Coherent Control of Multiphoton Transitions in the Gas and Condensed Phases with Shaped Ultrashort Pulses

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Marcos Dantus
Department of Chemistry and Department of Physics, Michigan State University, East Lansing MI 48824
dantus@msu.edu

1. Program Scope

Controlling laser-molecule interactions has become an integral part of developing devices and applications in spectroscopy, microscopy, optical switching, micromachining and photochemistry. Coherent control of multiphoton transitions could bring a significant improvement of these methods. In microscopy, multi-photon transitions are used to activate different contrast agents and suppress background fluorescence; coherent control could generate selective probe excitation. In photochemistry, different dissociative states are accessed through two, three, or more photon transitions; coherent control could be used to select the reaction pathway and therefore the yield-specific products. For micromachining and processing a wide variety of materials, femtosecond lasers are now used routinely. Understanding the interactions between the intense femtosecond pulse and the material could lead to technologically important advances. Pulse shaping could then be used to optimize the desired outcome. The scope of our research program is to develop robust and efficient strategies to control nonlinear laser-matter interactions using ultrashort shaped pulses in gas and condensed phases. Our systematic research has led to significant developments in a number of areas relevant to the AMO Physics group at DOE, among them: generation of ultrashort phase shaped pulses, coherent control and manipulation of quantum mechanical states in gas and condensed phases, behavior of isolated molecules under intense laser fields, behavior of condensed phase matter under intense laser field and implications on micromachining with ultrashort pulses, coherent control of nanoparticles their surface plasmon waves and their nonlinear optical behavior, and observation of coherent Coulomb explosion processes at 10^{16} W/cm^2. In all, the research has resulted in 36 publications (five journal covers) and nine invention disclosures, five of which have continued on to patenting:

Method to decode the spectral phase of ultrafast pulses, Marcos Dantus, 6/10/2002
Laser Selective Excitation, Marcos Dantus, Vadim Lozovoy, 1/27/2005
Laser Material Processing System, Marcos Dantus, 11/29/2005
Phase characterization and correction for ultrashort pulses, Marcos Dantus, 2/22/2006
Laser plasmonic system, Marcos Dantus, Jess Gunn, 5/5/2006

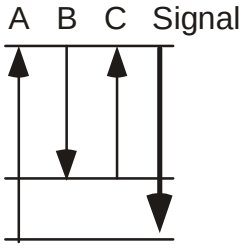
From these the DOE has indicated special interest on Laser Material Processing.

2. Coherent control with Four-Wave Mixing

We began our work with the manipulation of electronic and vibrational quantum states in iodine vapors with the ultimate goal of demonstrating molecule-based information processing. In those experiments we used three beams that were crossed in the Forward-Box geometry. Beam A is used to launch a wave packet in the excited electronic state, beam B is a shaped 800 nm pulse that controls the stimulated Raman transition to high vibrational levels of the ground state (v~22-24). The beam C brings the wave packet back to the excited state where it emits the FWM signal, which is spectrally dispersed, and time integrated. Our approach to coherent quantum state manipulation based on the photon echo process published in 2002 (shown in Fig. 1) is still considered one of the best approaches to optical manipulation of quantum states. This is because the photon echo signal is independent of inhomogeneous broadening. This leads to the observation of much longer coherence times.

Fig. 1

Experimental Data Obtained with the VIrtual Echo Setup



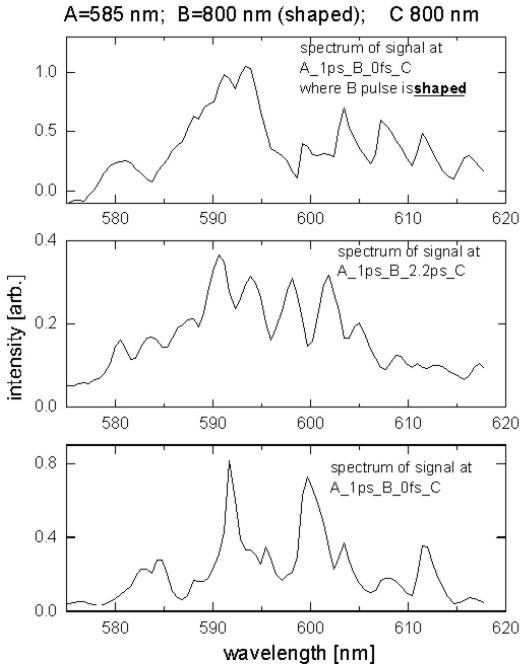


Fig. 1. Experimental data for a VE setup. The *pump* (A) was centered at 585 nm, the *Stokes* beam (B) was shaped and centered at 800 nm, the *probe* beam (C) was centered at 800 nm, and the signal was centered near 590 nm. The signal beam was frequency dispersed. Three different spectra resulting from different conditions are shown. The variables for each spectrum are indicated in the upper right corner. Clearly the time between the pulses and shaping of the *Stokes* beams changes the outcome in the observed signal.

