FY10 MURI THIRD YEAR REPORT

Mathematical Modeling and Experimental Validation of Ultrafast Nonlinear Light-Matter Coupling Associated with Filamentation in Transparent Media

AFOSR Grant No: FA9550-10-1-0561

TITLE: Mathematical Modeling and Experimental Validation of Ultrafast Nonlinear Light-Matter Coupling Associated with Filamentation in Transparent Media

PRINCIPAL INVESTIGATOR: Professor Jerome V Moloney

LEAD INSTITUTION: University of Arizona

AFOSR PROGRAM MANAGER: Dr. Arje Nachman, AFOSR, (703) 696-6565, arje.nachman@afosr.af.mil

PROGRAM OBJECTIVE: The objective of the program is to develop a first-principles, rigorous scientific theoretical and mathematical basis for understanding the nature of the interaction between intense ultrafast laser pulses with gaseous and condensed media. The program is designed to elucidate the physics associated with extreme nonlinear optical events causing 3D pulse spatio-temporal deformations, enable the creation of photo-ionized anisotropic nonequilibrium plasmas with concomitant white-light generation, and predict the evolution of the pulse over extended propagation paths. An overarching goal of the program is to exploit the spatial and temporal degrees of freedom in 3D laser pulses to design new classes of robust propagating conical wave based nonlinear wavepackets that can circumvent turbulent scattering and be delivered in a controlled manner at range.

SCIENTIFIC APPROACH: Prior work on the theoretical study of intense, ultrashort pulse filamentation in gases and condensed media has relied on using outdated parameterized phenomenological models proposed in the sixties and seventies designed for much longer duration or continuous wave laser propagation. As a consequence, attempts to explain ultrafast femtosecond light-matter coupling deviations require even further parameterization based on a flawed starting model and completely lack any predictive features. Consequently, the level of fundamental scientific understanding necessary to enable promising applications in remote delivery, sensing and spectroscopic applications has been lacking. The basic scientific approach of this program is to revisit such ultrafast nonlinear applications at the fundamental quantum level and explore unconventional laser pulse waveforms that may be more resistant to diffractive spreading, scattering and losses over realistic propagation paths. Firstly, team members are developing rigorous quantum mechanical models and carrying out simulations of intense field photo-ionization of individual atoms and molecules with the goal of removing ad hoc parameterization and understanding nonlinear saturation, generation of nonequilibrium electron/ion distributions, and anisotropy in the response to probes (THz or optical). Furthermore Coulomb mediated scattering of the highly anisotropic freed electrons/ions requires that they move beyond the single particle description and address the full many-body quantum Boltzmann scattering problem. Secondly, the team is addressing the generation and propagation of unconventional (Bessel, Airy, Vortex, etc) 3D pulse waveforms in the fully nonlinear regime as a means of overcoming the limitations of finite nonlinear focal volumes, scattering and absorption losses. Here they face a number of challenges: 1) Understanding the role of nonlinear propagation on otherwise linear robust conical wave pulsed waveforms, 2) extending the nonlinear interaction zone in the laboratory by employing either ignitor/heater schemes or dressed filaments and 3) porting these laboratory scale developments to multi-km range. Thirdly, a critically important aspect of this MURI program is to engage world class experimental teams to both challenge the level of understanding by discovering novel unexplained effects and validate the theoretical/mathematical understanding of the many competing physical processes competing in the nonlinear interaction volume.

MOST RECENT GOVERNMENT REVIEW AND GOVERNMENT PARTICIPANTS:
Albuquerque, NM (near Kirtland AFB), September 18, 2012
Dr Arje Nachman (AFOSR), Dr Enrique Parra (AFOSR), Dr Thomas Gavrielides (AFRL/RD), Dr Matthew Grupen (AFRL/RY), Dr James Harvey (ARO), Dr Danhong Huang (AFRL/RV), Dr Thomas Hussey (AFOSR), Dr Joseph Penano (NRL), Dr Tony Ting (NRL), Dr Nicholas Usechak (AFRL/RY), Dr Iyad Dajani (AFRL/RD), Dr Erik Bochove (AFRL/RD)

MURI CONSORTIUM RESEARCH TEAM MEMBERS:
Jerome V Moloney (U Az)--PI
Margaret Murnane (U Colorado)
Andreas Becker (U Colorado)
Robert Levis (Temple)
Alexander Gaeta (Cornel)
Demetrios Christodoulides (U Central Florida)
Charles Durfee (Colorado School of Mines)

FINANCIAL EXECUTION:

3-YEAR BASE PERIOD – FY10: $367,158.00 (2 months) **FY11:** $1,362,728.00 (12 months) **FY12:** $1,467,825.00 (12 months) **FY13:** $1,284,516.00 (10 months)

2-YEAR OPTION PERIOD – FY13: $222,020.00 (2 months) **FY14:** $1,500,938.00 (12 months) **FY15:** **$**1,294,694.00 (10 months)

As of 11/29/12, $3,120,435.17 of the Base Period funds has been expended against a current budgeted allocation of $3,197,721. As of 11/29/12, the PI projects that when the 3-year Base Period ends, the team will have spent well over 90% of the Base Period funds. It is recommended that no funds should be withheld when the two year option period begins.

