

Third harmonic generation in self-focused filaments in liquids

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Abstract: Intense filaments of third harmonic generation have been observed in a variety of liquids when pumped by 1300 nm wavelength femtosecond light. The onset of third harmonic generation coincides with the threshold for self-focusing as deduced from n_2 measurements performed on the liquids using the z -scan technique. We have found self-phase modulation above the threshold leading to continuum generation at about 3 times threshold. In the third-harmonic regime, the third harmonic output power is independent of the input power. Our observations are consistent with those for similar processes in gases as described by a phase-locking model. The measured conversion efficiency was approximately 10^{-6} .

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References and links

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1. Introduction

Self-focusing phenomena have generated considerable interest for decades [1]. The availability of powerful ultrafast femtosecond (10^{-15} s) laser sources provided by amplified Ti:Sapphire lasers, and the explosion of interest in continuum generation [2] and spatial solitons [3] has reinvigorated interest in these phenomena. Self-focusing has been observed in many materials including solids, liquid, gases, and liquid crystals.

The formation of bright filaments can be an important consequence of self-focusing, which arises from a balance between the optical Kerr lens focusing effect in a positive nonlinear refractive index (n_2) material and defocusing due to either diffraction or plasma generation. This interplay results in a threshold for self-focusing. Under the assumption of a continuous-wave Gaussian beam, the critical power for self-focusing is given by [4]

$$P_{cr} = \frac{3.77\lambda^2}{8\pi n_0 n_2} \quad (1)$$

Here, the λ is the wavelength; and n_0 and n_2 are the linear and nonlinear refractive indices, defined through $n = n_0 + n_2 I$, where I is the laser intensity. This also somewhat applies to the pulsed case, except that it will be complicated by the temporal properties of the laser pulses [5].

Inside the self-focused filament, the intensity can be extremely high (for example, the intensity inside the filament in air can be as high as $5 \times 10^{13} \text{ W/cm}^2$ [5]). As a result, several nonlinear optical processes can be induced with supercontinuum generation the most notable [2]. Self-focusing third harmonic generation (THG) has been reported by several research groups the gas phase in atmospheric air [6,7,8], noble gases [9,10], and methane [11]. A high conversion efficiency of 0.1% and 0.2% in air has been observed.[6] In addition, phase-locking theory for this phenomenon was used to explain the high conversion efficiency [8].

To our knowledge, THG in self-focused filaments has only been reported in the gas phase. Here, we report on the observation of relatively efficient THG in a near-infrared (1300nm) self-focusing filament in five different liquids: toluene, chloroform, acetone, cyclohexane and water. We have found that the critical power for THG onset coincides with the threshold for self-focusing as the z-scan technique has been used to measure the second-order nonlinear refractive indices, n_2 of the measured solvents. The evolution of the self-focused THG with increasing fundamental power in toluene will also be discussed.

2. Experimental

Tunable laser pulses were provided by an optical parametric amplifier (TOPAS-OPA, Quantronix) pumped by a regeneratively amplified Ti:Sapphire laser (CPA2010, Clark-MXR). This laser system can produce laser pulses with a peak power of about 10^{12} W and a repetition rate of 1 KHz and 220fs duration. A pair of crossed polarizers with a half-wave plate has been used to continuously control the incident laser peak power. The wavelength was fixed at 1300nm and the spectral FWHM of the fundamental light is about 7nm. The

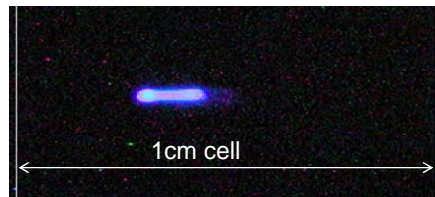


Fig. 1. Self-focusing THG filament in toluene. The fundamental wavelength was 1300nm (THG wavelength: 433nm) and the peak power is about 7 MW. The sample cell is 1cm long.

solvent samples were contained in a 1cm square cuvette. A 2.54 cm focal length lens focused the beam into the sample serving to induce the self-focusing process.

