COLTRIMS studies of strong-field laser-matter interactions

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We use coincident momentum imaging technique ("reaction microscope" or COLTRIMS) to study experimentally interactions of intense (10^{13}-10^{16} W/cm^2) ultrashort (6-60 fs) laser pulses with atoms and molecules in gas phase. In particular, we are interested in such aspects of laser-matter interactions as (i) interplay between non-resonant and resonant pathways of electronic excitation and ionization; (ii) ultrafast dynamics of strong-field processes; and (iii) interactions of strong-field with aligned molecules. To explore these aspects we (i) measure wavelength dependence of various strong-field processes; (ii) conduct pump-probe studies with short intense pulses; and (iii) employ rotational wavepacket technology to produce strongly anisotropic ensembles of molecules to interrogate.

1. Multi-photon resonant effects in strong-field ionization: origin of the dip in experimental longitudinal momentum distributions - *A.S. Alnaser, C.M. Maharjan, P. Wang and I.V. Litvinyuk*

We studied ionization of neon and argon by intense linearly polarized femtosecond laser pulses of different wavelengths (400 nm and 800 nm) and peak intensities, by measuring momentum distributions of singly charged positive ions in the direction parallel to laser polarization. For Ne the momentum distributions exhibited a characteristic dip at zero momentum at 800 nm and a complex multi-peak structure at 400 nm. Similarly, for Ar the momentum distributions evolved from complex multi-peak structure with a pronounced dip in the center at 400 nm, to a smooth distribution characteristic of pure tunneling ionization (800 nm high intensities). In the intermediate regime (800 nm, medium to low intensities), for both atoms we observed recoil ion momentum distributions modulated by quasi-periodic structures usually seen in the photoelectron energy spectra in multi-photon regime (ATI spectra). Ne did show a characteristic "dip" at low momentum, while the longitudinal momentum distribution for Ar exhibited a spike at zero momentum instead. The spectra did dramatically change at 400 nm, where both ions show the pronounced dip near zero momentum. Based on our results, we conclude that the structures observed in Ne and Ar momentum distributions reflect the specifics of atomic structure of the two targets and should not be attributed to effects of electron re-collision, as was suggested earlier. Instead, as our results indicate, they are due to effects of multi-photon resonant enhancement of strong-field ionization. The report of these studies is submitted for publication in J. Phys. B [1].

2. Resonant excitation during strong-field dissociative ionization - *A. S. Alnaser, M. Zamkov, X. M. Tong, C. M. Maharjan, P. Ranitovic, C. L. Cocke, and I. V. Litvinyuk*

We studied dissociative ionization of oxygen by intense femtosecond laser pulses with central wavelengths between 550 and 1800 nm. We measured kinetic energy release spectra and angular distributions of fragments resulting from symmetric dissociation of doubly charged molecular ions ($O_2^{2+} \rightarrow O^+ + O^+$ channel). In the kinetic energy release spectra we identified a number of distinct excited states of the molecular ion. Angular distributions for all but one of those states were consistent with re-collision excitation following single ionization of O_2. For the remaining ($B\ ^3\Pi_g$) excited state we observed a characteristic resonant dependence of its relative yield on the central optical frequency of the pulse, with the yield peaking at around 890 nm. This presents unambiguous evidence in support of importance of resonant excitation channels in strong field ionization of molecules. This study is reported in [2].

3. Momentum-imaging investigations of the dissociation of D_2^+ and the isomerization of acetylene to vinylidene by intense short laser pulses - *A. S. Alnaser, I.V. Litvinyuk, T. Osipov, B. Ulrich, A. Landers, E. Wells, C. M. Maharjan, P. Ranitovic, I. Bocharova, D. Ray and C.L. Cocke.*

We measured momentum-images of the ionic products from the ionization of D_2 and C_2H_2 by short laser pulses. For D_2 we use a pump-probe approach to investigate the dependence of the enhanced ionization on the internuclear distance. Evidence for two (not well separated) regions of enhancement are found near internuclear distances of 6 and 10 atomic units. In the case of acetylene we report clear evidence for the production of both acetylene and vinylidene dications with kinetic energy releases similar to those reported earlier by core electron removal. We also find very different angular distributions for the fragments in the two channels, consistent with a finite time for the isomerization. These studies are reported in [3].