We have made a number of measurements regarding the electronic decoherence rate in iodine vapor. These measurements are required to determine the ratio between the time it takes for coherence to be lost through random collisions and the time it takes to coherently manipulate information (write, compute, read). Our measurements have led to measurements of the fundamental decoherence cross section in the presence of different buffer gasses. The electronic dephasing cross sections are extremely large even compared to Van der Waals radii. Systematic measurements of the rate of decoherence were made as a function of buffer gas. From a number of such measurements, we calculated experimental cross sections for the long-range interactions. These values range from 144 $Å^2$ for Helium to 1370 $Å^2$ for I_2. We find that the cross sections can be modeled by dispersion interactions and show no dependence on the number of degrees of freedom in the buffer gas molecules. We reviewed the theory of electronic coherence dephasing and obtained an expression that can be reduced to the gas phase and the liquid phase limits. Despite the large values for electronic dephasing cross section, we find that even at room temperature we can obtain a coherence ratio in excess of 10^4, and using a molecular beam in excess of 10^7.

3. Coherent control in condensed phases

In 2002-2003 when we carried out these measurements, it was still unclear if one would be able to achieve coherent control in large molecules in solution. Our research based on understanding the physics of nonlinear optical processes, allowed us to achieve the desired control. Multi-photon transitions are optimized when the central bandwidth of the laser pulse, ω_0, is some fraction (half for two-photon, a third for three-photon etc.) of the total energy of the transition. The effect of pulse shaping on the probability amplitude for two-photon absorption (2PA) can be calculated as follows assuming sharp transitions,

$$A_2(\Delta) \propto \int_{-\infty}^{\infty} E(\Omega)E(\Delta-\Omega)\exp[i\{\varphi(\Omega)+\varphi(\Delta-\Omega)\}]d\Omega \qquad (1)$$

For three-photon absorption (3PA) a similar formula can be derived,

$$A_3(\Delta) \propto \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} E(\Omega_1)E(\Omega_2)E(\Delta-\Omega_1-\Omega_2)\exp[i\{\varphi(\Omega_1)+\varphi(\Omega_2)+\varphi(\Delta-\Omega_1-\Omega_1)\}]d\Omega_1 d\Omega_2 \quad (2)$$

where amplitudes and phases are introduced for two different detuning values Ω_1 and Ω_2, and $\Delta = \omega - n\omega_0$. The signal resulting from an n-photon absorption process is calculated using the equation

$$S_n \propto \int_{-\infty}^{\infty} g(\Delta)|A_n(\Delta)|^2 d\Delta, \quad (3)$$

which convolves the spectral amplitude of the laser pulse with the absorption spectrum of the molecule $g(\Delta)$. A rational approach to introduce intrapulse interference in multiphoton transitions of large molecules requires phase functions that are comparable to the homogeneously broadened absorption spectrum and the spectral width of the pulse. Conversely, in the time domain, the shaped pulse should be comparable to the relaxation dynamics of the material. We have been using a formula for defining the phase function that has a limited number of parameters,

$$\varphi(\Omega) = \alpha Cos(\gamma\Omega - \delta) + \tfrac{1}{2}\beta\Omega^2, \quad (4)$$

where α is the phase amplitude, β is the quadratic chirp, γ is the modulation in the frequency domain $2\pi N/(\Omega_{max} - \Omega_{min})$ with period N, and δ is the position of the phase mask with respect to the center of the pulse spectrum.

The results in Figure 2 show the changes in two- and three-photon induced fluorescence as a function of scanning the chirp parameter β and the mask position δ. The data were obtained using a femtosecond laser system producing 810 nm, ~ 50 fs pulses, shaped using a spatial light modulator at the Fourier plane of a zero-dispersion stretcher. The shaper was calibrated so that only phase delays were introduced without changes to the output spectrum, intensity, or polarization. The pulses 0.5-1 μJ were focused onto quartz cells with a 200 mm focal length lens.

