2D (time-frequency) femtosecond four-wave mixing at 10^{14} W/cm^2: Molecular and electronic response

Igor Pastirk, Matthew Comstock, Katherine A. Walowicz, Vadim V. Lozovoy and Marcos Dantus

Department of Chemistry and Center for Fundamental Materials Research, Michigan State University, East Lansing MI 48824,
E-mail: dantus@msu.edu

Abstract: The response of atoms and molecules to intense 10^{13} – 10^{15} W/cm^2 lasers is investigated using two-dimensional (time-frequency) four-wave mixing. The data reveal a transition from a molecular response to a purely electronic (plasma) response.

1. Introduction

High-intensity laser fields have been used to generate high harmonics, to cause above threshold ionization and to generate plasmas. In most studies, only the high order outcome is detected, here, we use a four-wave mixing (FWM) method to probe the sample after high-intensity laser interaction. For our measurements, we use two high-intensity laser pulses followed by a weak probe pulse. The signal corresponds to the diffraction of the probe laser from the transient grating formed by the two intense pulses at the sample [1]. Spectral dispersion of the signal reveals changes in frequency as a function of time delay. For low intensity excitation, rotational dephasing can be observed, however, for high-energy excitation, the molecular response disappears and diffraction like response is observed. Our observations at high intensity are consistent with self-phase modulation occurring in a plasma filament.

2. Experimental

The laser source was a Kapteyn-Murnane oscillator amplified by an Evolution X-pumped Spitfire producing 850 ?J per pulse at 1 kHz, centered at 807 nm. The near transform limited pulses were 50 fs in duration. The experiments were carried out by splitting the output from the amplifier into three laser beams that are combined at the sample cell in the forward-box geometry. The first two laser pulses where overlapped in time while the third pulse was delayed by a computer controlled actuator. The energy of the first two laser pulses was varied from 10 to 200 ?J per pulse. The third pulse was maintained at 10 ?J per pulse. The three pulses were focused onto a 30 ?m diameter. The signal was collected background-free at the phase matching geometry. A spectrometer was used to spectrally disperse the signal and a nitrogen cooled CCD was used for dispersed spectra detection. The 6" long quartz cell contained the sample gas O_2 (500 Torr), CS_2 (100 Torr) or Ar (760 Torr).

3. Results

For low intensity excitation, the early time response of the molecular samples showed a time-zero feature corresponding to the instantaneous polarizability of the medium. This feature appears vertical in the wavelength-time plots, indicating the overall laser chirp to be minimal. The data shown in Fig. 1, correspond to excitation intensities below the threshold for filament formation. Both O_2 and CS_2 show time delayed 'wings' that extend to several hundreds of femtoseconds. These features correspond to the O (?*J*=-2, blue shifted) and S (?*J*=+2, red shifted) branches and are caused by multiple rotational Raman transitions. Notice that for CS_2 the dephasing is much slower because of its much smaller rotational constant.