ACCOMPLISHMENTS TO-DATE:

Overall, the project to date has established that the status quo of current filamentation modeling is not adequate and that highly nonlinear quantum saturation phenomena accompanied by photo-ionization do not follow the established phenomenological parameterization widely used in the literature. The team has begun to build a sophisticated quantum description of ultrafast light-matter coupling at the single particle and many-body level, to establish a formal mathematical framework for describing extreme nonlinear optical events beyond paraxial and envelope approaches, to explore a new class of novel 3D robust pulse propagation scenarios based on the manipulation of spatial and temporal degrees of freedom and to experimentally demonstrate the first ever table-top coherent X-ray source based on extreme nonlinear optical generation of more than 5000 harmonics,

Intense Field Quantum Photoionization

Recent breakthroughs in velocity imaging techniques make it possible to capture these very intense field photo-ionized electron/ion angular distributions with femtosecond resolution. The resultant angular momentum distributions display a sensitive dependence on the exciting optical carrier frequency/wavelength and carrier-envelope phase. The team has developed a hierarchy of full 3D quantum simulation models of localized intense few cycle pulse photo-ionization of atoms/molecules with the goal of quantitatively verifying these experimental observations and also of providing first principles light-matter coupling sources for propagation studies. The computed photo-ionized electron/ion angular distributions have shown agreement with the velocity imaging measurements (electron holograms) for a variety of pulse excitation conditions including carrier-envelope phase (CEP) sensitivity.

This rigorous quantum approach has also made it possible to unravel the fundamental physics of ultrafast nonlinear saturation, distinguishing between bound-bound and bound-free contributions to the nonlinear optical response. The saturation effect is always accompanied by some degree of photo-ionization in contrast to some recent claims in the literature. This study has enabled the team to begin to understand the contributions of both bound-free and freed electrons in modifying the local response to an intense applied laser pulse which additionally has deep implications for higher harmonic generation under extreme conditions. The team has successfully employed this quantum approach to confirm that the strong photo-ionized electron anisotropy evident in velocity map imaging and its subsequent evolution should be detectable via delayed mutually orthogonal optical probes.

On the experimental front, in support of this effort, the team has characterized the temporal phase and amplitude of an 800nm filament as a function of propagation length and laser intensity using transient grating XFROG measurements. These measurements form the first experimental map of the propagation dynamics within a filament. Further spectrally-resolved transient birefringence measurements confirmed grating-induced negative birefringence as the source of saturation rollover in support of their theoretical quantum simulations.

A simple analytic quantum model based on a contact potential with a single ground state coupled to a continuum provides a qualitative picture that reflects some of the key quantum signatures of first principles simulations. The goal is to further expand this model to include multiple bound states and use this as a first step in building a comprehensive EM pulse propagator. The single bound state + continuum model has been incorporated into a 1D EM propagator and preliminary results demonstrate qualitative agreement with many features observed in experiments. It represents the first stepping stone in introducing a new computational approach for femtosecond pulse propagation in the transparency region of gases that permits full resolution in three space dimensions plus time while fully incorporating quantum coherence effects such as high-harmonic generation and strong-field ionization in a holistic fashion.

A full 3D quantum Schrödinger solver has been developed and optimized to run in parallel across the multiple CPU cores on large machines available to the MURI team. The numerical approach can, in particular, capture the excitation and dynamics in a large number of excited states (incl. Rydberg states) and has been applied to various atoms as well as light fields covering a large range of frequencies, intensities, and pulse shapes. This program provides a testbed to validate the accuracy of various reduced mathematical models.

Nonequilibrium Evolution of Highly Anisotropic Plasmas Generated by and Coupled to Intense Electromagnetic Fields

The single particle angularly (momentum) resolved anisotropic photo-ionized electron distributions computed using the 3D quantum simulations have been used as initial data for the first full 3D quantum Boltzmann study of the evolution of nonequilibrium electron/ion distributions towards a quasi-equilibrium isotropic plasma state. The complexity of this 3D many-body problem has made it necessary to decouple the initial photo-ionized electron/ion distribution creation from the Coulomb mediated electron/electron and electron/ion scattering problem. The photo-ionized electron/ion initial data are generated using few cycle pulses where it is assumed that no significant mutual correlations are established in the initiation phase. Their results clearly show that for typical carrier densities in a filament in air, the loss of anisotropy takes on the order of a few picoseconds, a time typically longer than the duration of the ionizing pulse. They note the existence of two scales: 1) fast scattering of the electrons with the ions on the order of 50-100fs and 2) a slower scattering to an isotropic plasma state.