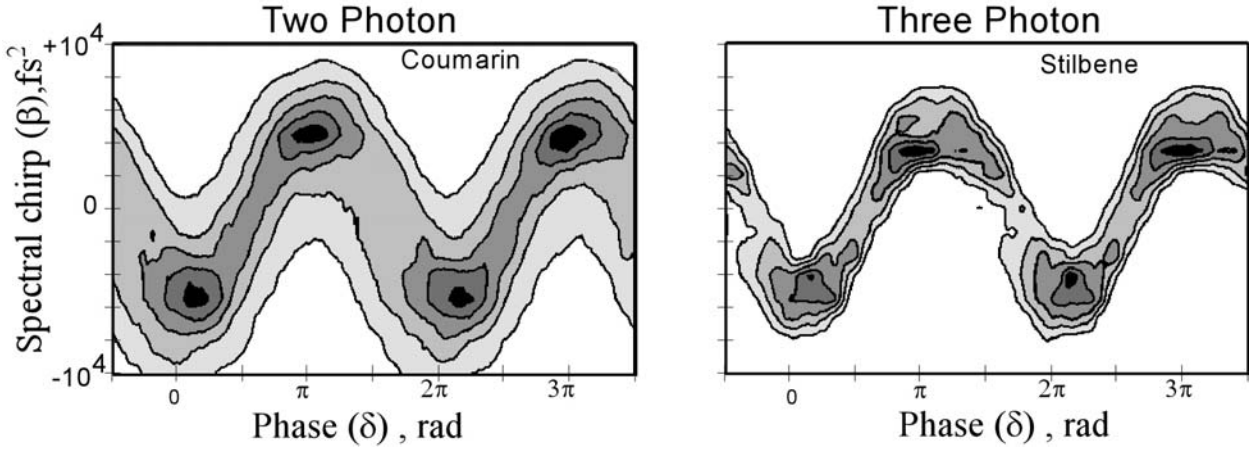


Fig. 2. Experimental measurement of the two- and three-photon induced fluorescence as a function of spectral chirp and phase mask position.

The data in Fig. 2 indicates that this concept of phase modulation originally proposed for controlling atomic transitions, is useful for controlling large molecules in condensed phases. Although the dependence of two- and three-photon induced fluorescence on the phase mask parameters is similar, there are some differences. The differences allow one to design a phase mask that can enhance one process while the other is suppressed. We have demonstrated control of multiphoton processes including continuum generation using this rational approach towards pulse shaping. We have tested this method on large organic molecules including green fluorescent protein, and concanavalin A protein.

4. Manipulation of multiple quantum mechanical states in small isolated molecules

The goal of these experiments is to explore controlled excitation and manipulation of electronic and vibrational wave packets by coherent nonlinear optical methods. The research has focused on gas phase iodine where four-wave mixing (FWM) methods have been used to coherently prepare vibrational wave packets in the ground and excited states. That work, together with a thorough theoretical analysis, was published in collaboration with Mukamel. One

of the motivating aspects was the possibility for storing and manipulating information in a quantum mechanical system. The project called for accurate measurements of the electronic decoherence rate (the time over which one can manipulate quantum states coherently without loss of information). We have measured the cross section for the long-range elastic collisions responsible for decoherence in neat iodine and for iodine in the presence of a number of buffer gases. These fundamental measurements provide a foundation for understanding the dephasing interactions between molecules at distances that are up to ten times their van der Waals radii. Based on very encouraging results, we proposed a method for the coherent storage and manipulation of information that maintained coherence for nanoseconds. The setup combined a three-pulse photon echo arrangement and a pulse shaper. With this system, we showed that the spectrum of the signal beam provided a reliable output, reporting on the different transitions induced by the shaped second pulse. This work has some similarities to the work on quantum computing using multiple pulse NMR. However, we have pointed out a number of advantages for using an optically based method. In the optical domain, quantum transitions are significantly more energetic than kT. NMR transitions are about eight orders of magnitude less energetic. This difference eliminates thermally induced errors in an optically based system even at room temperature. The train of optical pulses takes a couple of picoseconds instead of several seconds for the RF pulses needed in NMR, making an optical based system much faster. The next step in our research project is to determine experimentally the accuracy with which one can encode two arrays of numbers and read out their product. These measurements will determine the scalability limits of the setup. This research project will be continued during the next granting period.

We carried out a research project on the molecular dynamics and interconversion between NO_2 and N_2O_4. The study was carried out using the transient grating method. We observed rotational and vibrational wave packet dynamics that allowed us to determine the rotational constants of the molecules. We also observed the symmetric stretch of the unusually long N-N bond in N_2O_4. Our most significant finding was the absence of half revivals in the rotational wave packet dynamics of N_2O_4. This absence implies that the molecule is not perfectly planar as currently thought. Half revivals are only observed in molecules where symmetry imposes different population of even and odd rotational levels based on nuclear spin statistics. We observed that a large number of the N_2O_4 molecules dissociated into two NO_2 molecules in the presence of the far off-resonance laser fields. This implies that the barrier to dissociation is very small or not existent for certain geometrical configurations. We plan to follow up this study with additional measurements and collaboration with Professor Piotr Piecuch, who will calculate an even more accurate ground state potential energy surface for this molecule using the method of moments. This project will be completed before the end of the present granting period.

5. Coherent control of large molecules in condensed phase

Instead of following the approach based on pulse shapers and learning algorithms suggested by Rabitz and implemented by Wilson, Gerber, Levis, Bucksbaum, and others, our approach towards coherent control of large molecules in the condensed phase has followed a completely different strategy. We have paid attention to changes in the power spectrum that are the direct result from pulse shaping. For a particular electric field E(t) inducing an n^{th} order optical transition, one can calculate the n^{th} order power spectrum. For example, the second order power spectrum contains frequencies proportional to $2\omega_0$, which are responsible to for two-photon excitation. The power spectrum depends on the spectral phase of the femtosecond pulses. This dependence can be calculated, therefore, the phase required to optimize a specific transition can be calculated and implemented using a pulse shaper. We have used this principle to demonstrate experimentally coherent control of two-, three- and higher order multiphoton excitation of large molecules including laser dyes and proteins. We have expanded on this technology to demonstrate selective excitation of probe molecules in different micro-chemical environments. We have, more recently, demonstrated selective two-photon microscopy (see Figure 3).

