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Studies of complex systems in intense, ultrafast mid-infrared laser fields

Cosmin Blaga
OHIO STATE UNIVERSITY THE

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Dr. Cosmin Blaga
blaga.1@osu.edu

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14. ABSTRACT

This project investigated the behavior of complex matter (molecules and solid state) with strong, ultrafast laser fields operating at midinfared wavelengths. Areas of research addressed: ionization and fragmentation of hydrocarbons with vibrationally resonant pulses at/near the C-H stretch mode frequencies, high harmonic generation is solids, generation of strong, ultrafast midinfrared pulses with orbital angular momentum, femtoseccond laser damage in semiconductors, attosecond electron interferometry.

15. SUBJECT TERMS

Strong field laser, mid-infrared, ionization, femstosecond, attosecond

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"STUDIES OF COMPLEX SYSTEMS IN INTENSE, ULTRAFAST MID-INFRARED LASER FIELDS"

Award number: **FA9550-15-1-0203**

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PI: Cosmin I. Blaga (Ohio State University**)**

Collaborators: L. F. DiMauro **(**Ohio State), P. Agostini (Ohio State), J. Xu (Ohio State), E. Chowdhury (Ohio State), K. Schafer (Louisiana State), F. Catoire (CNRS, Bordeaux, France) and M. Kling (MPQ, Garching, Germany).

1. ABSTRACT

Technological advancements have recently led to the development of novel intense, ultrafast laser systems operating into the midinfrared (MIR) regime. As virtually all bedrocks of ultrafast, strong-field physics have been laid employing visible and near infrared (VIS/NIR) laser sources, it became imperative to test if and how fundamental concepts apply in the new regime. Perhaps not surprising, for atoms our previously developed strong-field tools (strong-field approximations or numerical integration of the time dependent Schrödinger equation) were found to be largely applicable in the MIR regime, albeit at higher computational costs giving increased ponderomotive energies [Colosimo2008]. On the other hand, as most molecules possess vibrational modes in the MIR band, at even moderate laser intensities vibrational effects, usually neglected in most theories, are expected to become significant. During the last five years, at OSU we have developed tunable MIR lasers and began investigating hydrocarbons interacting with pulses resonant with the C-H stretch mode centered near 3.3 μm. We discovered extremely strong vibrational effects, with fragmentation channels enhanced up to two orders of magnitude compared to the nonresonant case. The main goals of the current grant were four-fold: *i)* collect comprehensive experimental data sets on- and off- resonance as a function of various laser parameters in order to *ii)* support the development of a more general theoretical framework including molecular vibrations, *iii)* explore and exploit the favorable ponderomotive scaling laws that favor longer wavelengths – such as production of soft X-rays via high harmonic generation (HHG) and *iv)* identify future, desirable avenues for MIR ultrafast laser science and technology.

2. REVIEW NARRATIVE

2.1. Brief overview. During the award period we pursued several investigative directions studying the interaction of strong MIR pulses with complex matter, the original purpose of the grant. Towards this goal, we collected fragmentation data for methane (CH_4) with respect to its isotopic counterparts ($CH_{4-x}D_x$), which allowed us to control the number of C-H molecular bonds that were resonantly "tugged" during ionization while keeping the same electronic configuration. This effort concluded the first purpose of the grant, namely to collect a comprehensive data set for strong-field ionization of hydrocarbons with vibrationally resonant pulses. Although the experimental data set is complete and ready for publication, theoretical quantum chemistry calculations based on time dependent density functional theory (TDDFT) are necessary to decipher all experimental results. A collaboration with the group of Prof. John Herbert - theoretical chemist from OSU in Department of Chemistry – is well underway and first theoretical results are expected towards Fall 2018. In a separate investigative direction, we completed a collaboration with Dr. Enam Chowdhury's group to study laser damage mechanisms in semiconductors. A third investigative pathway was dedicated to the development of MIR laser technologies. Towards this goal high harmonic generation (HHG) in carrier-doped semiconductors and HHG in a photonic crystal were pursued, two collaborative projects developed at OSU as part of Prof. Louis F. DiMauro's AFOSR-MURI efforts. However, significant efforts were dedicated towards a fourth investigative pathway. Driven mainly by the current grant, we demonstrated a novel, highly promising electron wavepacket attosecond interferometric technique for probing molecular dynamics on ultrashort (10 attoseconds) time scales. Driven with tunable lasers, the new technique is based on rescattering electron wave-packets and unlike traditional HHG methods it works for *isolated* atom/molecules and is thus free of any macroscopic effects such as phasematching. The method is also far less demanding experimentally both on the laser and the vacuum side. Although we benchmarked this novel method for argon with VIS/NIR lasers for computational reasons, its application for complex systems (molecules) will require MIR pulses, in accordance to the purpose of the grant.