Spectral measurements were carried out at 90° using a spectrometer coupled to an intensified CCD (ICCD) camera (Princeton Instruments). For power dependence experiments, a photomultiplier tube (Hamamatsu R2255) coupled to a boxcar integrator replaced the spectrometer. Various combinations of filters have been used in front of the PMT detector as follows: (1) for determination of the critical power of self-focusing: one narrow-band filter centered at 433nm with 10nm bandwidth, two KG3 and one 1.0 O.D. neutral density filters; (2) for measuring the critical power of supercontinuum generation: one narrow band filter centered at 630nm with 10nm bandwidth was used and two KG3 and one 1.0 O.D. neutral density filters; (3) for measuring the total emission power dependence against the incident power: two KG3 and one 1.0 O.D. neutral density filters. The KG3 filter transmits in the visible (~300nm-850nm) but absorbs the infrared pump wavelength.

The THG efficiency was measured after transmission through the sample. The fundamental power was directly measured by an optical power meter. The THG was measured by a pre-calibrated photo diode (Thorlabs 210) with three KG3 color filters placed in front of the photo diode to block the fundamental light. The calibration of the photo diode was carried out by using a calibrated source of visible light (530nm) and one 3.0 O.D. neutral density filter, and measuring the input and output powers using the optical power meter and the photo diode respectively.

Z-scan was used to measure n_2 of all the five solvents [12]. A spatial filter has been built in front of the z-scan setup to ensure a Gaussian beam profile. The beam quality was constantly monitored by a laser beam profiler (Spiricon). The diameter of the beam following the lens ($f=10\text{cm}$) was measured with a knife-edge: $w_{0y}=38.5\mu\text{m}$, $w_{0x}=46.3\mu\text{m}$ (defined by $1/e^2$ of intensity). The Rayleigh range z_0 is estimated to be 3.58mm, while the sample cell thickness is either 1mm or 2mm. Depending on the magnitude of the n_2 values, two different intensity levels at the focal point have been used in the z-scan experiments: $70\text{GW}/\text{cm}^2$ and $199\text{GW}/\text{cm}^2$. Since the n_2 of the water and the quartz cell nearly equal, a 2mm thick cell was used to enlarge the nonlinear refractive response of water.

3. Results

A photograph of the self-focused third-harmonic filament is shown in Fig. 1. The threshold phenomenon of self-focusing THG is shown in Fig. 2, which depicts the emission spectra of toluene with the fundamental power of 4.3 MW and 4.5 MW, respectively. The two incident powers were intentionally selected to be slightly below and slightly above the threshold

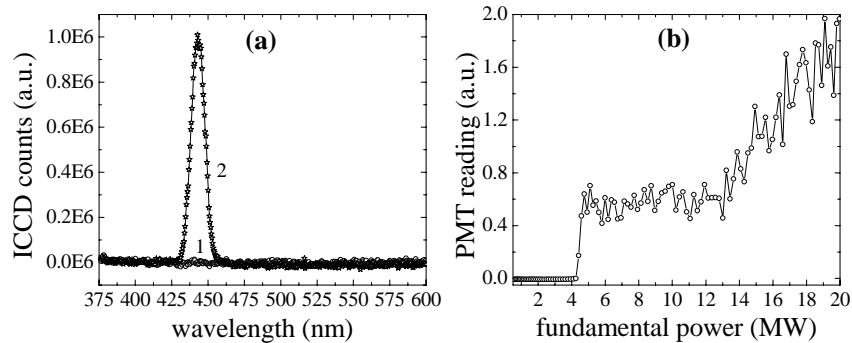


Fig. 2 Self-focusing THG at 1300nm in Toluene. In (a), curve 1 is the emission spectrum at a fundamental power of 4.3 MW and curve 2 is the emission spectrum at a fundamental power of 4.5MW; and (b) depicts the total power dependence on the incident power.

power. This graph clearly demonstrates that this third harmonic generation phenomenon is a threshold process. The self-focusing THG efficiency just above the critical power was estimated to be approximately 10^{-6} . Figure 2(b) depicts the power dependence of the total emission in toluene. In this case, no narrow-band filters were used (Two KG3 and neutral density filters were used to eliminate the fundamental.). In this figure, a critical point is evident at an incident power is 4.4MW. Above the threshold, a kink at about 13.2 MW divides the emission curve into two parts, corresponding to two different processes. It was found that the kink was found to be the critical power for supercontinuum generation, which was verified by a power dependence experiment using a narrow band filter away from the third harmonic centered at 630nm with 10nm band width and two KG3 and one 1.0 O.D. neutral density filter. Above the kink power, conical emission [5], visually observed as colorful rings behind the sample, confirms the appearance of a supercontinuum.

