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Applications of ultrashort shaped pulses in microscopy and for controlling chemical reactions

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Abstract

This article presents a new perspective on laser control based on insights into the effect of spectral phase on nonlinear optical processes. Gaining this understanding requires the systematic evaluation of the molecular response as a function of a series of pre-defined accurately shaped laser pulses. The effort required is rewarded with robust, highly reproducible, results. This approach is illustrated by results on selective two-photon excitation microscopy of biological samples, where higher signal and less photobleaching damage are achieved by accurate phase measurement and elimination of high-order phase distortions from the ultrashort laser pulses. A similar systematic approach applied to laser control of gas phase chemical reactions reveals surprising general trends. Molecular fragmentation pattern is found to be dependent on phase shaping. Differently shaped pulses with similar pulse duration have been found to produce similar fragmentation patterns. This implies that any single parameter that is proportional to the pulse duration, such as second harmonic generation intensity, allows us to predict the molecular fragmentation pattern within the experimental noise. This finding, is illustrated here for a series of isomers. Bond selectivity, coherent photochemistry and their applications are discussed in light of results from these systematic studies.

Keywords: Two-photon microscopy; Photo bleaching; Femtosecond laser; Mass spectrometry; Coherent control; Shaped pulses

1. Introduction

Using lasers to control matter has been in the minds of scientists since lasers were invented. Originally, it was thought that monochromatic coherent light would permit the direct cleavage of chemical bonds. This dream could not be realized because of ultrafast intramolecular vibrational energy redistribution (IVR). This prompted the quest for femtosecond laser pulse sources that could beat the timescale for IVR and permit selective bond cleavage [1]. The first such experiments proved that one could indeed interact with molecules while in their transition states [2]. In general, pump-probe experiments can be considered laser control experiments in which the probe causes a transition to a new potential energy surface from which different products are observed.

The molecular dynamics view of a localized wave packet that can be transferred to different potential energy surfaces using ultrashort pulses inspired the concepts of pump-dump and optimal control of chemical reactions [3,4]. Given that sufficiently accurate potential energy surfaces, ground and excited states, were not available, Judson and Rabitz proposed the use of a computer algorithm that would take advantage of the feedback gained from the laser-molecule interaction and thus learn to stir the reaction to a desired product target [5]. By 1998, stable laser sources and pulse shaping technology became available and the Gerber group demonstrated the first implementation of this technology to control the outcome of a laser-initiated chemical reaction involving a rather complex organometallic compound [6]. This landmark work inspired several groups to pursue laser control using

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learning algorithms [7]. The successful changes in the ratio of one product over a competing product suggested that bond selective laser control of chemistry was at hand. Understanding the success, especially when isolated molecules interacted with shaped near-IR pulses has remained nebulous.

In parallel to the learning algorithm work, pulse shaping experimental and theoretical research on the effect or phase shaping on nonlinear optical processes took place. This research led to the elegant demonstration of coherent control over two-photon transitions in atoms [8] and coherent anti-stokes Raman scattering (CARS) [9]. The application of these concepts to control molecular systems in excited electronic states required ultrashort shaped pulses, this in turn required accurate spectral phase measurement and elimination of phase distortions.

Interestingly, phase measurement, generation of ultrashort accurately shaped pulses, and their application to controlling nonlinear optical processes all occurred at once in an experiment carried out by the Dantus research group [10–13]. The experiment used spectral phase to control the resulting second harmonic generation (SHG) spectrum and two-photon excitation of large organic molecules in solution. Constructive or destructive interference was found to occur as predicted by a simple formula that could be used to predict the outcome of two- and three-photon processes. A number of reviews have been written about this process dubbed multiphoton intrapulse interference (MII) and its use for controlling nonlinear optical process [13–15]. This process, because it is intrinsic to the electric field of the pulse, is essentially medium-independent. The nonlinear optical (NLO) response to phase shaped pulses is accurately determined by MII, within a perturbative multiphoton description. This deterministic quality of MII implies that the NLO response could be used to measure the spectral phase of pulses. The method, known as MII phase scan (MIIPS), measures the NLO response for a number of reference phase functions and determines the spectral phase of the pulses. If desired, the spectral phase can be eliminated to leave transform limited pulses [16–18]. Being a single beam method, MIIPS can provide accurate phase measurement at the sample location, at the focus of a microscope [17] or a telescope[19].