The team has further established that the strong anisotropy can be detected with a delayed THz probe by developing a model that extends the above quantum Boltzmann approach to perturbatively incorporate a weak probe THz field. The computed results predict that the probe absorption and refraction should show marked differences for different polarization states. These differences persist for times at least on the order of a few hundred femtoseconds. Very recent unpublished experimental work shows preliminary evidence of this THz probe anisotropy and further measurements are being planned to confirm the observations.

Many atom correlated quantum wavefunctions play a critical role in establishing the true nature of the coupling of the intense EM pulse to the partially ionized medium during propagation over extended paths. Additionally, such correlated quantum wavefunctions play a central and not fully understood role in promoting very high harmonic generation in high pressure gases. The team is currently extending the many-body theory to include simultaneous intense field photo-ionization and mutual correlation of photo-ionized electrons aimed at elucidating the nature of such fundamental physical interactions.

Asymptotics, Multiple Scales Analysis, Blow-up and EM Shocks

The team has derived a hierarchy of nonlinear partial differential equation (PDE) carrier-resolved propagation models that go beyond established nonlinear envelope propagators and allows for analysis of singular blow-up, carrier shock formation and automatically includes carrier-envelope phase (CEP), generalized linear and nonlinear dispersion, and quantum polarization sources. The formal equation hierarchy includes bidirectional vectorial nonparaxial propagation equations that enable them to identify novel scattering/generation events in an extreme nonlinear interaction zone. For example, they find that longitudinal electric field components directly sourced by nonlinear terms can become significant under conditions of extreme intensities, ultrashort pulses and strongly nonparaxial fields – these modes are somewhat analogous to classical plasma electrostatic fields but appear here in the absence of plasma and are nonlinearly sourced. The effect of such a mode polarized along the beam axis is to transport energy away from the collapse region and provide a novel additional source of collapse regularization. In the weakly-nonlinear, slowly-varying limit they show explicitly that the effect of the longitudinally polarized mode is to restrict the range of transverse modulationally unstable wavenumbers and to act as a defocusing lens in the collapse region.

A new class of beams, based on conical wave superpositions (Bessel, Airy, etc), representing a radical departure from conventional Gaussian beams, can be synthesized readily with linear optical components and exhibit a remarkable robustness to large perturbations (even opaque objects) encountered along their propagation path. The team has implemented singular perturbation methods to mathematically study the effect applying a finite Gaussian apodization to such an ideal nondiffracting wave and a multiple scales approach to derive amplitude equations for weakly nonlinear conical waves from a governing equation of cubic nonlinear Schrödinger type. From these equations they obtain asymptotic solutions for the linear and the weakly nonlinear problem from which several uniform estimates are obtained that describe the deviation from the ideal nondiffracting solution. Moreover, a numerical simulation based on an implementation of their amplitude equations supports their analytical results.

Experiments to explore means of propagating such beams have been carried out by team members. Vortex and Bessel beams have been generated using appropriately fabricated phase masks capable of handling the very high power densities in the initial ultrashort laser pulse at 800nm. Modulation instability along the most intense circular Bessel ring and vortex ring lead to the formation of rings of intense light filaments with accompanying white light and plasma generation. In parallel, an ignitor (intense femtosecond laser pulse) and heater (lower power but higher energy nanosecond duration pulse) have been employed in a series of experiments aimed at densifying the initially dilute plasmas generated by the ignitor pulse. Necklace beams in hollow waveguides, with discontinuous phase jumps of π around their circumference, have been shown to be resistant to filamentation far above the critical power for filamentation.

Filament Induced Coherent X-rays

Team members have generated bright high-harmonic x-ray supercontinua with photon energies spanning the EUV to 1.6 keV (<7.7 Å) by focusing 3.9-mm wavelength pulses from a tabletop femtosecond laser into a waveguide filled with He gas. This represents an extreme >5000-order nonlinear process while also demonstrating fully phase-matched frequency upconversion. They discovered that the multi-atmosphere pressures necessary for efficient x-ray generation also supports laser beam filamentation, enhancing the x-ray yield by another order of magnitude. They observe coherent, laser-like x-ray beams, despite the fact that ultrahigh harmonic generation occurs in a regime where the laser-driven electrons encounter many neighboring atoms before they re-encounter their parent ions. Preliminary calculations indicate that the kilo–electron volt—bandwidth coherent supercontinuum has a well-behaved chirp that, when compensated, could support a single–x-ray–cycle 2.5-attosecond pulse duration. Finally, they show that in the kilo–electron volt region, a much higher-order nonlinear process is required for phase matching than is required for harmonic emission from a single atom.

