# **MURI** activities

Cocke group, 2007-2009

Major objective:

-To launch dynamics in an atom or molecule by ionizing/exciting an electronic transition with an EUV attosecond pulse or (attosecond pulse train) and to follow/control the motion of the development of the wave packet so-formed with a phase-locked IR field.

-Systems studied to date:

Atomic system: He , electronic wave packets launched Molecular system: H<sub>2</sub>, vibrational wave packets launched -Personnel supported directly by MURI:

-K.Singh (postdoc)

-Personnel at in driving roles, not supported directly by MURI:

-P.Ranitovic, C.L.Cocke, S.De, S.Chen, X.-M.Tong (theory)

## Setup for APT/IR experiments



An attosecond pulse train (APT) (~4 cycles long) is generated by an 800 nm IR beam focused into a gas filled capillary where harmonics between the 11<sup>th</sup> and 27<sup>th</sup> (17-42 eV) are produced. These, and an inteferometrically correlated IR beam, are focused by a toroidal mirror onto a diffusive gas jet and the ions and electrons are momentum analyzed by a COLTRIMS spectrometer.

### Ionization of He by APT in presence of an IR field: Energetics



Participants:

P. Ranitovic, Xiao-Min Tong, B. Gramkow, S. De, B. DePaola, K. P. Singh, W. Cao, M. Magrakvelidze, D. Ray, I. Bocharova, H. Mashiko, A. Sandhu, E. Gagnon, M. M. Murnane, H. C. Kapteyn, I. Litvinyuk and C. L. Cocke

The 11<sup>th</sup> through 15<sup>th</sup> harmonics excite the He . Only the 17<sup>th</sup> can ionize it directly. Two lasers were used in this study with central wavelengths near 785 and 800 nm, and the overlap with the 13<sup>th</sup> and 15<sup>th</sup> harmonics with the resonances in He is different for the two cases. This has a profound effect on the ionization of He by the combined EUV/IR field.

### Ionization of He in presence of an IR field: He<sup>+</sup> yield



Experiment (Ranitovic et al., submitted)

Theory (X.-M. Tong)

As a function of APT/IR delay, three regions are found, with significant He<sup>+</sup> yield only if the IR comes during or after the APT. The latter situation results from resonant excitation of He followed by IR ionization and only occurs strongly for one of the laser wavelengths. The yield in the former situation is also laser-wavelength sensitive, which is seen more clearly in theoretical calculations for the process. For this case the yield oscillates as a function of the IR-APT delay with a period of half the IR optical period.

# Ionization of He in presence of an IR field: He<sup>+</sup> yield and photoelectron spectrum in overlap region with two IR beams



When two IR fields with different phases are applied, the oscillation period in the overlap region switches to a full IR period because of the modulation of the IR intensity . Both 13<sup>th</sup> harmonic + 3 IR photons and 15<sup>th</sup> harmonic + 1 IR photon contribute to the yield. The calculated results are very wavelength dependent , presumably because to the sensitivity of the different *np* resonances is very wavelength dependent.

# Localization of electron wave function during dissociation of D<sub>2</sub> by EUV/IR field



This experiment is performed using an APT generated by a two-color (400/800 nm) field, producing a single AP for each optical period of the 800 nm IR.

The harmonics in the APT, shown by the blue arrows, are sufficient to ionize  $D_2$  to the  $1s\sigma_g$  (lowest) potential curve of  $D_2^+$  only. The vibrational wave-packet so-launched is then coupled by a superimposed IR field to the  $2p\sigma_u$  potential. The linear combination of these potential curves finally results in an electron wave packet localized on the left or right in the fragmentation process, leading to asymmetric emission of the H<sup>+</sup> ions.

Personnel involved: K.Singh, W.Cao, P.Ranitovic, S.De, F.He, D.Ray, S.Chen, U.Thumm, A.Becker, M.M.Murnane, H.C.Kapteyn, I.Litvinyuk and C.L.Cocke

#### Localization of electron wave function during dissociation of D<sub>2</sub> by EUV/IR field: The COLTRIMS spectrum of ions emitted



The time of flight with no IR shows D<sup>2+</sup> ions and a small D<sup>+</sup> peak due to ground state dissociation. When the IR is added, much stronger emission of D<sup>+</sup> ions is generated by bond-softening/above threshold dissociation. Ions emitted to the right and left are easily distinguished by their flight times.

#### Localization of electron wave function during dissociation of D<sub>2</sub> by EUV/IR field: Asymmetry of emission



The left-right asymmetry of D<sup>+</sup> emission oscillates as a function of the relative delay of the APT and IR. This occurs because the relative phase of the  $1s\sigma_g$  and  $2p\sigma_u$  potential curves at the end of the dissociation depends on the phase of the IR at which the APT launches the vibrational wave packet. The red curve is a theoretical calculation by F.He based on the  $1s\sigma_g$  and  $2p\sigma_u$  coupling by the IR field. This data shows that the localization of the electronic wave function on the dissociating D<sub>2</sub><sup>+</sup> molecule can be manupulated/ controlled by the application of an IR field during the dissociation.

### **Publications**

- "Control of electron localization in molecules using XUV and IR pulses", K.P.Singh, W.Cao, P.Ranitovic, S.De, F.He, D.Ray, S.Chen, U.Thumm, A.Becker, M.M.Murnane, H.C.Kapteyn, I.Litvinyk and C.L.Cocke, submitted, NJP, (2009).
- "IR-Assisted Ionization of Helium by Attosecond XUV Radiation", P. Ranitovic, Xiao-Min Tong, B. Gramkow, S. De, B. DePaola, K. P. Singh, W. Cao, M. Magrakvelidze, D. Ray, I. Bocharova, H. Mashiko, A. Sandhu, E. Gagnon, M. M. Murnane, H. C. Kapteyn, I. Litvinyuk and C. L. Cocke, submitted, PRL, (2009).
- "Control of electron localization in a molecule using XUV and IR pulses", K. Singh, P. Ranitovic, W. Cao, S. De, D. Ray, S. Chen, I. Bocharova, M. Magrakvelidze, H. Mashiko, F. He, U. Thumm, A. Becker, I. Litvinyuk, C.L. Cocke, BAPS.2009.DAMOP.S5.8 (2009).
- "IR-Assisted Ionization of He by Attosecond XUV Radiation", P. Ranitovic, X.-M. Tong, B. Gramkow, S. De, B. DePaola, K. P. Singh, W. Cao, M. Magrakvelidze, D. Ray, I. Bocharova, H. Mashiko, E. Gagnon, A. Sandhu, M. M. Murnane, H. C. Kapteyn, I. Litvinyuk and C.L. Cocke, ICPEAC XXIV contributed abstracts, Mo 167 (2009).
- "Control of electron localization in a molecule using XUV and IR pulses", K. Singh, P. Ranitovic, W. Cao, S. De, D. Ray, H. Mashiko, S. Chen, F. He, A. Becker, U. Thumm, I. Litvinyuk, C. L. Cocke, ICPEAC XXIV contributed abstracts, Th 184, (2009).