Building on the successes of MII to control NLO processes, we attempted to extend its use to laser control of chemical reactions. The initial work was based on binary phase shaped pulses which were shown by us to be the most efficient means to achieve MII [20]. Our laser control of chemical reactions based on binary phase pulses showed that large changes could be found in the fragmentation patterns of isolated polyatomic molecules [21]. However, close scrutiny of three dimensional maps obtained after exhaustive evaluation of binary phase functions revealed that maximum changes were found when comparing transform limited (TL) pulses and highly modulated pulses [22]. No evidence was found for a multiphotonic resonance. In this article we present a brief account of a comprehensive study recently completed where we found that when isolated molecules are irradiated by intense near-IR lasers, the duration of the laser-molecule interaction determines the resulting fragmentation pattern [23]. Within the interaction time, the precise time–frequency structure of the pulse was found to have little or no effect. For example, a large number of very differently shaped pulses were found to produce identical fragmentation patterns. This observation contradicts the expected dependence on phase and amplitude properties of the pulse based on pump-dump and optimal control theories. We are beginning to develop a model that explains this unexpected behavior. In the mean time, we have shown that order-of-magnitude control over molecular fragmentation can be achieved by a number of single parameter approaches, for example linear chirp. The rather simple dependence on pulse duration makes it possible to predict how a molecule will fragment when irradiated with an arbitrarily shaped laser pulse. The fact that the fragmentation pattern depends on molecular structure makes us confident that pulse shaping adds a new dimension to mass spectrometry, for example for isomeric identification [24,25]. Intrigued that we failed to find the expected phase sensitivity that has been anticipated by most scientists in the field, we searched further and found a single coherent photodissociation pathways, discussed at the end of the article.

2. Experimental

In order to study the effects of pulse shaping it is extremely important to be able to synthesize pulses that are completely free of phase distortions. As mentioned earlier, we use MIIPS to accomplish phase measurement and compensation of phase distortions. MIIPS is presently the most accurate means for measuring and eliminating the chromatic dispersion introduced by optical elements such as quartz, glass, and more recently air and water [16,17,26].

The experiments presented here were obtained using two different femtosecond laser systems. For biological imaging, we used an oscillator producing pulses with a 110 nm FWHM bandwidth centered at 800 nm. The pulse shaper uses a 640 pixel phase only spatial light modulator (SLM). The beam enters an inverted microscope and is focused by a high numerical aperture objective on the sample. A pair of prisms is used to reduce the burden of removing linear chirp. Because a prism pair can only compensate for group delay dispersion (GDD), we refer to this setup as GDD-only compensation. High-order phase distortions (HOD) introduced by all the optics in the beam path are measured and eliminated using MIIPS based on SHG signal that is collected from a thin KDP crystal placed at the focus of the microscope objective. For imaging we used a thick mouse kidney tissue section (over 100 μm) stained with DAPI (cell nuclei), Mitotracker-488 (mitochondria), and Phalloidin-568 (actin). A series of up to 50 optical sections (section distance of 1 μm each) were recorded (512×512 pixels per image, scanning speed 1 sec per

image, 8 images were averaged for each depth), in a sequential mode. For selective imaging, mouse embryo fibroblast (MEFs) cells were co-stained with phalloidin-conjugated to alexa fluor 350 dye (Invitrogene). Mitochondria were labeled with a primary biotin antibody using a FITC-conjugated goat anti-mouse secondary (Jackson Immunoresearch). The objective used was Nikon 60×1.45 NA, with working distance of 130 μm.

For the experiments on laser control of chemical reactions, the broad bandwidth output of the Ti:Sapphire oscillator goes through a pulse shaper with a 128 pixel phase only pulse shaper and then enters a regenerative amplifier producing 35 fs TL pulses with 0.7 mJ per pulse. For these experiments, we eliminate phase distortions that accumulate in the amplifier and those that are introduced by all other optical elements using MIIPS at the focus of the beam inside the mass spectrometer sample chamber or at an equivalent point that is configured to mimic the inside of the chamber (identical focusing optic). This ensures that we know precisely what phase is delivered to the sample. The experiments presented here were obtained with a 0.15 mJ/per pulse energy, the peak power density at the focus is estimated at 10^{15} W/cm^2 when the pulses are TL. Additional information about the samples, and the mass spectrometer is given elsewhere [23].