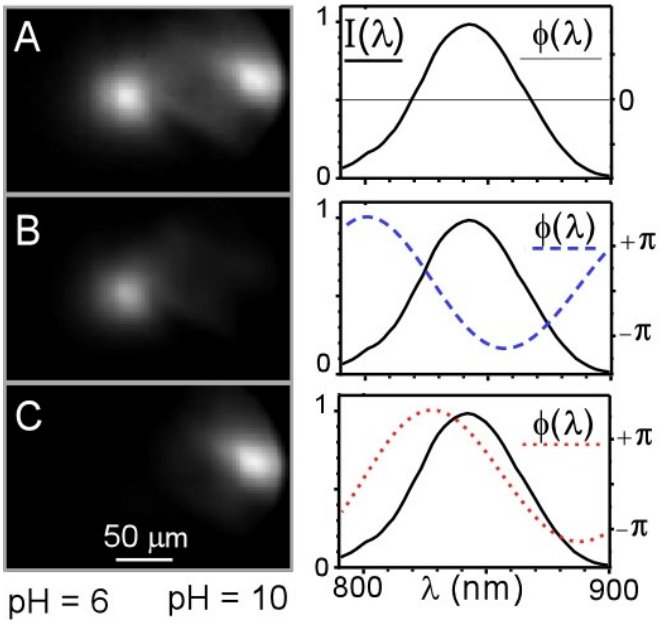


Figure 3, Experimental demonstration of pH-sensitive selective two-photon microscopy. The sample imaged has an acidic (left side of the frame at pH 6) and a basic (right side of the frame at pH 10) region, both labeled with HPTS. **A** Image of the sample obtained with transform-limited pulses. The diagrams on the right show the spectrum of the 21-fs laser pulses, centered at 842 nm, and the spectral phase of the pulse (blue dashed line or red dotted line, that maximize pH 6 or pH 10 fluorescence, respectively). **B** Image of the same sample and location obtained with pulses that have been optimized for selective excitation of HPTS in an acidic micro-environment. For this image $\alpha=1.5\pi$, $\gamma=20$ fs, and $\delta=0.75\pi$. **C** Image of the same sample and location obtained with pulses that have been optimized for selective excitation of HPTS in a basic micro-environment. For this image $\alpha=1.5\pi$, $\gamma=20$ fs, and $\delta=0.25\pi$.

6. Characterization of ultrashort pulses using MIIPS

The work described above would not have been possible without a significant amount of effort being invested in the development of pulse characterization and pulse shaping technology in our group during the present granting period. Our present setup is capable of taking in ultrashort femtosecond pulses, characterizing the phase distortions, compensating for the distortions and rendering transform limited pulses shorter than 5 fs in duration. These steps are now done automatically using a method developed by our group, that we have called multiphoton intrapulse interference phase scan (MIIPS). This method was patented and is now commercially available. It is being used in more than 20 femtosecond laser laboratories in different parts of the world. The impact of this work spans physics, chemistry, electronic engineering, materials processing, and biomedical imaging.

MIIPS has turned out to be an extremely accurate method for measuring arbitrary phase distortions in the spectral phase of femtosecond pulses with unprecedented 0.5 fs^2 accuracy. Because MIIPS uses a pulse shaper, it can compensate spectral phase distortions to produce routinely transform limited pulses within 0.3% of the theoretical limit. We have compared MIIPS to other widely used methods for phase retrieval and MIIPS has been found to be more accurate than SPIDER, an order of magnitude more accurate than FROG, and even more accurate than white-light interferometry for measuring group velocity dispersion. MIIPS is now integrated in our laser systems for automatically optimizing the output until it is transform limited, and then carrying out phase shaping projects. The figure on the right shows an SHG FROG trace on the left and a MIIPS scan for the same 18 fs, transform-limited pulses.

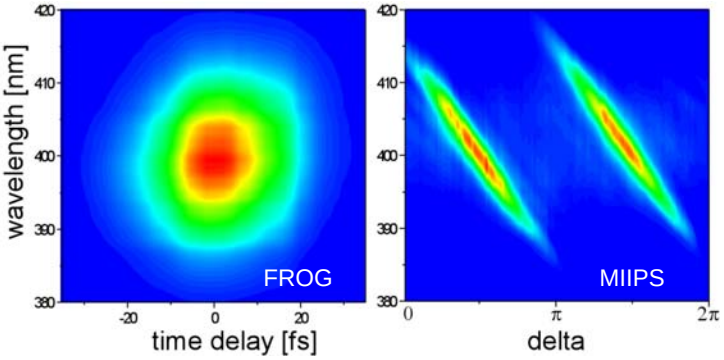


Figure 4, Experimental results obtained from 18 fs TL pulses using SHG-FROG and MIIPS.

