Generation of ultrafast continuum mid-infrared laser pulse

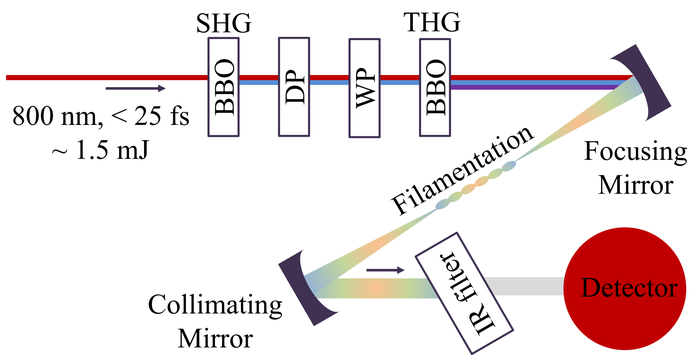
Chai Ho Lim[1,2], Kwanghee Park[1,2], Youngseok Chae[1,2], Kyungwon Kwak[1,2]*, Minhaeng Cho[1,2]*

[1] Center of Molecular Spectroscopy and Dynamics, Institute for Basic Science(IBS)
[2] Department of Chemistry, Korea University

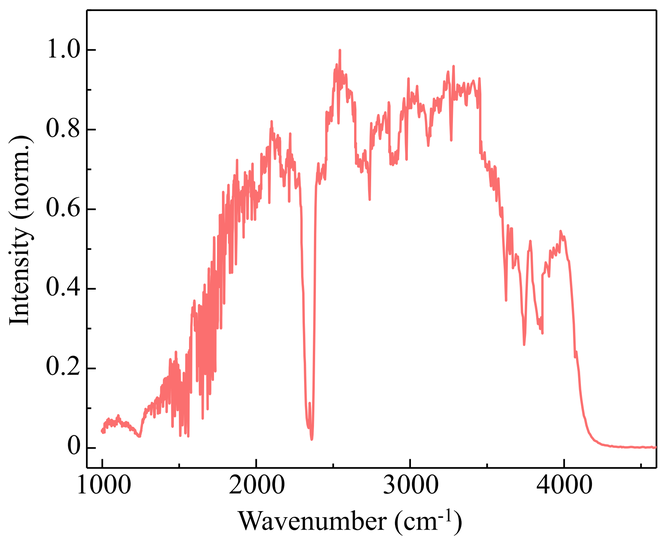
Introduction

INTRODUCTION

Ultrafast infrared spectroscopy performs study of various chemical reactions and molecular dynamics that occur at the femtosecond level. Most of the mid-IR pulse used in the ultrafast infrared spectroscopy have been generated by signal and idler from OPA through difference frequency generation. The limitation of mid-IR pulses generated by OPA-dfg is that the bandwidth is relatively narrow to observe entire molecular vibration region. Recently, Tokmakoff's group and several groups have generated a continuum IR pulse via two or three color pulse mixing in plasma that can cover the most of molecular vibration region (1000-4000 cm-1) and succeeded in applying it as a probe pulse in 2D-IR spectroscopy. We have also succeeded in producing continuum IR in the same way and we plan to apply it to ultrafast IR spectroscopy.



Generation of continuum IR source starts from doubling(400 nm) and tripling(266 nm) of fundamental source(800 nm, 25 fs, ~1.5W). Fundamental source with second harmonic and third harmonic are focused in the air. When the air is in the intense electric field which is in this case laser pulse, air break down into plasma. In the air-plasma, mixing of three UV-vis pulse generates plasma induced continuum IR pulse.



Theoretical Explanation

TRANSIENT ELECTRON CURRENT MODEL

Phenomenon of continuum IR generation can be explained by transient electron current model. It explains the generation via directional current of the free electron in the plasma. Plasma under electric field of fundamental, second harmonic and third harmonic laser field can be expressed as,

$$E_L(t) = E_\omega \cos\omega t + E_{2\omega}\cos(2\omega t + \theta_{2\omega}) + E_{3\omega}cos(3\omega t + \theta_{3\omega})$$