3. Results and discussion

3.1. Shaped pulses for improved two-photon fluorescence microscopy

In 1990, Denck and Webb showed that two-photon excitation could replace the conventional confocal laser microscopy [27]. The use of femtosecond lasers for two-photon microscopy was a natural consequence of the inverse relation between pulse duration and two-photon excitation efficiency. Note that despite this relation, the pulses used for microscopy are typically 100–150 fs, and several articles in the literature conclude that shorter pulses fail to deliver greater signal and that shorter pulses induced greater damage [28]. This paradox between the theoretical prediction and experimental findings motivated a study in which we measured the signal from our microscope under two different conditions GDD corrected pulses, and pulses corrected for GDD and HOD using MIIPS. The first condition mimics the majority of microscopes presently in use, in particular those that include a prism pair to correct the GDD introduced by the microscope objective. We found that the GDD only compensated measurements agree with the literature in that no signal gains were obtained by using shorter pulses. However, the TL pulses for which MIIPS was used to remove HOD's achieve the expected linear increase in signal as a function of spectral bandwidth [29]. The increased signal has been demonstrated under imaging conditions [30].

We illustrate the importance of measuring and correcting phase distortions during two-photon imaging of biological samples with ultrashort pulses in Fig. 1, where increased penetration depth is observed when higher-order dispersion is compensated for, compared to when only GDD is compensated for. The images show proximal kidney tubule cells with a typical apical brush border (distinct high intensity inner area of the cells). These structures can be seen clearly when applying MIIPS compared to GDD-only compensation. The images shown for GDD-only compensation have been intensified by a factor of two, otherwise they were too dark to show any structure. Image quality was assessed by calculating signal-to-noise ratio similar to Xi et al. [29]. We found that using TL pulses allows us to image a depth range that is three times greater than GDD-only compensation.

Given the signal increase realized with TL pulses, one can then reduce the intensity of the laser and thus reduce photon induced damage, while maintaining a constant signal to noise ratio. Laser intensity for both types of laser pulses was adjusted until TL and GDD only corrected pulses produced exactly the same signal. The factor of ~2 attenuation of the TL pulses resulted in decreased photobleaching of the sample. TL pulses were found to cause less photobleaching [29]. We attribute this difference to the different laser intensity used for TL pulses. This observation

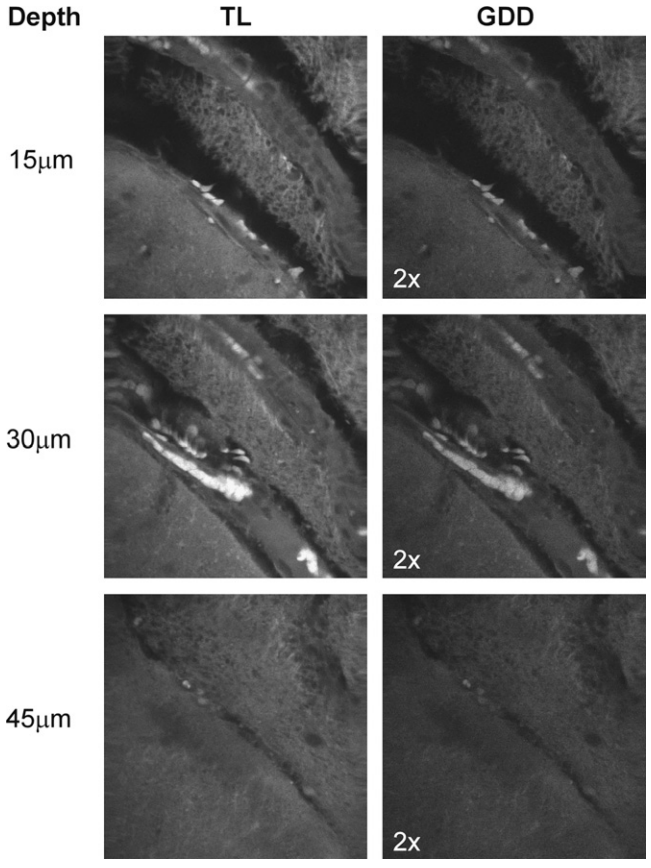

Fig. 1. Cross section of mouse brain labeled with MitoTracker 488, Phalloidin 568, obtained at different depths, shown for TL pulse (left) and GDD compensated pulses (right). The brighter features are red blood cells inside blood vessels. The signal intensity for GDD only compensation was increased by a factor of two. Image size is 100×100 μm.