The critical powers of self-focusing in five different solvents have been measured. These are listed in the fourth column of Table 1. In order to verify that the third harmonic threshold corresponds to the self-focusing threshold, the nonlinear refractive indices n_2 of the five solvents were measured using the z-scan technique. The results are listed in the third column of Table 1. Using Eq. (1), the theoretical self-focusing critical powers were calculated and listed in the fifth column of Table 1. The values from the fourth column and the fifth column are generally in good agreement, indicated a coincident self-focusing threshold. We note that the values of n_2 show the same trend as reference [13], although the values are different, which we attribute to the different measurement techniques. We note that our femtosecond laser will only probe the electronic contribution, while those used in reference [13] would allow other effects (especially orientational) to contribute to the measurement.

By taking the self-focusing THG emissions spectra in toluene with various laser intensities, we examined the evolution of the self-focusing THG from pure THG to red-shift broadened THG and finally to the supercontinuum. As shown in Fig. 3(a), just above the threshold (e.g. curve 2), the THG peak has a narrow (instrumentation limited) spectrum (FWHM=11nm) at 433nm. As the fundamental power increases, the THG peak shows a red-shift and broadening. This is in contrast to that reported in atmospheric air [6,7], showing a blue shift of the self-focused THG with increasing incident power. When the incident power is about 3 times above the THG threshold, supercontinuum starts to appear as colored rings behind the sample. As the incident power increases, so does the supercontinuum. At 20 times above the threshold, the visible supercontinuum covers a broad spectrum range from 500nm-900nm (see the inset of Fig. 3(a)). The evolution of self-focusing THG can be described as followed: (1) below the threshold, no third harmonic generation is evident; (2) just above the threshold, strong THG appears; (3) above threshold, the THG spectrum starts to red-shift and broaden; (4) the supercontinuum appears at about 3 times above the threshold; (5) as power is increased further, an intense broad supercontinuum dominates the emission spectrum.

We also plot the spectral broadening of self-focusing THG as a function of the incident power in toluene in Fig. 3(b). This exhibits two linear broadening regions. From the self-

Table 1. Optical constants and critical powers of liquids.

<i>Solvent</i>	n_0 [13]	$n_2(10^{-20}m^2/W)$	$P_{cr}(exp.) (MW)$	$P_{cr}(cal.) (MW)$
Toluene	1.43	5.31	4.4	3.3
Chloroform	1.44	2.89	6.3	6.1
Acetone	1.35	1.85	7.9	10.2
Cyclohexane	1.44	1.43	10.0	12.3
Water	1.33	0.81	21.7	23.5

focusing critical power to the supercontinuum critical power, the broadening is fit by a line with slope 1.16nm/MW; above the supercontinuum critical power, the slope of the fitted broadening is 0.16nm/MW.

Shown in Fig. 2(b) is the power dependence of the generated light. Again, a kink is observed when supercontinuum generation begins to occur. In the first range, from the self-focusing critical power to the supercontinuum critical power, the emission is pure THG, and the total SHG power is constant although the THG spectrum broadens with increasing incident power. Above the supercontinuum critical power, the power dependence is more or less linear as a function of the incident power.

4. Discussion

In the measurements of n_2 , our values differ from those in reference [13]. Our values are more relevant for the THG experiments since they were carried out with the same femtosecond laser (including wavelength), with only electronic contributions

The self-focusing thresholds for THG observed here are several orders of magnitude lower than that observed in gases as Eq. 1 would suggest given the approximately 3 orders of magnitude larger n_2 in liquids as compared with gases. The high efficiency of the THG inside the filament can be understood using the theory of phase-locking [8]. Let us assume that light propagates in the z direction with the electric fields of the fundamental (ω) and THG (3ω) light written as $A_{\omega,3\omega} \exp(i\phi_{\omega,3\omega})$. The electric field amplitudes of both the fundamental and THG light, $A_{\omega,3\omega}$ are determined by the peak powers through $\sqrt{2P_{\max} / \pi w_0^2}$, where w_0 is the filament diameter, and $\phi_{\omega,3\omega}$ the phases of the fundamental and THG light. The possibility of phase locking can be shown from a calculation from reference [8],