Once ionized at $t = t_i$, the free electrons in subject to the external laser field accelerate at a velocity of

$$v(t) = \int a dt = -\frac{e}{m_e}\int_{t_i}^{t} E_L(t)dt$$

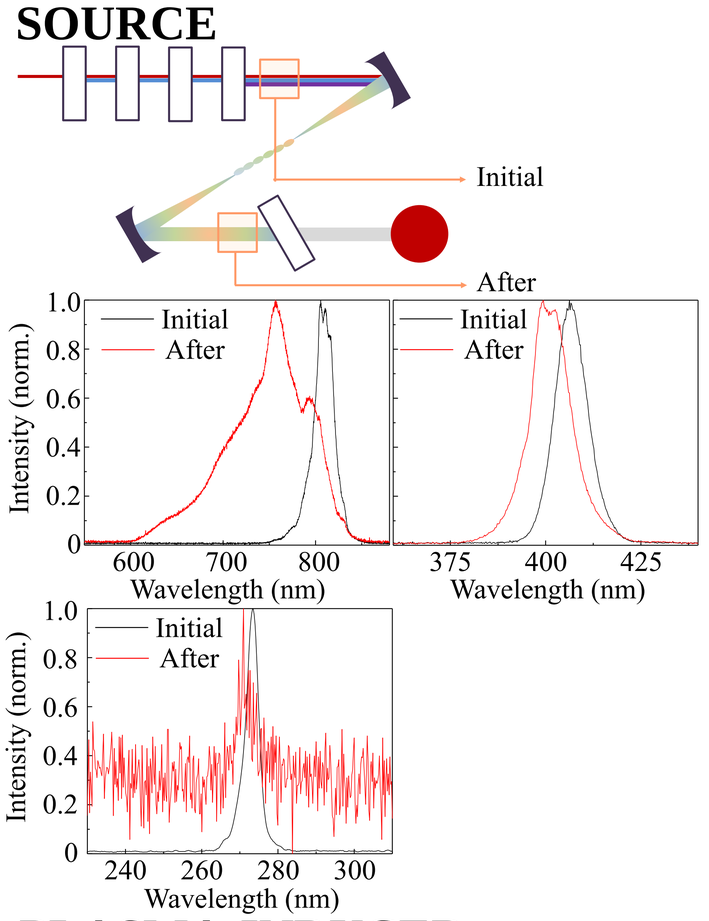
Electron current density can be computed as

$$J(t) = -\int ev(t,t')dN_e(t')$$

where $dN_e(t')$ is the change in electron density in the interval between $t'$and $t' + dt'$. This current surge gives rise to radiation, whose electrical field is proportional to $dJ(t)/dt$. Finally, the radiation spectrum can be obtained from a Fourier transform of $dJ(t)/dt$.

Plasma Induced Spectral Blue Shift

SPECTURUM OF INPUT SOURCE



PLASMA-INDUCED SPECTRAL BLUESHIFT

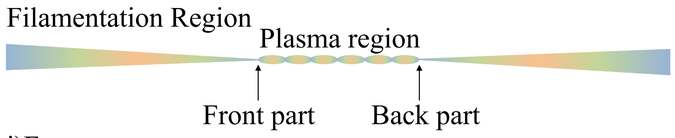
During propagation in air-plasma medium, ultrafast laser pulse experiences varying of refractive index in medium. Variation in refractive index induces a phase shift in the pulse, leading to a change of the pulse's frequency spectrum which is called self-phase modulation(SPM). Assuming the wave is a plane wave, wavefunction and frequency shift is given by,

$$F(z,t) = \exp\left\{i\left[\left(\omega_0 t - \frac{\omega_0 n_0}{c}z\right) + \int_0^t \Delta\omega dt\right]\right\}$$

$$\Delta\omega = \frac{\partial}{\partial t}\left(-\frac{\omega_0 \Delta n(t)}{c}z\right) = -\frac{\omega_0}{c}z\frac{\partial[\Delta n(t)]}{\partial t}$$

where $\Delta n(t)$ is the variation of refractive index, c is the light speed and ω_0 is the initial frequency.

Filamentation Region



i)Front part

Neutral gas dominates, neglecting the plasma contribution,

$$\Delta\omega = -\frac{\omega_0 z}{c}\frac{\partial[\Delta n(t)]}{\partial t} = -\frac{\omega_0 z}{c}n_2\frac{\partial I(\text{front part})}{\partial t} < 0$$

Since pulse is focusing, the pulse has a positive temporal slope. Hence front part of the pulse contributes to red shift broadening.

ii)Plasma

Frequency shift broadening due to the plasma term is

$$\Delta\omega = +\frac{2\pi z e^2}{c m_e \omega_0}\frac{\partial N_e}{\partial t}$$

The rate of change of the electron density is proportional to N_0, the density of the neutral air and to I^m.

$$\frac{\partial N_e(t)}{\partial t} \propto N_0 I^m$$

Substituting to frequency shift broadening due to the plasma term, we obtain

$$\Delta\omega = +\frac{2\pi z e^2 N_0 w}{c m_e \omega_0} I^m(t)$$

Showing blue shift broadening of the frequency and it is large partially because of the nonlinear dependence on the high intensity inside the self-focal region.