2.1.1. Tunnel ionization of molecules with vibrationally resonant MIR pulses.

If during the first half of the grant we studied the role of vibrational resonances by tuning the laser wavelength near the C-H stretch mode (~ 3.3 μm) of various hydrocarbons, in the second half we focused on isotopic effects, studying hydrocarbons for which the normal hydrogen (m_H = 1 a.u.) was substituted with deuterium atoms (m_D = 2 a.u). This allowed us to study molecular counterparts with the same electronic configurations but different vibrational manifolds ionized by identical laser fields. As illustrated in Figure 1, the fragmentation of CD_4 at two nonresonant drivers (2.9 and 3.3 μm) is identical with the nonresonant case (2.9 μm) in CH_4. In these nonresonant cases, the mass spectrum is dominated by the unfragmented parent ion (CH_4^+ or CD_4^+) whereas the CH_3^+ or CD_3^+ fragment amount to approximately 10% of the total signal. In addition, the total ionization rates for CD_4 and CH_4 at the 2.9 μm nonresonant case are identical, allowing us to conclude that vibrational levels play no significant role in the nonresonant tunnel ionization of molecules. As a corollary, we also concluded that coupling laser energy into vibrational degrees of freedom is minimal. However, when the MIR laser wavelength matches a vibrational resonance, as is the case for CH_4 at 3.3 μm, we observed large fragmentation enhancements and an overall increase of the total ionization rate by a factor of five.

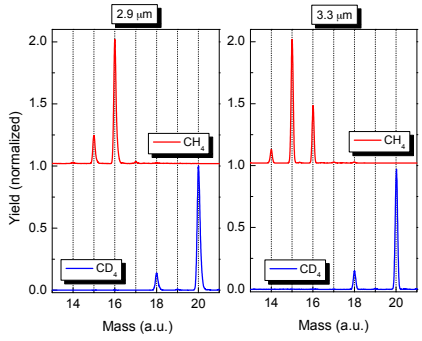

Figure 1: Strong field ionization of methane isotopes. **Left –** Mass spectra of CH_4 (top) and CD_4 (bottom) with both molecules ionized with 2.9 μm pulses (off-resonance) display identical fragmentation ratios. **Right –** same, with CH_4 (top) resonantly excited and CD_4 (bottom) off-resonant.

In addition to methane, we have also obtained isotopic data in ethane and deuterated ethane reaching identical conclusions. The experimental data sets are complete and we are currently expecting the first theoretical modeling results from our collaboration with Prof. John Herbert, theoretical chemist at Ohio State to elucidate the interaction of molecules with vibrational resonant pulses. The main theoretical difficulty rests on the fact that small hydrocarbons are large enough for ab initio quantum calculations (i.e. brute force numerical methods for the time-dependent Schrödinger equation) but small enough for mean-field based methods life TDDFT. We plan to publish the results later this year, with the exact timing depending upon the completion of the theoretical work.

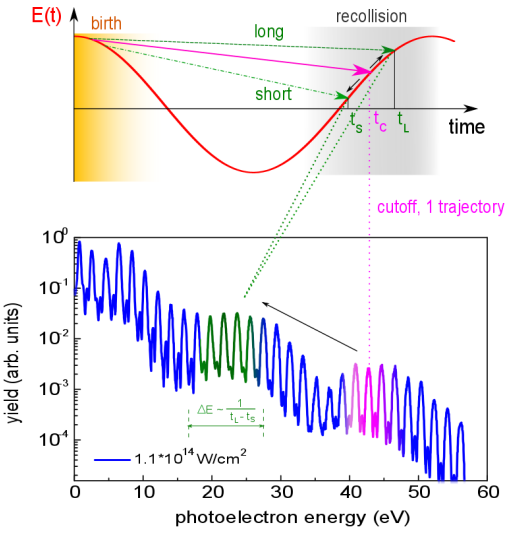

Figure 2: Principle of long and short electron trajectory interferometry. (**top**) – Electric field of the laser and the classical trajectories. (**bottom**) – photoelectron spectra along the laser linear polarization, calculated via numerical integration of the time-dependent Schrödinger equation (TDSE) for a 5-cycle flat top pulse at 650 nm. The color-coded trajectories are responsible for the two enhancements visible in the spectra.

2.1.2. Photoelectron interferometry driven with tunable lasers. During the second half of the grant, we dedicated significant efforts towards a new project, namely the development of an electron interferometric method that could have broad applications in ultrafast molecular dynamics. The key ingredients of the method are interferences between returning photoelectron trajectories (the quantum equivalent of classical long and short trajectories, illustrated in Figure 2) and the existence of leading intensities that are due to channel-closure yield enhancements in the laser focal volume [Miloševic2007].