7. Selective excitation using binary phase shaped pulses

While most scientist in the field of laser control decided to pursue optimization projects using computer-controlled evolutionary learning algorithms, we decided to fully understand the physics of nonlinear optical processes and to develop a methodology to achieve the optimization. Our efforts paid off. Given a 128 pixel pulse shaper capable of phase and amplitude modulation, there are 10^{300} different pulses possible. Evaluating these different pulses would take times that are many orders of magnitude greater than the lifetime of the universe. However, we have learned that all one needs is to select between two phase values (zero and π) for each pixel and no amplitude control is needed. This reduces the search space immediately by 262 orders of magnitude.

We embarked on a systematic study of the effects of phase shaping on multiphoton transitions. This study primarily focused on selective two-photon excitation, the competition of two- versus three-photon transitions, and on selective CARS excitation. This study explores the most efficient methods for controlling multiphoton transitions. Starting from first principles we review a number of different phase functions and emerge with very valuable conclusion. The most valuable being that selective excitation requires the pixels of the pulse shaper to take only two phase values, and not a whole range of values as previously believed. Binary phase shaping, when the difference between the two phase values is π, represents a quantum leap in our progress towards controlling multiphoton processes and the design of robust applications using coherent control. Theoretical work in our laboratory has progressed at the same rate as our experiments. We are presently able to calculate optimum phase functions using a computer operating at 3 GHz, and then implement those phase functions in the lab. We can alternatively make experimental measurements and also simulate them accurately using theory. To demonstrate our present capabilities we evaluated 2^{16} different 16-bit binary phase functions for their ability to generate SHG intensity at 400 nm within a narrow 0.5 nm spectral width, while keeping the background SHG outside that spectral window to a minimum. The results from that research project (shown in Figure 5) are mapped on the left (theoretical prediction on the left and experimental result on the right). The measured figure of merit ranged from red (factor of 2.5 signal-to-background) to black (factor of 0.5 signal-to-background). Beyond these results, we learn from our theory that optimal selectivity depends on binary numbers with minimal correlation. Finding numbers with these properties with more than 10 bits is not a trivial task. However, these numbers are used for cryptography and for secure communications. Based on the availability of published numbers with up to 213 bits, we are now able to simply write the optimal solution without the need of experiments. In summary, we went from the need to evaluate 10^{300} possible phases, to evaluating only10^{38} phases, to finally not having to carry out experiments because we know what the optimal answer is. This research published in Phys. Rev. A (pub. #20) and is revolutionary. It will probably take five years for other groups in the field to realize that the problem of optimizing low-order (2-3) nonlinear optical processes has been solved.

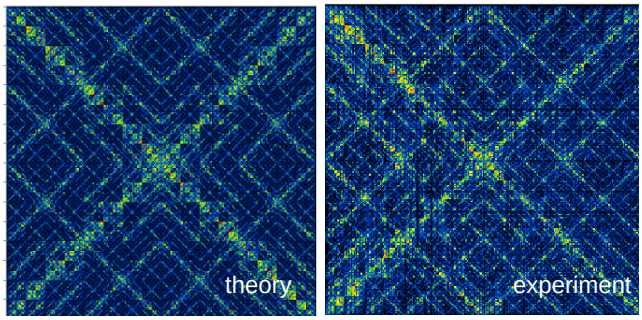


Figure 5, Proof that binary phase shaping optimally achieves control over second order nonlinear optical excitation. Results from 2^{16}=65536 calculations and experiments are in perfect agreement on the selective two-photon excitation at a narrow frequency using 16-bit binary phases.

The systematic evaluation of binary phase functions and mapping the results has now become a standard in our group that we have applied to a number of projects such as the identification of chemical warfare agents. This work had a significant impact when we applied it to the standoff detection of explosives through a JIEDDO funded project. We demonstrated our ability to selectively excite single vibrational modes of molecules in thin films placed 36 ft away from the laser system. This project is also several years ahead of any other group in the world.

Projects using selective multiphoton excitation on large molecules led us to demonstrate selective two-photon microscopy. In these projects, we demonstrated selective excitation of two different chromophores or a single chromophore but in different environments. The latter experiment sparked the idea of using phase shaping for functional imaging. The goal was to use a two-photon active chromophore that was sensitive to a chemical gradient such as pH. We then use phase shaping to control the excitation of the chemically sensitive chromophore. Results from the initial measurements were very encouraging, especially when working with HPTS, a large organic molecule with a pH dependent two-photon cross section spectra that always emits at the same wavelength. While we

have optimized two-photon microscopy with ultrashort (10 fs) pulses, most groups in the world are still struggling to get sub-30 fs pulses through a high numerical aperture objective.