The very high gas density required puts these experiments in a regime of HHG from non-isolated emitters: Spread of the ionized electron quantum wave packet over its few-femtosecond free trajectory means that the electron will encounter many neighboring atoms. This contrasts with emission from dilute, isolated atoms for UV or EUV harmonic generation. For kilo–electron volt harmonics, the electron wave function in the continuum extends to ~500 Å, whereas the separation between the He atoms is ~15 Å at 10-atm pressure. However, the ionization levels are low at ~0.03%. For VUV/EUV harmonics, the electron typically extends ~2 to 20 Å between ionization and recollision, whereas the separation between atoms is ~70 Å at ~0.1 atm pressure, and phase matching occurs at ~10% ionization levels. Thus, HHG driven by mid-IR pulses liberates 0.001 as much of the electron wave function into the continuum compared with visible driving lasers, though it is spread over a 100-times-larger distance. Fortunately, their experimental results indicate that rescattering of this large and diffuse recolliding electron wave packet from other atoms seems not to adversely influence the coherence of the emission, likely because the medium is weakly ionized. Evidence for this includes the well-formed, spatially coherent x-ray beams and the remarkable quadratic growth that continues from 0.2 atm (when the rescattering electron wave packet can begin to encounter neighboring atoms) to more than two-orders-of-magnitude higher pressure.
In a second extremely favorable convergence of extreme nonlinear optics, the multi-atmosphere gas pressures required for phase-matched x-ray generation also overlap with the parameter range where laser pulse filamentation is possible. The measured x-ray beam profile also dramatically narrows as the gas pressure increases, indicative of filamentation of the driving laser. Essentially, the x-ray HHG beam, imaged at the exit of the fiber, shrinks to less than one-third of its former diameter, whereas the x-ray signal increases tenfold (integrated over all orders) at pressures seven times greater than those required for phase matching.

Spatial and Temporal Chirping, Dressed Optical Filaments and Remote Delivery

By exploiting the full spatial and temporal degrees of freedom of ultrashort laser pulses (USPs), the team has designed new classes of USPs with the unique property that they can be focused precisely in a tight focus – the latter focus does not move towards the source with increasing pulse power as observed with conventional USPs. The construction algorithm involves simultaneously imposing a spatial and temporal chirp (simultaneous spatial and temporal focusing SSTF) on the initial laser pulse and has the advantage that the pulse propagates essentially linearly over most of the propagation path until it reaches the target zone. This algorithm can be adapted to produce extended focused pulses or any general pulse spatiotemporal waveform if desired. Additionally, the team has explored the possibility that pulses with Bessel beam transverse cross-sections (or other beams with ring-like far-fields) can extend the self-trapped filament range in air by utilizing the Bessel (or other) beam as a dressing beam. The idea is to offset the weak loss of power from the filament due to plasma absorption that would allow the filament to drop below critical power, and sustain the profile above critical over extended paths. Their preliminary results indicate that the filament range can be extended by an order of magnitude or more. A very recent preliminary experiment by members of the team has confirmed that such beam dressing works over laboratory scales.

Experiments by team members have demonstrated robust propagation of femtosecond SSTF laser pulses and those with Bessel and vortex transverse profiles over the few meter propagation lengths available in the laboratory. Phase masks for vortex beams designed and fabricated in fused silica to withstand terawatt class pulses have been tested and will be delivered to the Air Force Research Laboratory TeraWatt laser laboratory at Kirtland AFB in Albuquerque for field testing. An ignitor-heater scheme, whereby an ultra-intense femtosecond laser pulse with moderate energy in the multi-mJ range creates a dilute seed plasma channel via filamentation, is heated and densified through the application of an delayed co-propagating nanoscecond multi-Joule laser pulse was demonstrated in the laboratory.

They were the first to successfully demonstrate the experimental application of this approach in ambient air.

Another important accomplishment of the project has been the initial theoretical demonstration that apodized Bessel, Vortex or other beams can be reconstructed remotely at km range by using an optimization algorithm based on an extension of the Gerchberg-Saxton Fourier integral relation. The essence of this algorithm is to assume a magnified far-field ring-like amplitude profile near the launch point and run an iterative mapping where the phase profile is allowed to vary such as to yield the desired solution at target. Their initial results show that they can stipulate an on-axis uniform extended amplitude profile at the target and obtain a convergent solution at km range. For example, they have explicitly shown that a Bessel beam at source can be imaged at 1km to give a near uniform on-axis intensity extending over 200m.

Lastly, they have performed the first studies of the phase properties of beams that undergo filamentation and find that the phase of the beam after undergoing filamentation is highly sensitive to the input energy of the pulses. This "loss-of-phase" effect results in the output phase effectively being random from shot-to-shot due to small fluctuations in the input pulse. These results are essential for controlling filaments and understanding the dynamics of multiple filamentation for beams that undergo multiple filamentation (e.g., necklace, Bessel, etc.)