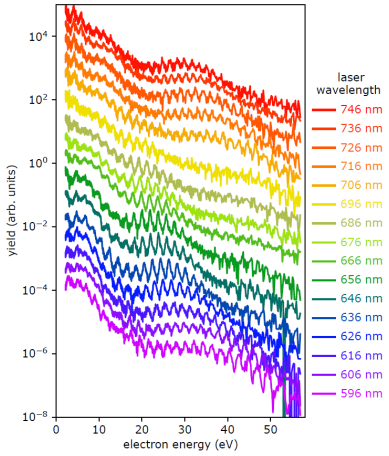

Figure 3: Electron wave packet interferometer tuning. Photoelectron spectra at various laser wavelengths, displaying the evolution of the interference maximum, seen as the enhancement in the 15-40 eV region.

Although the existence of the leading intensity is beneficial as it allows the enhancement (green-coded region in Figure 2) to survive focal averaging, it also "fixes" the relative phase between different quantum trajectories at a given wavelength [Paulus1994]. Consequently, at a given laser wavelength, the interferometer formed by different quantum orbits is practically "stuck". The solution that allows us to scan the interferometer relies on finely scanning the laser wavelength, as seen in Figure 3 for argon atoms.

To interpret our results, an extensive set of numerical integrations of the time-dependent Schrödinger equation for a wide range of laser parameters were performed using the Ohio Supercomputer Center in collaboration with Prof. Kenneth Schafer from Louisiana State University. In addition, an analytical formula based on the strong field approximation developed by Frolov, Manakov and Starace (FMS) [Frolov2009] was found to reproduce the experimental results and confirm the interferometric interpretation. These results can be seen in Figure 4.

The most important characteristic of the interferometer is its temporal resolution. As seen from Figure 4, the enhancement position changes with ~50±2% eV/fs for the 13-photon channel closure, which for a rather standard time of flight precision of 1% (0.3 eV at 30 eV) translates into 10 as temporal resolutions. Compared to a typical X-ray HHG attosecond apparatus, time of flight spectrometers

are extremely simple, table top and cost effective, requiring only modest pulse energies (few tens of μJ in Figure 3). Additionally, the presence of the leading intensity renders the interferometer immune to typical <5% shot to shot intensity fluctuations.

Although we presented here only the results for argon, we have measured the interferometric signal in all noble gases, but more importantly, in molecules such as N_2, methane, ethane and others. It is in these molecular systems where interferometric methods promise to be extremely valuable tools for attosecond molecular dynamics, charge migration, molecular imaging, etc. A manuscript with the results outlined in this document is complete and will be submitted to *Science* next month (July 2018).

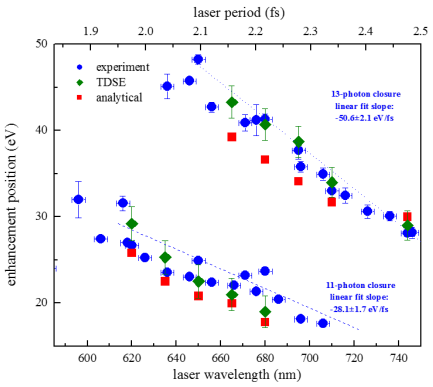

Figure 4: Interference maxima as a function of laser wavelength. Experimental data (blue) vs. TDSE (green) and the FMS analytical formula (red). The enhancement position is extremely sensitive to wavelength (or laser period), demonstrating interferometer sensitivity better than an atomic unit of time (24 as).

Finally, motivated in part by this project we have conducted an extensive survey to benchmark the ionization pathways in various targets as a function of laser wavelength [Lai2017]. In particular, we investigated the transition from tunneling to multiphoton ionization and established general guidelines for the applicability of various ionization theoretical models. Thus, the survey has far-reaching implications as it pertains to many strong field phenomena since ionization is a critical step in a wide array of applications (HHG spectroscopy, laser filamentation, ultrafast molecular imaging, etc.).

2.1.3. Plasmonic field enhancements in fullerenes. Field enhancements around metallic nanotips have been demonstrated to produce high-energy electrons even when irradiated with weak femtosecond pulses [Hommelhoff2006]. The origin of the field enhancement is the presence of free electrons in the metal, collectively reacting to the external laser field in accordance to Maxwell's equations. During the project, we have observed similar enhancements in C_{60}, a macromolecule with 60 unpaired electrons that act in an analogous manner to the nanotip. The signal of interest appears as large yield enhancements for *direct* electrons, especially near the classical $2U_P$ cutoff (here, U_P is the ponderomotive potential, the cycle average kinetic energy of a free electron placed in an electromagnetic field) but also as a suppression of the low energy structure (LES) [Blaga2009], a dephasing effect on the *rescattered* electron wavepacket due to the induced dipole. These results have been successfully reproduced numerically using classical electron trajectory Monte Carlo simulations at the Ohio Supercomputer Center. In addition, our collaborator, Dr. Fabrice Catoire (CNRS, Bordeaux, France) is currently developing a time-dependent density functional theory (TDDFT) for C_{60}. The results of this project will be published later this year (2018). We expect to publish these results towards the end of the year (late 2018).

