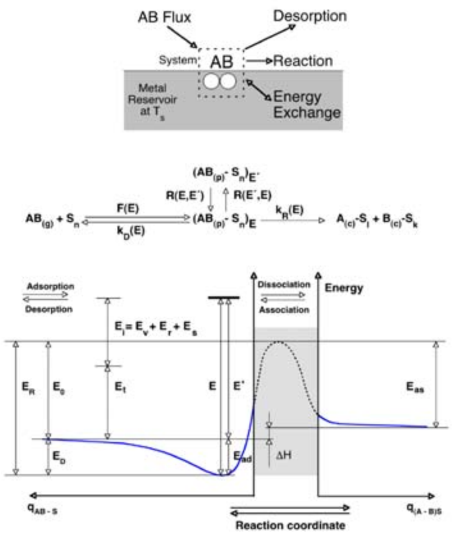
Kristin Buck - Surface photochemistry and spectroscopy: Broadband ir/visible sum frequency generation, ultrafast photochemistry of adsorbates

Associates:

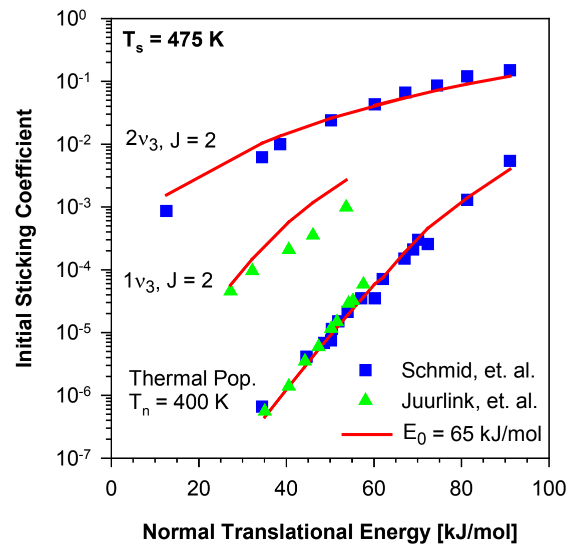
Rob Zehr, Neel Samanta -Dynamics of photoinduced electron transfer chemistry of adsorbates probed with nanosecond lasers.

Todd Schwendemann, Leticia Valadez - Electron transfer chemistry of adsorbates studied by scanning tunneling microscopy.

Microcanonical Unimolecular Rate Theory at Surfaces – IR Photochemistry in Catalysis



Schematic depiction of the kinetics and energetics of activated dissociative chemisorption. Central to catalytic steam reforming of CH_4 and chemical vapor deposition of Si and Ge from SiH_4 & GeH_4.



IR-laser pumping increases reaction probability by many orders of magnitude!

Dissociative chemisorption of a CH_4 molecular beam incident on a Ni(100) surface with and without laser excitation of the ν_3 antisymmetric C-H stretching vibration. Comparison of experiments to predictions of a 3-parameter model with a threshold energy for dissociation of E_0 = 65 kJ/mol (c.f. 432 kJ/mol for dissociation of gas phase CH_4).