8. Effect of pulse shaping on micromachining

Our ability to control nonlinear optical excitation at low and intermediate intensities had to be tested at higher laser intensities (10^{14} to 10^{18} W/cm^2) where perturbation theory is no longer applicable. This research started by exploring the interaction of intense femtosecond pulses on metallic surfaces [12]. We focused the beam on a continuously refreshed surface (rotating the target) and measured the laser induced breakdown spectroscopy (LIBS) atomic emission. Because we used 30 fs pulses, we noticed that the threshold for LIBS was very low, far lower than the threshold reported by groups using longer pulses. Instead of requiring ~100 mJ of pulse energy we were able to obtain stable signals with ~5 μJ of energy per pulse. When we explored different pulse shaping strategies we found very small effects compared to those found earlier for multiphoton excitation. Although some degree of selectivity between copper and aluminum surfaces was found when using binary phase shaping, the extent of the effect was very modest (~30 %).

The most intriguing finding was that as 35 fs pulses were stretched to a pulse duration of 10 ps, the overall LIBS signal did not disappear; it actually was ~20% higher than for transform limited pulses. This was a surprising finding because the laser intensity was near threshold and we had assumed peak intensity was critical to observe LIBS near threshold. If the pulse duration of ultrashort pulses was not responsible for the very low LIBS threshold we observed, then it had to be their broad bandwidth. We tested this conclusion and found that the LIBS threshold depends on the inverse of the bandwidth of the pulses (see right panel of figure). This observation has important implications in the field of micromachining and surface processing by lasers. We now know that the reason for the higher signal obtained with chirped pulses comes from more efficient melting and evaporation achieved by the longer pulses. This is demonstrated by the results in Figure 6 obtained for 35 fs TL (top) and chirped (bottom) pulses with the same energy per pulse.

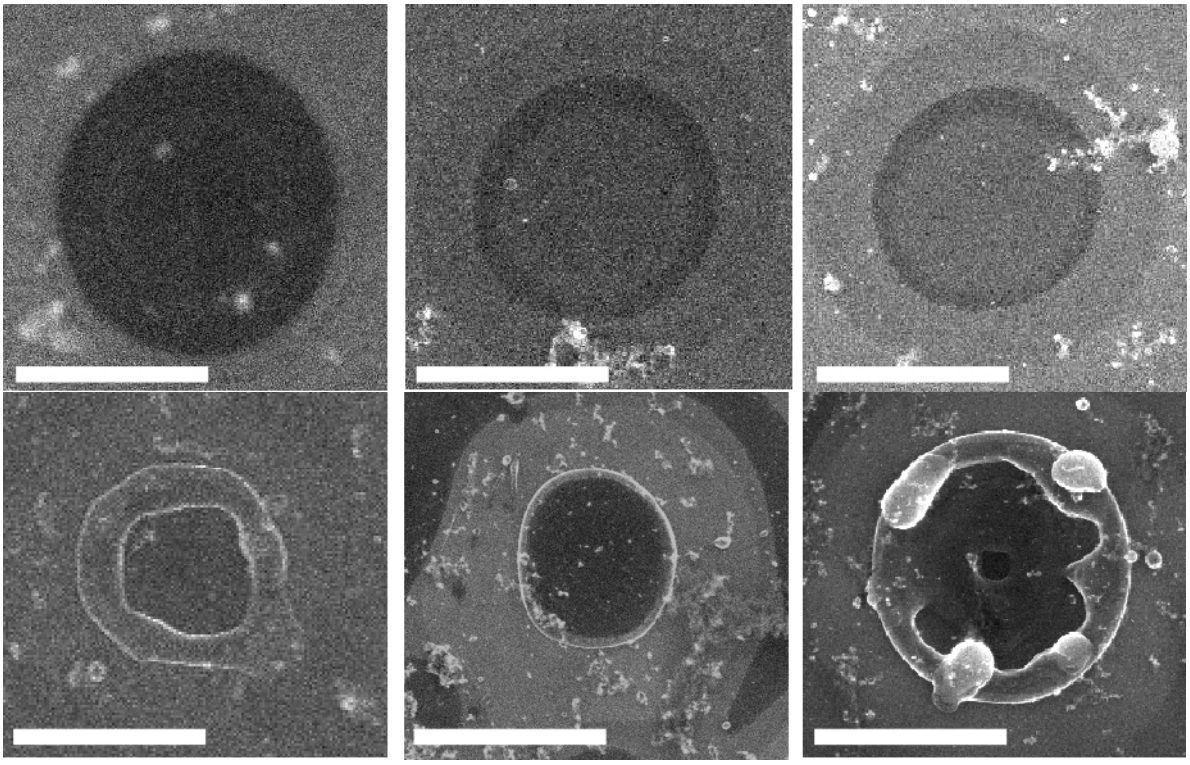


Figure 6. Micromachining results comparing TL (top) and chirped (bottom) single pulses impinging on a silicon wafer. The scale bar is 200 nm.