iii)Back Part

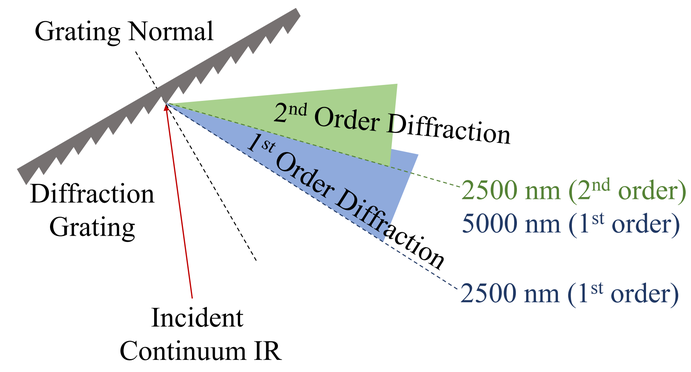
$$\Delta\omega = -\frac{\omega_0 z}{c}n_2\frac{\partial I(\text{very steep back part with negative slope})}{\partial t}$$

It shows blue shift broadening by same analogy of front part.

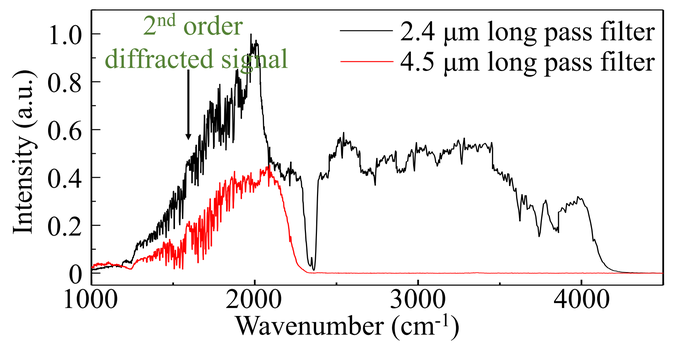
Plasma-induced spectral blue shift makes it possible to mix nonharmonic three-color pulse, resulting continuum IR. Without it, it will generate terahertz pulse.

Second Order Effect From Monochromator

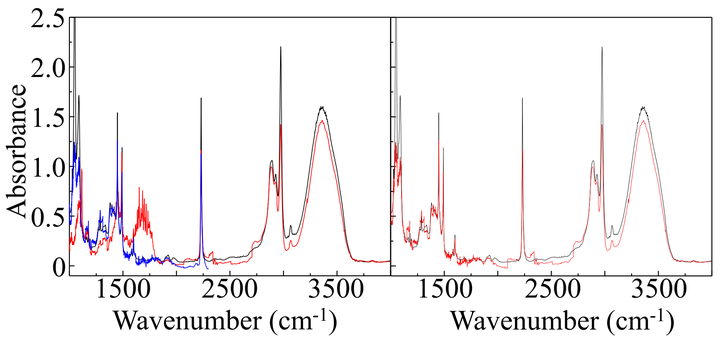
SECOND ORDER EFFECT FROM MONOCHROMATOR



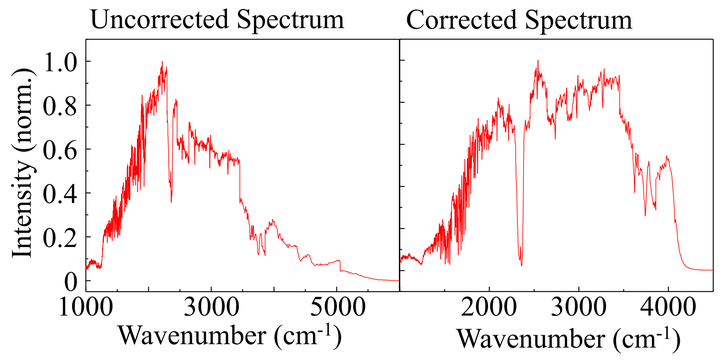
Detection through the monochromator uses diffraction grating which spectrally resolve the laser pulse. The 1st order diffraction signals are usually used for the frequency resolved signal. However, in the case of broadband pulse, it is unable to avoid signal from the 2nd order diffracted beam resulting distortion of the spectrum. To obtain undistorted spectrum, we measured the spectrum in two parts with two long pass filter.



Absorbance spectrum of water and benzonitrile mixture from continuum IR pulse and commercial FT-IR spectrometer were compared to check the obtained spectrum were proper or not.



Below are before and after corrected spectrum of continuum IR pulse



Conclusion & Acknowledgement

CONCLUSION

We were able to obtain broadband IR source via simple colinear geometry of nonlinear crystals using fundamental source from Ti:Sapphire regenerative amplifier. Since the obtained source was extremely broad, unwanted 2nd order diffraction signal from high frequency component overlapped with 1st order diffraction signal. After using two long pass filters, it was able to solve the detection problem occurred form diffraction grating. We are now able to generate continuum IR source to use for variable applications in ultrafast IR spectroscopy.

ACKNOWLEDGEMENT

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