Education and Outreach

The complex physics associated with USP filamentation in condensed media and gases requires that students and even senior researchers be educated in the most advanced techniques in theoretical physics and mathematics. As their understanding of these processes evolves, it is important that this knowledge be disseminated to the general body of researchers within and beyond the MURI program. The team has organized 2 intensive three-day March summer schools that were run immediately prior to the semi-annual review in Tucson in March 2011 and March 2012. The school topics covered advanced asymptotics, multiple scales analysis, nonlinear PDEs and computational methods under the Applied Mathematics heading, theory of intense field photo-ionization of atoms and molecules, quantum many-body physics of plasma evolution in gases and novel beams based on conical wave superpositions. In addition, team members have participated as lecturers in the STELLA "International School for Training in Experiments with Lasers and Laser Applications", June 20 to July 8, 2011 held at the University of Insubria, Como, Italy.

The 4-th International Symposium on Filamentation (COFIL 2012), hosted in Tucson from Oct 7-12, 2012 was partially supported through the MURI program and was chaired by the PI of the MURI. This symposium, held biannually since 2006, was the first to be held in the U.S. and brought together world experts on filamentation science from North America, Europe, and Asia. Prior meetings were held in Quebec, Canada (2006), Paris, France (2008) and Crete, Greece (2010). The MURI was well represented through a plenary talk, 10 invited talks and 8 poster presentations.

PLANS FOR THE OPTION YEARS:

Intense Field Quantum Photoionization

The quantum photo-ionization models developed under this heading represent the key enablers for understanding the ultrafast physics under conditions of extreme nonlinear optics. Moreover, they provide the critical underpinning for the derivation of mathematically reduced, but physically self-consistent, source terms for electromagnetic propagators and provide the necessary insight for resolving the details of higher-harmonic generation under extreme conditions. The team plans to reconcile the full 3D quantum Schrödinger approach with the alternative Strong Field Approximation – the latter treats the free electron and intense field coupling in zero order and develops a perturbative Green's function approach to include the Coulomb (or, for example, a short range delta, Yukawa etc) potential in an iterative manner. At lowest order, the problem couples the ground state of an atom or molecule to its continuum. An open problem that they plan to address is the level to which higher order iterations of the SFA include important bound state (bound-free and bound-bound) contributions to the nonlinear response. Operationally, the form of the SFA is much more amenable to incorporation as a quantum driving polarization source in a field propagation model. They will also explore quantum calculations of the nonlinear polarizability of the oxygen and nitrogen molecular ions produced by the leading edge of the filamenting pulse. Such polarizabilities form the basis for additional terms in the propagation equation to enable predictive calculations.

An important step in the Years 4 & 5 option is to systematically decouple the local ultrafast nonlinear physics from cumulative propagation effects in order that they identify the key quantum contributions and enable quantitative comparison to theory. They propose to extend this approach to measure quantum coherence and memory effects in ultrafast strong field ionization. Recently, it has become possible to generate ultra-fast waveforms with sub-cycle duration, with controllable temporal shapes. This can be achieved by coherent combination of several harmonics, with controlled spectral phases and powers. The result is a pulse-stream in which each of the peaks has a pre-defined shape. In particular, it is possible to expose a studied system to a suitably designed series of impulse-like excitations, study its response and how it depends on the previous history of excitation. Experiments will be pursued through collaboration with Professor D. Faccio at Herriot Watt, Scotland. The pulse synthesizer system capable of creating controlled waveforms will be available in D. Faccio's laboratory in the near future. It will be utilized as a drive for nonlinear response in rare gases and/or condensed media in conjunction with electrical or optical probing of freed electron densities. Similarly, optical probes can measure the effective nonlinear susceptibility. In the current approach, such observables should be the same for excitation with fixed spectral composition but different relative spectral phases. Yet, model calculation show that coherent memory effects should be observable through e.g. effective ionization rates.

The potential impact of such investigations is quite significant - it can be a new alternative way for probing nonlinear responses on sub-cycle time scales.

The full ab-initio quantum simulations, which offer a rigorous test of their mathematical models, will be used to identify the importance of bound-bound, bound-free, and free-free transitions on photo-ionization, high-harmonic generation and other nonlinear responses of the medium. In particular, the effect of resonant via non-resonant transitions as well as a careful and detailed characterization of the state of the medium during and after the interaction with a light pulse shall be studied. They further propose to extend their numerical approach to molecules. This requires first the inclusion of multi-center wavefunctions in the current basis-state expansion and then treatment of vibrational and rotational dynamics within the Born-Oppenheimer approximation.