9. Nonlinear optical plasmonics and their control using shaped ultrashort pulses

Having developed all the tools necessary for selective nonlinear optical microscopy, we turned our attention to the study of multiphoton excitation of silver nanoparticles. Our first finding was that dendritic silver nanoparticles have a very large cross section for two-photon induced luminescence. When excited at 800 nm they exhibit strong two-photon induced luminescence near 550 nm. When imaging the dendritic silver nanoparticles in the microscope, we noticed that excitation at one location resulted in emission from multiple nanoparticles, some at distances up to 100um, or approximately 100 focal-spot diameters away.

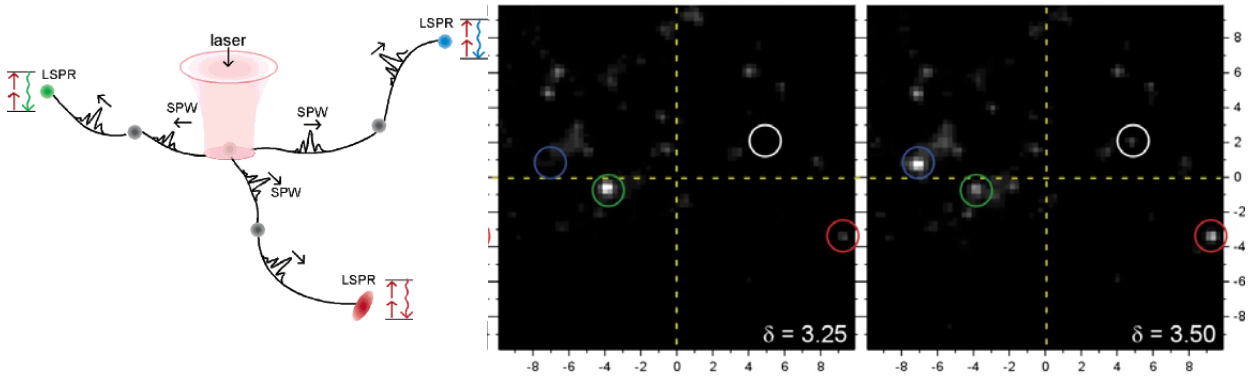


Figure 7. (left) Schematic drawing of the experiments. The laser is focused to a spot that is 0.4 micron in diameter. The two-photon induced emission is measured tens of microns away. (right) Experimental results showing the focal spot of the laser (center of dashed line crossing) and the emission from nanoparticles several microns away from the laser focus. The two panels correspond to two different phase functions which are causing different nanoparticles to light up.

The dendritic nanoparticles are interconnected during their random deposition on a quartz substrate during sample preparation. The remote emission results from surface plasmon waveguiding and subsequent localization at the end of the chain where localized surface plasmon resonance emission takes place. What is most intriguing is that the remote emissions can be controlled through the phase or polarization of the excitation beam. Control by phase shaping is shown in Figure 7. To be able to obtain these images we had to use MIIPS to compensate all phase distortions introduced by the 60X 1.45 NA objective, and we also had to attenuate the beam to 0.1 pJ/pulse to avoid damaging the nanoparticles. The positions of the sample and laser were fixed, with the laser focused to a spot of about 0.5 μm diameter at the center of the crosshairs. A sinusoidal phase function was introduced to shape the ~12 fs pulses. We see that different phases preferentially cause two-photon luminescence at locations far from the focal spot (colored circles). This phenomenon, its implications to plasmon waveguiding, and energy transfer between molecules at long distances (up to 100 um) will be studied in detail. By tracking the spectral phase dependence of the remote emitters, we are able to determine the dispersive properties of the nanowires that transmit the laser pulse energy. We have measured the dispersion of these plasmonic waveguides and in most cases they have negative dispersion. The implication of these measurements is that these nanowires can compensate for positive dispersion and serve as pulse compressors. We have recently demonstrated the propagation of 7 fs pulses in these plasmonic waveguides (manuscript in preparation). By following the interference between the pump and probe beams (both same wavelengths) we find that the further the particles the longer the interference observed. Typical values go from 7 fs near the focal spot to 100 fs for particles far from the focal spot. These measurements further indicate that propagation of the field in the nano plasmonic waveguides preserves the coherence of the electromagnetic waves. The pulse broadening appears to be caused primarily by dispersion in the plasmonic waveguides.