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suggests that photobleaching is caused by absorption of one or more photons by the excited state.

Ultrabroad bandwidth is one more advantage to using ultrashort pulses for biomedical imaging because pulses can be easily tailored to preferentially excite different chromophores. Selective excitation is presently accomplished with tunable lasers; however, as the laser is tuned there is a risk that images obtained will not register, a condition also known as 'pixel shift'. This issue is important because one of the critical aspects of imaging is collocation of subcellular structures. If image registration cannot be ascertained, collocation cannot be determined with certainty. With ultrashort pulses phase shaping can be used to achieve selective excitation, taking advantage of MII [10,12]. We have shown that selective excitation is possible even when exciting fluorescent chromophores through a millimeter of scattering biological tissue [31].

Selective excitation is illustrated in Fig. 2, on a sample of mouse embryo fibroblast cells. The cells were co-stained with Phalloidin-350 (actin filaments), and with biotin antibody followed by FITC secondary (mitochondria). Spectral amplitude shaping at the pulse shaper was used to get selective excitation of the sample dyes. The pulses used were obtained from the higher (blue) or the lower (red) spectral frequency regions of the pulses. Control experiments with single dye staining were employed to ensure selective (exclusive) excitation $> 95\%$ for each dye by the blue or red pulses (data not shown). In Fig. 2 actin filaments were visualized using the blue pulse (left image), while the mitochondria was imaged by the red pulse (middle image). The merged image (right image) clearly demonstrates typical sub-cellular localization of each structure.

3.2. Control of chemical reactions

Control of chemical reactions, and in particular fragmentation of isolated molecules with shaped near-IR pulses, is not as simple to understand as experiments involving resonance excitation. When the laser pulses are resonant with electronic transitions, phase and amplitude changes can be directly related to the phase and amplitude of the resulting quantum rovibronic states, as nicely demonstrated by Leone and coworkers [32,33]. The time evolution of this type of coherent superposition is fully understood. The case discussed here is quite different because it involves molecules without one or two-photon resonances with the carrier frequency of the pulses. The experiment was carried out in a mass spectrometer; therefore, only ions directly generated by the single shaped pulse were detected. We picked this set of conditions inspired by pioneering experiments using learning algorithms to control chemical reactions [6,34]. The goal of our study was to determine if certain types of phase shaping functions could be used to reveal general trends in laser control of chemical reactions.

This research project led us to evaluate tens of thousands of phases on a total of sixteen different molecules. Instead of using a learning algorithm we used exhaustive evaluation of different sets of phases inspired by a number of different physical models. For example, we used chirp because it has been used for inducing rapid adiabatic passage [35], and for wavepacket following [36]. We used sinusoidal functions with different periods to create trains of pulses with different temporal spacing, these were inspired by pump-probe type of experiments. It was expected that a certain interpulse duration would cause a significant change in the fragmentation pattern, similar to what would be expected if this were a pump-dump or pump-probe experiment. In addition to these strategies we used several others, such as random phases, random binary phases, and a series of phases that represented polynomial functions that can be described with cubic, quartic and quintic frequency dependences. We also tested combinations of phase and amplitude shaping. A full account of these results is published elsewhere [23]. Here we focus on representative results obtained for three molecular isomers.