4. Strong-field ionization of dynamically aligned molecules – *C.M. Maharjan, A.S. Alnaser, I. Bocharova, P. Ranitovic, D. Ray, C.L. Cocke and I.V. Litvinyuk*

This study is motivated by the recent molecular tomography experiment of Corkum and co-workers. In this experiment they reconstruct complete the HOMO wave-function of N_2 (including phase) from a series of high harmonics spectra measured with molecules aligned at various angles to the laser polarization. The reconstruction procedure uses normalization to the high harmonics spectra of a companion atom (Ar), and it is based on the assumption that re-colliding electron wavepacket has the same 3D structure for N_2 and Ar, and also for N_2 this electron wavepacket is independent of molecular orientation. The hypothesis is that molecular structure and orientation do not affect the momentum distribution of tunneling electrons (“all tunnels are alike”). We undertook to directly verify this assumption. We measured full 3D electron momentum distributions for aligned N_2 and O_2 molecules. In a pump-probe experiment we first created rotational wavepackets in target molecules with a weak pump pulse and then ionized molecules with a stronger probe pulse around the wavepacket revival time, when most molecules are aligned either parallel or perpendicular to the polarization direction of the pump. The probe polarization was perpendicular to the pump polarization. We measured high-resolution electron energy spectra and complete 3D momentum distributions of electrons

using COLTRIMS. In summary, for aligned N_2 and O_2 we did not observe a significant dependence of the electron energy spectra and 3D momentum distributions on the alignment of the molecular axis. Our results seem to provide experimental validation for the "all tunnels are alike" hypothesis behind the molecular tomography technique. However, for both molecules we did observe small but definite difference in momentum distribution projections on the two axes perpendicular to polarization direction – one along the alignment axis, another perpendicular to it. Along the alignment axis the 1D projection of 3D momentum distribution showed more pronounced cusp structure at zero momentum, than similar projection on the perpendicular axis. Such cusp structure is usually associated with "Coulomb focusing" of the electron wavepacket by the ion core potential. We attribute observed differences to different Coulomb focusing in the two directions due to the two-center attractive potential which breaks the cylindrical symmetry of the problem. Also in the electron energy spectra for both molecules, in addition to the usual ATI peaks with photon energy (1.55 eV) periodicity we observe multiple non-ATI peaks at low energy (0-5 eV) indicative that some resonance or resonance-enhanced ionization processes also playing a role. These peaks seem to be related to similar structures observed in Ar (see abstract by C.L. Cocke and [4]). The exact nature and production mechanism for these low energy electron peaks remains unexplained and will be a subject of further studies. The publication of these results is being prepared.

5. Time-resolved Coulomb explosion imaging of small molecules - *F. Légaré, K. F. Lee, I. V. Litvinyuk, P. W. Dooley, A. D. Bandrauk, D. M. Villeneuve, P. B. Corkum*

This is continued collaboration with Ottawa and Sherbrooke groups aimed at developing Coulomb explosion imaging as probe of time-dependent molecular structure. The experiments were conducted in Ottawa awhile ago, with collaborative work on data analysis and presentation resulting in two publications in 2005. In [5] we use intense few-cycle laser pulses to ionize molecules to the point of Coulomb explosion. We use Coulomb's law or *ab initio* potentials to reconstruct the molecular structure of D_2O and SO_2 from the correlated momenta of exploded fragments. For D_2O, a light and fast system, we observed about 0.3 Å and 15° deviation from the known bond length and bond angle. By simulating the Coulomb explosion for equilibrium geometry, we showed that this deviation is mainly caused by ion motion during ionization. Measuring three-dimensional structure with half bond length resolution is sufficient to observe large-scale rearrangements of small molecules such as isomerization processes. In [6] we image the dynamics of diatomic and triatomic molecules with sub- 5 *fs* and sub-Å resolution using laser Coulomb explosion imaging with 8 *fs* pulses. We obtain image information by measuring the vector momenta of all atomic ions produced by explosion of a single molecule. We image vibrating D_2^+ and of dissociating SO_2^{2+} and SO_2^{3+} . Images taken at 0 and 60 *fs* show that the dissociation of SO_2^{2+} produces an SO^+ rotational wave packet.

Publications (2005-06 and pending)

1. A.S. Alnaser, C.M. Maharjan, P.Q. Wang and I.V. Litvinyuk, "*Multi-photon resonant effects in strong-field ionization: origin of the dip in experimental longitudinal momentum distributions"*, submitted to J. Phys. B
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3. A.S. Alnaser, I.V. Litvinyuk, T. Osipov, B. Ulrich, A. Landers, E. Wells, C.M. Maharjan, P. Ranitovic, I. Bocharova, D. Ray and C.L. Cocke, "*Momentum-imaging investigations of the dissociation of D_2^+ and the isomerization of acetylene to vinylidene by intense short laser pulses"*, J. Phys. B: At. Mol. Opt. Phys. **39**, S485 (2006)
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10. I.V. Litvinyuk, F. Légaré, P.W. Dooley, D.M. Villeneuve, P.B. Corkum, J. Zanghellini, A. Pegarkov, C. Fabian, and T. Brabec, "*Shakeup Excitation during Optical Tunnel Ionization***",** Phys. Rev. Lett. **94**, 033003 (2005)
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