Nonequilibrium Evolution of Highly Anisotropic Plasmas

The quantum Boltzmann formulism and simulation carried out in the first 3 years represents the first stage in the development of a comprehensive many-body description that includes quantum photo-ionization, electron-electron and electron ion scattering, all within a single physically self-consistent framework. In this study, they decoupled the initial intense field photoionization from the subsequent many-body scattering interactions. Anisotropic photo-ionized electron-ion distributions derived from a full single particle quantum Schrödinger problem with a few cycle driving field were used as initial data in their 3D quantum Boltzmann scattering simulation. Whereas such an approach is reasonably well justified for ultrashort, i.e. a few femtosecond long strong-field ionizing pulses, they become very questionable for pulse durations in the tens of fs to ps range. Here, the characteristic time scales for the field changes and carrier equilibration become comparable. In order to deal with these experimentally very relevant situations, the team will have to extend the formal theory to self-consistently treat the field-ionization problem in a fully interacting many-electron-ion system. Here, the ionizing field simultaneously interacts with many atoms/molecules such that the electrons of a given atom are subject to the Coulomb and exchange interaction with those of all the other atoms. After all, quantum mechanical electrons are fundamentally indistinguishable such that the many electron-ion system has to be in a properly antisymmetrized many-body state. By applying nonequilibrium quantum-kinetic theory, the team will derive coupled equations for the propagating field and the material excitations. The complexity of the generalized Maxwell-Bloch type many-body problem will necessitate that they develop massively parallel computational algorithms that take advantage of mixed multi-core CPU and GPU hardware on their newly acquired (AFOSR DURIP) SGI UV2 supercomputer. Specific challenges that will be addressed include novel algorithm development to reduce the computational complexity of full quantum Boltzmann scattering contributions – the latter will include exploration of GPU dedicated hardware and asymptotic reduction based on physical scale separations observed in full scale simulations.

In a parallel effort to decouple ultrafast light induced photo-ionization coupled to many body electron-electron, electron-ion correlations from propagation induced filamentation, they propose to employ a high finesse femtosecond enhancement cavity (fsEC) recently developed as a source for XUV frequency comb and higher harmonic generation. The latter system, whose resonance properties are polarization dependent, can support strong peak power ultrashort pulse trains capable of ionizing the nonlinear medium. It can be used as a sensitive diagnostic tool to measure and dynamically track the anisotropic photo-ionized electron-ion distributions with femtosecond resolution and can serve as a diagnostic tool in a pump-probe regime providing sensitive measurements of a thin nonlinear medium's instantaneous reaction to the strong-field pump - accumulated nonlinear phase changes are deduced from the resonance properties of the cavity with respect to the probe pulse train. The impact of such a capability will be to assess the overall accuracy of their theory/simulations and help them devise reduced scale mathematical models.

Spatial and Temporal Chirping, Dressed Optical Filaments and Remote Delivery

They plan to further explore the spatial/temporal chirping approach as a means of delivering controlled intense pulse waveforms to target regions at both short and long range. A nontrivial combination of spatiotemporal chirp control at the pulsed laser source may lead to novel and applications-oriented delivery of preformed 3D pulses to any desired target. In particular, they plan to test the feasibility of generating a "plasma mirror" to redirect microwave radiation by taking advantage of the SSTF scheme to create an array of linearly extended plasma channels in the laboratory. Other schemes to be explored include, for example, the use of the recently developed class of accelerating autofocusing wavefronts--capable of abruptly delivering all their energy on target. These effects can be further enhanced using chirped waveforms for dispersion control.

In the context of remote delivery at km ranges, the team will need to address the following important theoretical and experimental challenges:

- Extend the study to include dispersion compensation

It will be very important to generalize the Gerchberg-Saxton mapping to the broader problem of full space and time dispersion compensation. It is well established that imposing a negative frequency chirp on an ultra-short laser pulse with a Gaussian-like transverse profile will delay the onset of critical self-focusing (critical collapse singularity) to further down range. In effect, one disperses the temporal waveform such that the long pulse sees a "linear medium" and through remote compression generates the full nonlinear effect at a desired remote location. While this simple "zero-order" negative chirp is still likely to work for the more complex 3D pulse waveforms that they are investigating here, it is likely that the very strong spatiotemporal coupling (space and time contributions cannot necessarily be written as a direct product) along the propagation path will require the creation of a new paradigm for simultaneous space/time dispersion compensation. The team proposes to further explore means of dispersion compensation along these lines keeping in mind that the transverse beam profiles may initially be nontrivial linear combinations of conical waves that become nonlinearly coupled under propagation. The use of an energy reservoir in time (time-dressing schemes) in order to further prolong the longevity of an optical filament will also be pursued during the course of this work. This could be accomplished by utilizing specially designed optical pulse sequences.