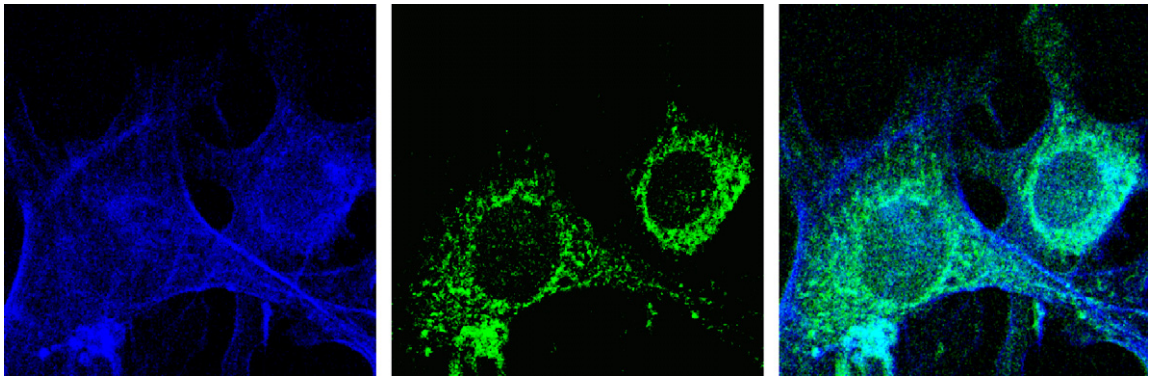

Fig. 2. Selective two-photon imaging of mouse embryo fibroblast (MEFs) cells with Phalloidin-350 (actin filaments), and with biotin antibody followed by FITC secondary (mitochondria). The cells were imaged with shaped pulses that favored two-photon excitation at bluer or redder frequencies. The images (100×100 μm) resulting from these measurements were false colored accordingly. The figure on the right merges results from the two previous images. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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For our experiments, rather than comparing the ratio between two photoproducts, we decided to plot the observed fragmentation trends as a function of the total ion yield (I_{MS}) for each of the different pulse shaping functions. In Fig. 3 we show results for *ortho*-, *meta*-, and *para*-nitrotoluene as a function of pulse shaping. The lines correspond to results obtained as a function of positive chirp, and the dots correspond to measurements obtained as a function of changing the period of a sinusoidal function. When represented as a Wigner function, these two methods for shaping the pulses are absolutely different. The sinusoidal pulses have multiple oscillations with regions of positive and negative interference, corresponding to trains of femtosecond pulses, in complete contrast to the linear frequency sweep of the chirped pulses. Despite these differences, when the percent yield for each fragment ion is plotted as a function of I_{MS}, a striking and surprising coincidence is revealed. The relative yield of every fragment as a function of I_{MS} coincides for chirp and for sinusoidal phase modulation. This finding has not been confirmed for more than 20 different molecules by our group [29].

In Fig. 3a we show the mass spectrum of *ortho*-nitrotoluene obtained using TL pulses, together with an assignment of the major ion products and their most probable structures. Comparing the results from Fig. 3b–d, we find that the fragmentation patterns are sensitive to molecular geometry, making femtosecond laser ionization useful for isomer identification [24,25]. Here, we see that the differences are not caused by a very specific shaped pulse but by the extent that the fragmentation pattern changes. For *ortho*-nitrotoluene, the product with *m*/*z* 120 results from the reaction between the methyl group and the nitro group. This intramolecular hydrogen transfer reaction leads to loss of OH and is favored for TL pulses. Linear chirp can reduce the yield by up to two orders of magnitude. The ortho hydrogen transfer reaction is favored by ionic states, this is why using TL pulses optimizes this reaction pathway [37]. As chirp is increased, the pulse lengthens and multiphoton excitation of intermediate electronic states becomes the dominant ionization–fragmentation process. Differences among the other isomers are more subtle. For example, the tropylium ion at *m*/*z* 91 is a major product for *meta*- and *para*-nitrotoluene, only *ortho*-nitrotoluene produces the tolyl ion at *m*/*z* 92 with significant abundance. The molecular ion for *meta*-nitrotoluene has a greater relative yield. The phenylium ion at *m*/*z* 77 is more likely to be produced from *para*-nitrotoluene than from the other two isomers.