Predictions for IR-FEL Enhanced Chemical Vapor Deposition at Surfaces

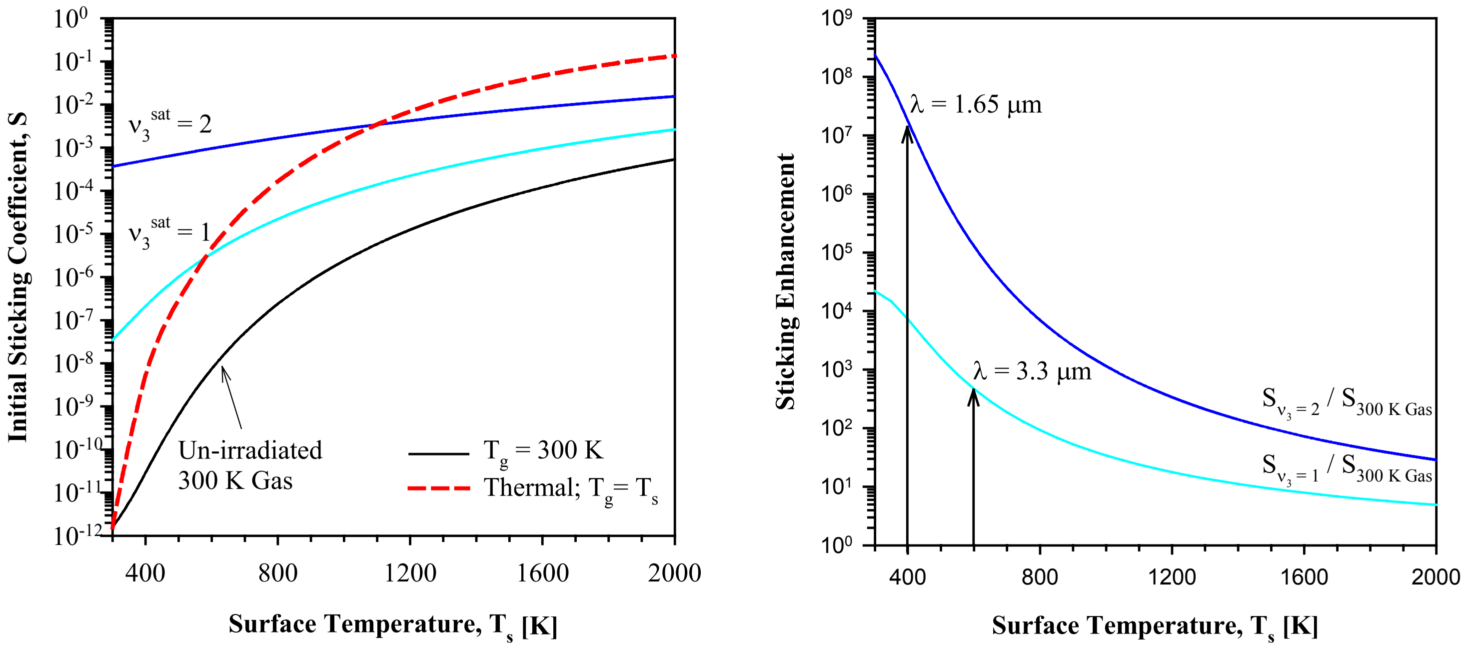
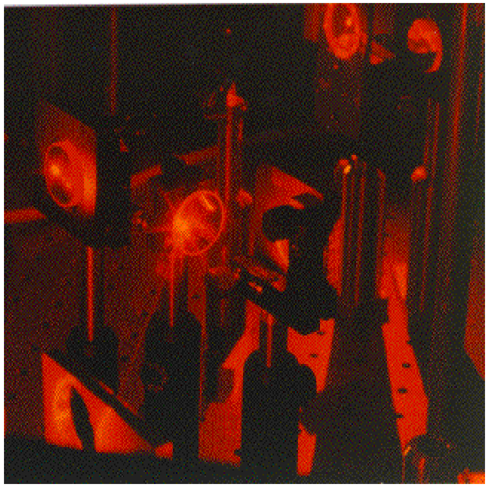
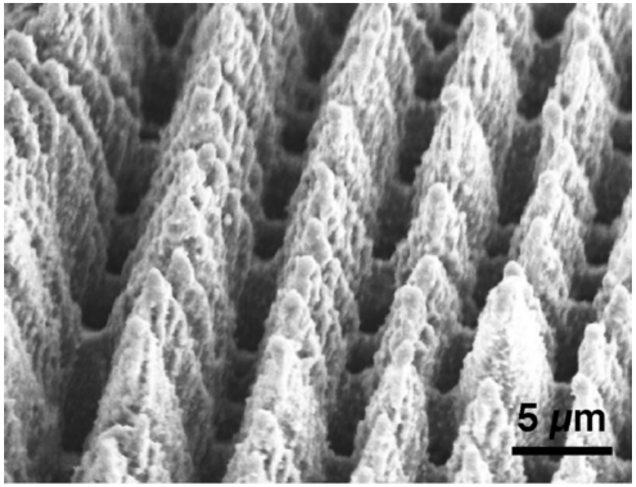


Fig. 1 (a) Dissociative sticking probabilities predicted for an ambient gas of methane impinging on Ni(100) as a function of surface temperature, with and without IR-FEL saturation pumping of selected ν_3 vibrational transitions. (b) Sticking enhancements for a 300 K ambient gas of methane pumped to saturation in the IR.

CAMOS Lab Director Kurt Kolasinski's Research on Si Pillar Formation





Laser irradiation of Si crystals under appropriate chemical & optical conditions can lead to the spontaneous formation of conical structures. These pillars can be tens of micrometers long. The tips, however, are on the order of a few hundred nanometers or less.

Current & Future Directions

- Increase use of the JLAB FELs as they become operating user facilities.
- Strengthen research & teaching ties between UVa, NSU, JLAB and other regional groups working with lasers.
- Dissemination of SELIM coursework through distance-learning tools.
- Increase research emphasis on FEL and advanced light source development – closer coupling to JLAB even when the FELs are not turned on (e.g., Accelerator Physics course at UVa).
- Work towards an IGERT renewal proposal this Spring.