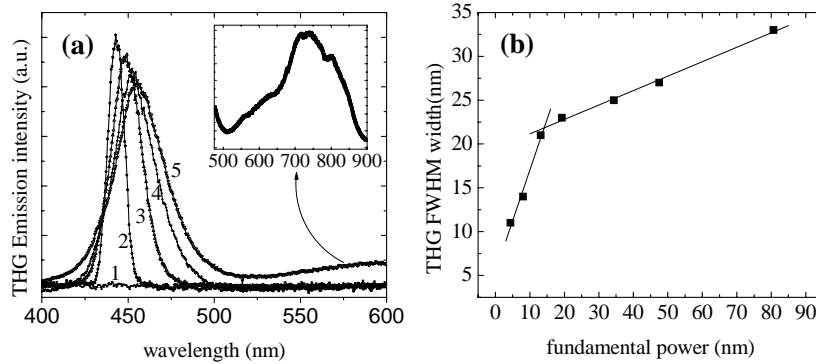


Fig. 3 Broadening and red-shift of the self-focusing third harmonic generation emission spectrum in toluene at 1300nm. The curves in graph (a) represent the following incident power: (1) 4.3 MW, (2) 4.5 MW, (3) 20 MW, (4) 50 MW, (5) 85 MW. Above 13.2MW, supercontinuum generation appears but is still negligible compared to the strong THG peak. When the power is about 80MW, the visually observed white light is intense and broad in spectrum. The inset depicts the spectral range of the supercontinuum. Graph (b) shows the FWHM spectral width of the THG in the incident power range between 4.4MW to 80MW. The kink at 13.2MW corresponds to the threshold for supercontinuum generation. The two power ranges in this graph are fit by linear function with slopes of 1.16nm/MW and 0.16nm/MW.

$$\frac{1}{2} \frac{\partial A_\omega^2}{\partial z} = -\frac{1}{4} \nabla_\perp (A_\omega^2 \nabla_\perp \phi_\omega) + \frac{L_{DF}}{L_{NL}} A_\omega^3 A_{3\omega} \sin(3\phi_\omega - \phi_{3\omega}). \quad (2)$$

Here $L_{DF} = k_\omega w_0^2 / 2$, $L_{NL} = (n_2 k_\omega I_0)^{-1}$. Below the THG threshold, $\frac{\partial A_\omega^2}{\partial z} \approx 0$ and $\frac{\partial \phi_\omega}{\partial z} \approx 0$, and all terms in Eq. (2) is zero. Above the threshold, in order to maintain coherence over great lengths, $\sin(3\phi_\omega - \phi_{3\omega})$ must be smaller than γ as defined by $\gamma = \left(\frac{L_{DF}}{L_{NL}} A_\omega^3 A_{3\omega}\right)^{-1}$. In our case,

γ is as small as 10^{-4} and, thus, the phase differences between two waves are either 0 or π since $\sin(3\phi_\omega - \phi_{3\omega}) \approx 0$ throughout the filament. Hence, based on the calculation, we believe the self-focusing THG that we observed is a result of phase-locking between the fundamental and the THG light waves.

The origin of the constant intensity of THG (Fig. 2(b)) above threshold in the filament is unclear, but a possible explanation is an effect similar to intensity clamping [14] as observed in filamentation of the fundamental. The self-focusing of laser pulses can be treated slice by slice. After the front part of the pulse reaches the clamped intensity, the later part of the pulse will be defocused by plasma generation and the corresponding energy will be transferred to the so-called background reservoir [5]. Increasing the fundamental intensity will result in an increase of the background reservoir but not in the intensity of the central self-focusing filament. The spectral broadening of the THG waves and its evolution from THG to supercontinuum can has been explained by self- or cross-phase-modulation [5].

5. Conclusions

We have reported on efficient third harmonic generation in self-focusing filaments in liquids. We have experimentally determined the self-focusing critical powers at 1300nm in five pure solvents: toluene, chloroform, acetone, cyclohexane and water. The z-scan technique was used to measure the nonlinear refractive indices, n_2 of the five solvents. Using the measured n_2 , the calculated critical powers for self-focusing in the five solvents show good agreement with the threshold for THG. A red-shift and broadening of the THG spectrum has been observed in toluene as the incident power increases. The broadening is linearly dependent on the incident power, about 1.16nm/MW. However, when the incident power is about 3 times higher than the self-focusing critical power, supercontinuum generation appears covering a broad spectral range from 500nm to 900nm. This represents an upshifted (relative to the laser fundamental) continuum suggesting new sources of short wavelength continua in condensed media. Phase-locking theory has been used here to explain the relatively high efficiency of the harmonic generation process.

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