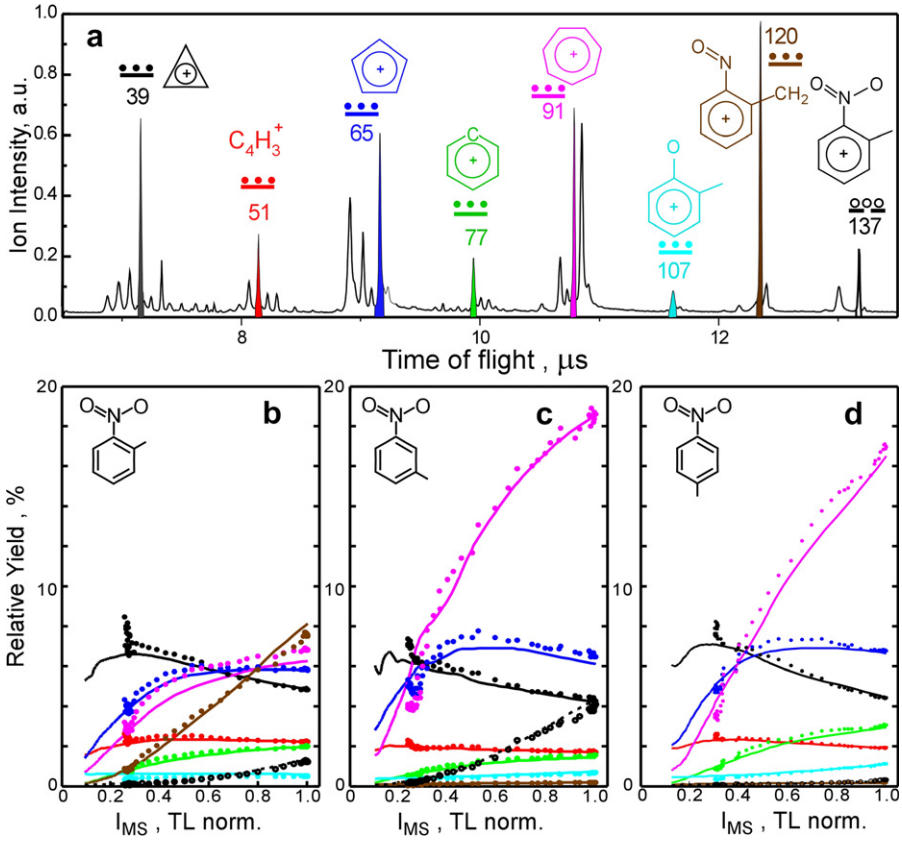

Fig. 3. Experimental results on the laser controlled fragmentation of nitrotoluene isomers. (a) The mass spectrum of *ortho*-nitrotoluene is shown together with the structure of the most common product ions. (b, c, d) Relative yield of the most common product ions for *ortho*-, *meta*- and *para*-nitrotoluene. Results for linear chirp (lines) and changing the period of a sinusoidal function (dots) are compared as a function of I_{MS}. Notice that the changes in the fragmentation pattern coincide for the two very different pulse shaping strategies (lines vs. dots). Changes in the fragment yields as a function of pulse shaping provides a clear distinction between the three isomers.

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The detailed ion chemistry involved in these experiments has not been fully unraveled. The main observation being made here is that two very different pulse shaping strategies (chirp and a sine function) were used and the results are remarkably similar. There is no evidence that the sinusoidal phase function, leading to trains of pulses was successful in influencing any of the different product channels beyond the signal-to-noise levels in this experiment. Note that for sinusoidal functions, our pulse shaper could not deliver phase function that would yield I_{MS} values below 0.3. This is because of a finite resolution. The extent of change induced by pulse shaping can be quantitatively expressed as a variance. We have dimension we have proposed elsewhere the use of variance as a robust dimension for mass spectrometric analytical applications [38].

The results shown in Fig. 3 show a significant degree of control as a function of pulse shaping. The degree of observed control could be increased much further with simple chirp. Equally significant is what we do not find in the data. There appears to be no difference attributable to the very different methods used to shape the laser pulses. Coherent vibrational dynamics would have favored pulse sequences that matched the molecular dynamics, but this was not observed. Our present explanation is that under off-resonance strong field excitation, the molecule is field ionized within the first few femtoseconds, probably within the rise of the pulse. The electromagnetic field causes a coherent superposition of electronic states. As the complex wave packet evolves different components reach an outer turning point from which additional photons are absorbed causing further dissociation. Strong field off-resonance excitation populates vibronic states that are as much as tens of thousands of wave numbers apart. The ensuing photofragmentation can be expected to be essentially insensitive to coherent effects due to the very large energy spread of states excited by the pulse. The rate of wave packet dephasing increases linearly with the range of energies it encompasses and the rate of vibrational redistribution increases linearly with the number density of states within the energy range of excitation. In other words, ultrafast IVR rates are expected to follow strong field non-resonant excitation. This behavior is very different when the initial excitation creates a localized wave packet in a well defined electronic state. In such cases, when the energy bandwidth of the coherent superposition is 10–500 cm^{-1} (instead of thousands of wave numbers) clear wave packet evolution can be observed [39–41].