- Mitigation against turbulence for extended ground level or vertical pulse delivery

Their current work indicates that a robust transverse beam waveform may be generated remotely at a remote target location. However, their initial calculations are restricted to linear propagation over extended paths without taking into account atmospheric turbulence. Moreover, they have ignored the dispersion of air on sustaining a targeted pulse temporal waveform and the potential that nonlinearity, although very weak, may have a cumulative influence on the remotely generated 3D laser pulse waveform. The team has access to expertise in designing adaptive optics systems and plans to calculate the effect of turbulence over both horizontal and vertical pathways and explore an adaptive optics solution for turbulence mitigation. Preliminary studies have suggested that specially designed wavefronts in the form of multi-beamlet Airy beams can substantially reduce atmospheric scintillation effects. They intend to explore these possibilities at both the theoretical and experimental level.

Asymptotics, Multiple Scales Analysis, Blow-up and EM Shocks

The main thrust of this part of the MURI effort in Years 4 & 5 will be to seek a unified mathematical description of quantum photo-ionization and many-body interactions that dispenses with current cumbersome and computationally intractable methodologies. The resultant light-matter models will retain all of the relevant physics while taking advantage of separations of spatial and temporal scales based on “lessons learned” from numerical simulations and experimental diagnostics. The strong field approximation (SFA) model, while more tractable from a computational perspective, still does not offer a sufficiently computationally transparent alternative to existing phenomenological approaches in EM propagation codes.

The extreme nonlinear interactions associated with the generation of 5000 harmonics-or more-of the driving field in coherent soft X-ray generation will require the development of new approaches to resolving blow-up singularities and carrier-wave shocks that lead to huge spectral broadening with concomitant attosecond/zeptosecond pulse formation. The team will extend carrier-resolved PDE models developed in years 1-3 and guided by numerical analysis, will develop time domain models capable of automatically resolving sub-cycle shock formation within the propagating pulse.

A second effort will be devoted to a systematic mathematical study using multiple scales analysis of the mutual interaction between multiple filaments being seeded by a broad quasi-linear background. Understanding how filaments interact will provide crucial insight into sustaining multi-filamentary patterns, seeded for example on an intense vortex ring, as extended plasma waveguides for guiding high-power microwaves over significant propagation ranges

Filament-Induced Coherent X-rays

Their research breakthrough in generating coherent soft X-rays from a mid-IR femtosecond source in the first three years of the project opens up a vast scope of potential applications and raises many unanswered fundamental physics questions. In particular, simulation of intense pulse filamentation in the high pressure capillary was limited to the case of HHG in Argon where experimentally they were able to observe a significant enhancement in beam quality and HHG flux (x7) due to filamentation. These simulations stressed their computational resources and they were forced to decouple the filament generation from the higher harmonic generation. In the next two years the team will investigate applying more sophisticated USP carrier-resolved propagation codes, coupled to quantum strong field material models to extend the theory/simulation to the He case where 5001 harmonics and beyond are generated. They will also compare these simulations with experiment for validation. Finally, they will explore filamentation in waveguides in different gases for different driving laser wavelengths to uncover a comprehensive, global understanding.

They will also investigate the photoelectron angular distribution as a result of strong field ionization using mid-infrared driving lasers. Very recent experimental and theory has shown that this distribution is very anisotropic and that some electrons are trapped close to the parent ion for many laser cycles before escaping. These electrons may excite the atom, leading to a different nonlinear response during filamentation.

Education and Outreach

The strong educational theme of the MURI will be continued with an international summer school on the "Theory and mathematical modeling of ultrashort pulse propagation" to be held at University College Cork, Ireland from July 28, 2013 to August 2, 2013. The school lecturers will be drawn from the MURI team and the same topics covered but in greater detail. The goal of this school is to bring together junior and senior personnel from the large European and U.S. groups who are world leaders in filamentation science.

Local MURI 3-day schools will be continued in March 2014 and 2015 in Tucson.

Applications.

TRANSITION PLANNING:

The scientific objective of the MURI program is to develop first-principles, rigorous understanding of the interaction of ultra-short intense laser pulses with gaseous and condensed media. While pursuing this fundamental objective, the MURI team leadership has contacted candidate users of the developing capabilities in the Department of Defense and in the civilian industrial sector.

There has been transition planning and substantial scientific and consultative contact with three military user candidates: The Directed Energy Directorate at Kirtland AFB, the Sensors Directorate at Wright Patterson AFB, and SOCOM in Tampa, Florida.

At Kirtland AFB, the MURI team is designing and testing custom hardware for generating TW-class pulses with robust transverse profiles. A fused silica phase mask to generate vortex transverse profiles will be delivered to AFRL for field testing at multi-km range. The MURI effort will subsequently transition the results of developed remote delivery methodologies following turbulence mitigation studies demonstrated at laboratory scale. Working with AFRL at Kirtland, the MURI team will explore pre-chirp to extend plasma propagation distances. Personnel at AFRL/Kirtland are interested in coupling radiofrequency and microwave signals to plasma channels to provide low probability of intercept communication capability and remote surface interrogation capabilities. Additionally, the plasma filament is a conductive path, and as such can assist in the highly local delivery of electromagnetic energy for the purpose of interacting with a target either for benign communication or destructive illumination.