10. Controlling molecular fragmentation with intense near-IR femtosecond pulses

Our most recent project on the interaction of intense near IR shaped femtosecond pulses with gas phase molecules has led to very significant findings. Essentially, we found that under strong near-IR irradiation the fragmentation pattern changes by one or more orders of magnitude upon pulse shaping. However, these changes depended only on pulse duration and not on precise phase-amplitude properties of the field. This finding goes against numerous publications where it was claimed that control over fragmentation required special computer algorithms that tested thousands of different shaped pulses. The figure on the right shows changes in the relative yield of fragment ions as a function of the total yield of ions (a quantity proportional to pulse duration) for two very different types of shaped pulses. The lines correspond to positive chirp and the dots correspond to varying the period of a sinusoidal phase function. The resulting pulses for these two phase functions are completely different, yet the results are the same. These findings have been confirmed on 13 different molecules, so we can generalize. We also found that there is no condition under which acetophenone produces toluene (92 m/z), this calls into question the most important article on laser controlled chemistry. Our findings are now published in a feature article in the Journal of Physical Chemistry. We also found a fragmentation pattern involving double ionization that is highly sensitive to pulse shaping. This pathway is presently under investigation in our group and it is one of the directions we would like to pursue with further DOE funding.

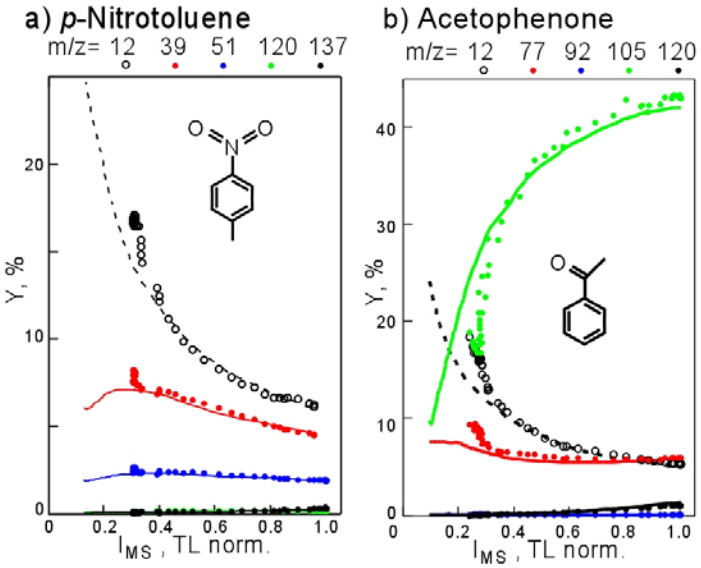


Figure 8, Experimental results on the controlled fragmentation of para-nitrotoluene (left) and acetophenone (right). The lines correspond to linear chirp, the dots correspond to experiments as a function of the period of a sinusoidal function.

Summary:

Our DOE supported research was highly innovative, in many cases revolutionizing how tasks like pulse characterization can be carried out with the highest accuracy. Our work has opened new venues of research such as nonlinear plasmonic control. Our research on coherent control is rooted in physics, and can be used to build applications because it is robust. This is in stark contrast to most of the work based on learning algorithms.

Scientists Supported:

High School Students: Sherdeep Singh and Andrew Gross
Undergraduate Students: Katherine A. Walowicz, Janelle Shane, Melinda Ewald
Graduate Students: Igor Pastirk, Matthew Comstock, Johanna Dela Cruz, Bingwei Xu, Jess Gunn, Michael Kangas, Xin Zhu, Yves Coello
Post Docs: Irene Grimberg, Vadim V. Lozovoy, Tissa Gunaratne

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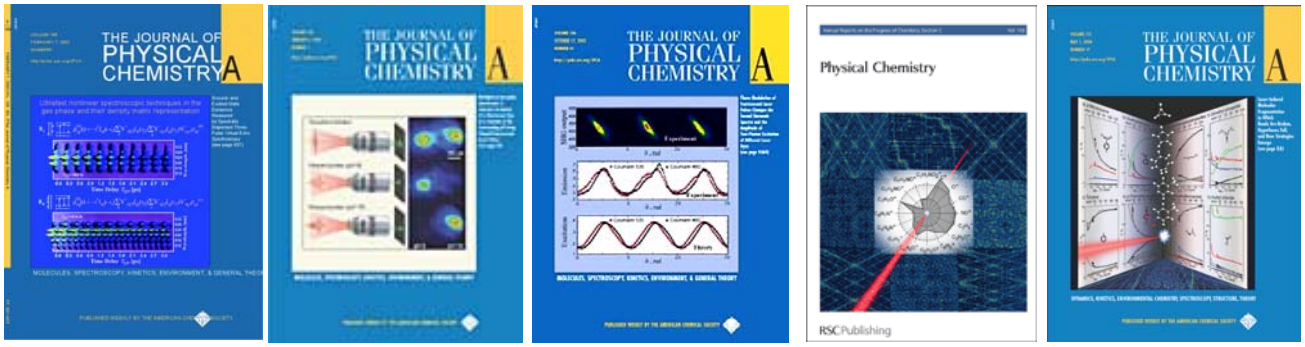

Publications Resulting from this Grant 2001-2008:

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3. I. Grimberg, V. V. Lozovoy, M. Dantus and S. Mukamel, "Femtosecond three pulse spectroscopies in the gas phase: density matrix representation," *J. Phys. Chem*. A, Feature 106, 5, 697 (2002). Our results featured in the cover.
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