The apparent lack of coherence found in our data, inspired us to search further for fragmentation pathways that were sensitive to the temporal symmetry of the shaped pulses. Such sensitivity would indicate wave packet evolution [42]. We first searched for a fragment yield that would not be symmetric with respect to the sign of chirp. We found in a number of molecules, that fragments resulting from multiply charged precursors with yields of 1–10%, were highly sensitive to chirp sign and magnitude, and show interesting dependence as a function of phase function used [23]. We have clear evidence that this is a coherent process that is highly sensitive to pulse shaping. The results for acetophenone appear in Fig. 4 where we show the dependence of ratio $[CH_3^+]/[C_6H_6CO^+]$ (15 m/z–105 m/z ratio) as a function of chirp (Fig. 4a) or pulse pair separation controlled by scanning the slope of two linear functions that delay the blue or red frequencies of the pulse (Fig. 4b) [42]. Notice that the relative yield of CH_3^+ increases significantly for $\beta = -7800\ fs^2$ and for a pulse separation of 300 fs. There is no corresponding enhancement of the relative yield for positive values. A full account of these coherent fragmentation pathways, occurring above 10^{15} W/cm^2, will be published elsewhere.

4. Conclusions

As the field of laser control matures, a number of important achievements are becoming clear. Most notably, there are already two commercial applications being used in laboratories around the world based on coherent control; these are the interference between ω and 2ω used for phase carrier stabilization [43] and MIIPS [17]. Pulse shaping is

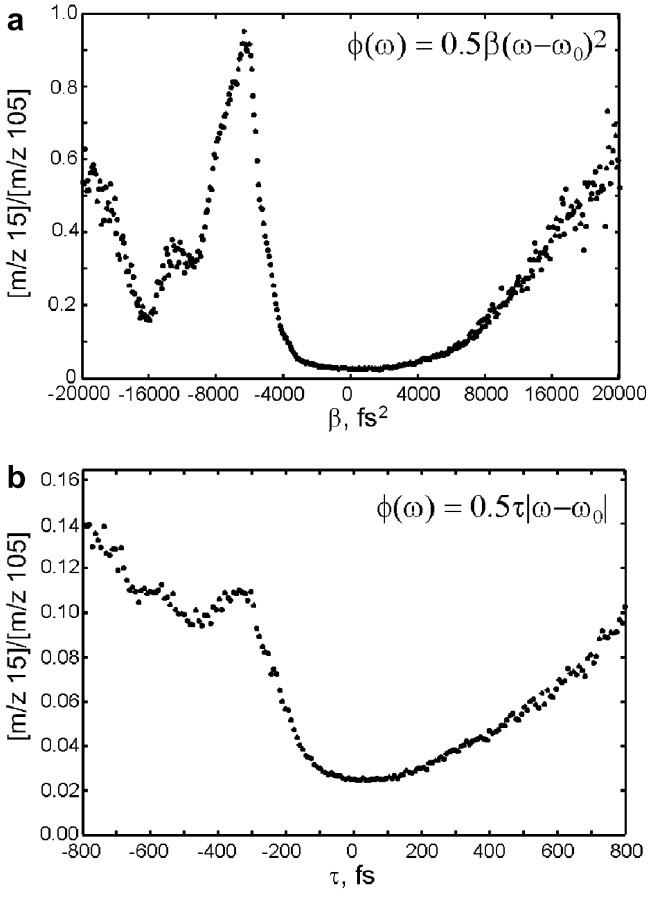

Fig. 4. Dependence of the relative [m/z 15]/[m/z 105] yields for fragmentation of acetophenone as a function of linear chirp (a) and as a function of the time delay between two sub-pulses (b). Notice the asymmetric dependence on pulse shaping, a signature of coherent wave packet evolution.

now used for improving NLO imaging based on two-photon microscopy and CARS imaging [9,44–46]. Similar methods are being developed in our group for remote CARS detection.

The lack of dependence on the pulse shaping strategy used for near-IR laser control of photofragmentation, implies that robust applications can be readily developed. Single parameter control implies that an instrument can be 'trained' to search for a molecule-specific fragmentation pattern in minutes. Our group is working on these types of analytical applications for biomedical and homeland security applications.

In our opinion, the field of laser control is about to experience tremendous growth. A wave that will follow the entrance of femtosecond lasers to industrial micromachining, surgical procedures and metrology. Applications will depend on robust, and reproducible results. This is because applications require a number of systems to always obtain a given response for a pre-determined shaped pulse.

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