The MURI effort has suggested that an array of filaments can serve as an effective mirror for radar radiation. This filament property may provide a useful tool to augment synthetic aperture radar capability. By properly placing the plasma filament array a transmitting aircraft can receive forward scattered as well as backscattered radar returns and this dual information path can significantly enhance the quality of military (and environmental) imaging. This development has been presented in some detail to the Sensors Directorate at Wright Patterson Air Force Base and a continuing dialogue is occurring as the capability develops. The radar processing scheme has already been developed and the physics of plasma filament array propagation is increasingly well understood within the MURI.

The United States Special Operations Command (USSOCOM) SOF Warrior project office, McDill AFB, Tampa Florida, seeks augmented visual capability for special operations activities. MURI personnel have visited the SOF Warrior Project office and briefed on optical and microwave capabilities. A need for wide area and "over the hill" optical viewing has been defined by the special operators and other combatants. A plasma ball launched upwards on a proper slant angle can serve as a spherical mirror and permit "over the hill" and wide area viewing. MURI leadership perceives this as an important transition path for the MURI science products.

The work of Professor Margaret Murnane in this MURI has resulted in a table top X-ray source. It is the MURI leadership's plan to present this emerging technical capability to the Materials Directorate at Wright Patterson Air Force Base. The table top capability provides a ready characterization of material structures and the study of material fatigue. The development of this table top capability coupled to the recent development by two groups addressing the phase problem in X-ray spectroscopy (Emil Wolf at Rochester and Emmanuelle Candes at Stanford) may lead to intensified progress in materials science.

The first three transition paths have involved personnel contacts between the MURI and potential users. The table top X-ray capability of professor Murnane has been related to the Materials Directorate program at Wright Patterson Air Force Base, but personnel contacts have not been made yet since the capability is not ready for daily laboratory use.
The following transition patterns have seen substantial discussion within the MURI but have not yet been related to a specific user.

When a plasma beam is created above ground level it will dissociate water in aerosols and excite impurities held in aerosols, specifically alkali elements such as sodium and potassium. Some of the excitation states of these elements consist of closely spaced doublets. These excited states can be acquired by an atomic line filter which serves for optics as a lock-in amplifier which detects narrowband signal in very high background noise. The exciting plasma would not be seen during the day. At night, non-visible laser light can be used without visible plasma. The transmitting system comprises the laser system and message encoding and may require a jeep-sized vehicle. The receiving system, built around the atomic line filter, could be potentially two-man portable or jeep portable. This communication system is considered for non-line-of-sight low probability of intercept communication.

Excitation conditions within the atmosphere air-based filament plasmas studied in this MURI result in electron temperatures in the 1eV range and associated electron densities of $\sim 10^{16}$ per cubic centimeter. The optical power densities within the nominal 100 micron diameter filaments are $\sim 5 \times 10^{13}$ watts per square centimeter. These power densities are sufficient to readily dissociate and excite molecular fragments and atoms. Interception of such plasma filaments with chemical or biological agents can thus provide spectral signatures that can be used as agent identifiers.

PUBLICATIONS AND PRESENTATIONS AND INDIVIDUAL AWARDS:

47 papers in archival journals – excludes conference proceedings

FULL LIST OF PUBLICATIONS:

2012

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Charles G. Durfee, Michael Greco, Erica Block, Dawn Vitek, and Jeff A. Squier, "Intuitive Analysis Of Ultrafast Pulse Propagation Through Angularly Dispersive Structures," **invited paper** to be published in New Developments in Photon and Materials Research, ed. J. I. Jang (NOVA Science, 2012).

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INDIVIDUAL AWARDS:

Professor Margaret Murnane, a winner of multiple prestigious awards, including a MacArthur Fellowship in 2000, won the following awards during the life of the MURI:
2012 Willis Lamb Award for Laser Science and Quantum Optics (shared with Henry Kapteyn)
2012-2014 Chair, President’s Committee for the US National Medal of Science
2011 Boyle Medal of the Royal Dublin Society
2010 Appointed to the President’s Committee for the US National Medal of Science
2010 R.W. Wood Prize of the Optical Society of America (shared with Henry Kapteyn)
2010 Arthur L Schawlow Prize in Laser Science by American Physical Society (shared w Henry Kapteyn)

15 Graduate and 3 undergraduate students have been supported by this MURI grant.
12 Post Docs (4 part-time) have been supported by this MURI grant.

ISSUES OF CONCERN:
No issues of concern have been identified by any of the panelists at the most recent review.

RECOMMENDATION FOR EXERCISE OF OPTION:
It is unequivocally recommended that this MURI be funded for the 4th and 5th years.