2.1.4. High harmonic generation in condensed media. During the last year of the project we began studying HHG in molecular clusters, liquids and crystals. For crystals, we investigated HHG from an electron-doped semiconductor through photoexcitation. By studying the effect of carrier density, we identified that interband transition dominates both the above- and below-bandgap HHG in zinc oxide and predicted that at longer wavelengths, e.g. far-infrared, that the emission will evolve to intraband Bloch transitions. These results have been reported in Nature Communications [Wang2017]. In a separate project, we reported on the generation of harmonic-like photon up- conversion in a $LiNbO_3$-based nonlinear photonic crystal by mid-infrared femtosecond laser pulses. Studying below bandgap harmonics for various driver wavelengths, we demonstrated a conversion efficiency reaching 13% for the 11^{th} harmonic when the laser was operated at 4 μm. We attributed our results to a cascaded three-wave mixing mechanism which reproduced well the general features of the observed spectra, including a plateau-like harmonic

distribution and the observed efficiency. These results were published in Optics Letters [Park2017].

2.1.5. Laser-induced optical damage at MIR wavelengths. Throughout the project, we established a collaborative effort with Dr. Enam Chowdhury's group at OSU, investigating laser damage formations in MIR-relevant optical materials. We observed the formation of high spatial frequency laser induced periodic surface structures (HSFL) in germanium between 2-3.6 μm. These results were explained by a modified surface-scattering model including Drude excitation and the optical Kerr effect. The results were submitted and published in *Journal of Applied Physics* [Austin2016].

3. PERSONNEL MILESTONES

Dr. Cosmin Blaga, principal investigator, will join the James R. MacDonald Laboratory at Kansas State University as tenure-track faculty starting the 2018/2019 academic year.

Dr. Kaikai Zhang, partially supported by this grant as a postdoc, joined Amplitude Systems as a laser engineer.

Ms. Stephanie Slaughter, a physics undergraduate junior student at Ohio State was supported from the current grant as a summer undergraduate research associate and developed a laser pointing stabilization system.

Ms. Xiaowei Gong, a physics undergraduate senior student at Ohio State earned a departmental summer research fellowship and worked with Mr. Yu-Hang Lai (graduate student supported from other sources) and successfully developed a MIR frequency resolved optical gating (FROG) apparatus. In Fall 2017, Ms. Gong joined the physics graduate program at Duke University.

4. PUBLICATIONS ACKNOWLEDGING THE GRANT

[Austin2016] Austin, D.R *et al.*, High Spatial Frequency Laser Induced Periodic Surface Structure Formation in Germanium, *J. Appl. Phys.,* **120**, 143103 (2016). DOI: http://dx.doi.org/10.1063/1.4964737

[Lai2017] Lai, H-L *et al.*, Experimental investigation of strong-field ionization theories for laser fields from visible to midinfrared frequencies, *Phys. Rev. A*, **96**, 063417 (2017). DOI: https://doi.org/10.1103/PhysRevA.96.063417

[Park2017] Park, H. et al., High-order harmonic generations in intense MIR fields by cascade three-wave mixing in a fractal-poled LiNbO3 photonic crystal, *Optics Letters*, **42**, No. 19, 4020 (2017), DOI: https://doi.org/10.1364/OL.42.004020

[Wang 2017] Wang, Z. *et al.*, The roles of photo-carrier doping and driving wavelength in high harmonic generation from a semiconductor, *Nature Communications,* 8: 1686 (2017), DOI: https://doi.org/10.1038/s41467-017-01899-1

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[Frolov2009] Frolov, M. V., Manakov, N. L. and Starace, A. F., Analytic formulas for above-threshold ionization or detachment plateau spectra, *Phys. Rev. A*, **79**, 033406 (2009).

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[Xu2014] Xu, J. at el., Diffraction using laser-driven broadband electron wave packets, *Nat. Commun.,* 5:4635 doi: 10.1038/ncomms5635 (2014).

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Abstract

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[Lai2017] Lai, H-L et al., Experimental investigation of strong-field ionization theories for laser fields from visible to midinfrared frequencies, Phys. Rev. A, 96, 063417 (2017). DOI: https://doi.org/10.1103/PhysRevA.96.063417

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[Wang 2017] Wang, Z. et al., The roles of photo-carrier doping and driving wavelength in high harmonic generation from a semiconductor, Nature Communications, 8: 1686 (2017), DOI: https://doi.org/10.1038/s41467-017-01899-1

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