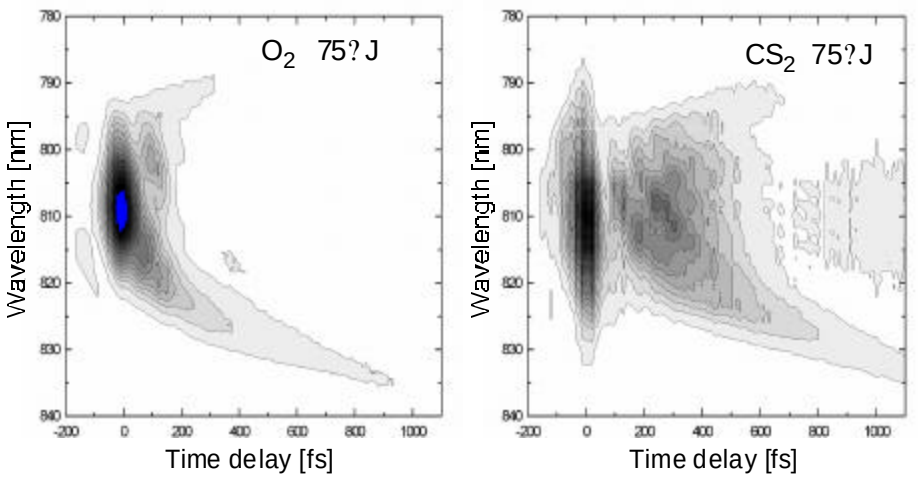

Figure 1. Experimental 2-dimensional data obtained at intermediate intensity

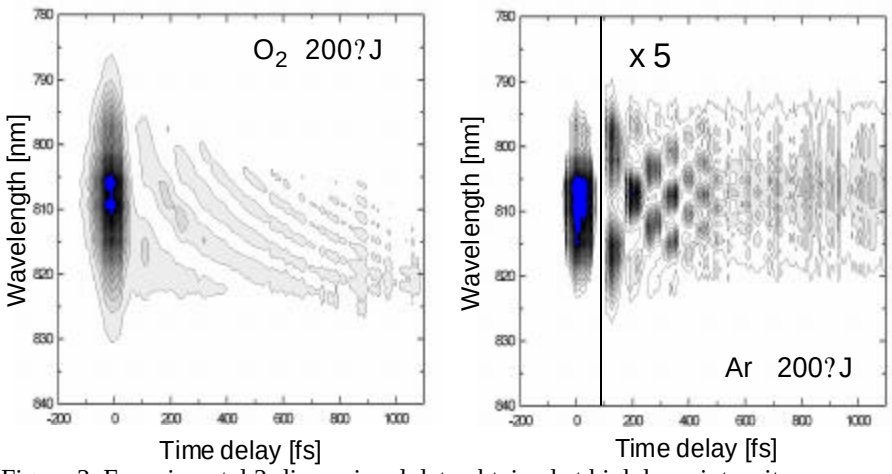

Figure 2. Experimental 2-dimensional data obtained at high laser intensity

When the laser intensity is increased above 10^{14} W/cm^2, a filament is visible in the cell and the signal no longer shows the molecular signature of rotational dephasing. Instead, a series of recurring features are observed. In Fig. 2a, we show data for O_2 obtained at high laser intensity. Notice that the blue and red wings seen in Fig 1a, are replaced by a series of parabolic red-shifting features. These features are caused by plasma formation in the sample. We replaced the molecular sample by argon and found that at lower laser intensities, the signal showed the instrument's autocorrelation. However, at high intensity, the signal (shown in Fig. 2b) shows a time zero feature, and a series of diffraction-like features indicative of plasma induced self-phase modulation. Damping of the plasma oscillation leads to a diminishing red/blue shift of the features. The Ar data is more symmetric than that of O_2 perhaps because of the Coulomb explosion that follows high intensity excitation of a molecular sample.

4. Discussion

At high intensities, the observed oscillation period seems to be independent of sample (O_2, N_2, CS_2 and Ar) and pressure (in the 100 to 760 Torr range). The period starts at ~ 90 fs and decreases to ~ 50 fs with a damping time of ~ 0.5 ps. The spectrum reveals two types of shifts; odd oscillations have a node at the carrier frequency while even oscillations do not. Temporal oscillations in the FWM signal under femtosecond plasmas excitation were predicted in the theoretical work of Vu *et al* [2], The oscillation frequency is proportional to the plasma frequency. From this we estimate a plasma density of ~10^{18} cm^{-3}. The second possible explanation for the observed oscillations in the FWM signal is related to a cyclotron motion of the plasma electrons caused by a self induced magnetic field. The estimated magnetic flux density is larger than 10 T [3]. Our data is consistent with their time-resolved simulation, unfortunately spectral changes have not been addressed. We find that the additional information obtained from two-dimensional FWM is important to observe the transition between a molecular response to an electronic response as a function of laser intensity. We have explored the effect of laser chirp. These results will be published elsewhere.

Acknowledgements: This work was supported by the National Science Foundation Grant CHE-9812584. Additional funding comes from a Packard Science and Engineering Fellowship, an Alfred P. Sloan Research Fellowship and a Camille Dreyfus Teacher-Scholar award. KW thanks the REU program at MSU.

References

1 E. J. Brown, Q. Zhang, and M. Dantus, J. Chem. Phys. **110**, 5772, 1999.

2 Q. T. Vu, L. Banayi, P. I. Tamborenea and H. Haug, Europhys. Lett. **40**, 323, 1997.

3 M.W. Wu and H. Haug, Phys. Rev. B **58**, 13060, 1998.