FUNDAMENTAL RESEARCH

Enhanced Super continuum generation in water in the presence of ultra dilute solutions

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Intense ultrashort laser beam, after propagation through a transparent medium (water in this work), experiences self-focusing, thereby increasing the intensities in localized area to such high values so as to induce nonlinear optical effects like the generation of white light super-continuum and multi-photon absorption. We have carried out experiments for measuring super-continuum generation in water that contains ultra dilute solutes (of ethanol). Our observations of generation of super-continuum spectra in various solutes indicate that our measurements are able to make a distinction between normal water and water mixed with such ultra-dilute solutes i.e. Homoeopathic medicines.

Keywords: White light generation; non-linear dynamics; Ultra dilute solutions

Introduction

It is well known[1] that when an intense laser beam is incident on a transparent material, the material tends to self-focus the beam, which can result in even higher intensities. At such high intensities, several nonlinear optical effects set in, such as white light super-continuum generation, multi-photon absorption, ionization.[2] Super-continuum generation using femtosecond lasers has been of contemporary interest, where white light generations in transparent solids such as air, water, barium fluoride, and glass have been studied.[1, 3-5] Recently the influence of addition of organic molecules, such as proteins, in water, on the white light generation in water has also been studied.[6] The results of the study by C. Santosh et al. indicated suppression of white light generation particularly in the blue side of the spectrum, when small concentrations (μM) of α-amylase was added to water even though this protein does not absorb at the incident laser radiation at 800 nm.

There has been lot of experimental work involving supercontinuum generation in water.[2] Earlier work on ultrafast supercontinuum generation in water carried out by Alfano and coworkers (see ref. 1 and references therein) have demonstrated enhancement of super-continuum generation upon addition of inorganic ionic dopants like Zn^{2+} and K^{+}.

Here, we investigate supercontinuum generation in water containing organic molecules like alcohol. The alcohol that we investigate is obtained after ultra dilution. In the case of ultra dilute solutions it is suggested that solutes bring about molecular re-orientations in the solvent, and these structural changes have been observed by transmission electron microscopy by Lo *et al.*[7,8] In this work, we investigate the influence of ultra-dilute alcohol on supercontinuum generation in water. Since the process of preparing ultra-dilute solutions is very involved, we used the homoeopathic medicines which are known to be prepared by processes of ultra dilution. We thus measure the supercontinuum generation in water treated with plain ethanol and water treated with ethanol which was prepared by ultra-diluting various mixtures (hereafter referred to as treated ethanol).

Experimental setup

The laser used was a Ti:sapphire system with 1mJ energy, 45fs pulse duration, 1 kHz repetition rate at 800 nm.[4] The energy of the laser used in the present set of experiments was ~70 μJ. The laser beam was loosely focused using a 30 cm lens inside a 1 cm long cuvette of fused silica containing the sample. Before focusing, the laser beam was split and one part of the beam was used to monitor fluctuations in the laser

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energy. The spectra were recorded using a fiber-optic based spectrometer (Ocean-optics HR 4000), a dual spectrometer that simultaneously monitored the white light spectra and the reference spectrum of the incident laser. The reference spectrum was used for comparing and normalizing the fluctuations caused by laser energy (rms <2%) throughout the experiment.

The water used in the experiment was type-I water obtained from MILLIPORE system (Model MilliQ). The ethanol samples were obtained from the homoeopathic pharmaceutical company, Willmer Shwabe India Pvt. Ltd. Five series of ethanol samples viz. *Pulsatilla, Cuprum Met, Lachesis, Natrum Mur* and *Ruta* were used. The gas chromatography was carried out to identify chemical composition of the samples and the measurement indicated that samples composed of water and ethanol only in slightly varying proportions.

Table 1. The different treated samples used for measuring white light spectra.

Sample	Series	Potency 30	Potency 200	Potency 1M	Potency 10M
Pulsatilla	A	-	A2	A3	-
Cuprum Met	B	B1	B2	B3	-
Lachesis	C	C1	C2	C3	
Natrum Mur	D	D1	D2	D3	D4
Ruta	E	E1	E2	-	E4

Results and Discussion

Figure 1 show a typical UV-visible light absorption spectra of untreated ethanol and one sample from each of the treated ethanol. The UV spectra were obtained from the Shimadzu UV 2100 spectrophotometer. In this case, 20 μl of ethanol was added to 1ml of water. As can be seen there is essentially no difference in the spectra of the sample under study. This indicates that the linear absorption spectra are not able to show the difference between the treated and untreated sample.

The white light generation spectrum was measured using plain water, followed by the measurement of spectrum using untreated ethanol of 20 μl being added to 3ml of plain water. This spectrum is called as the *standard spectrum* with which all the other spectra were compared.

The spectra using untreated ethanol were repeated 4 times and were compared with that of the standard spectrum, as a function of the wavelength which is shown in Figure 2. As it can be seen from these five spectra that by repeating the plain ethanol in water, the super continuum generated in all the trials is nearly same.

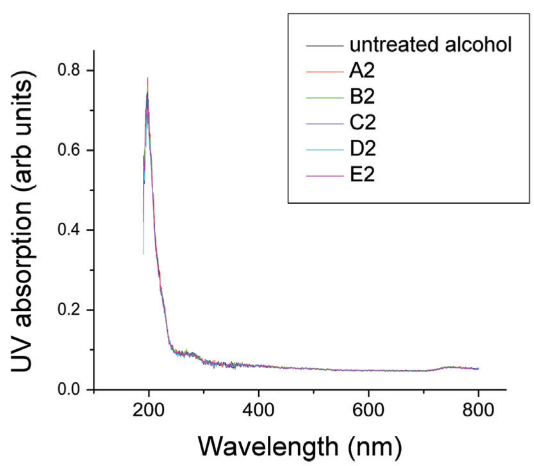

Figure 1: The ultraviolet and visible light absorption spectra carried out on the untreated and treated ethanol samples.

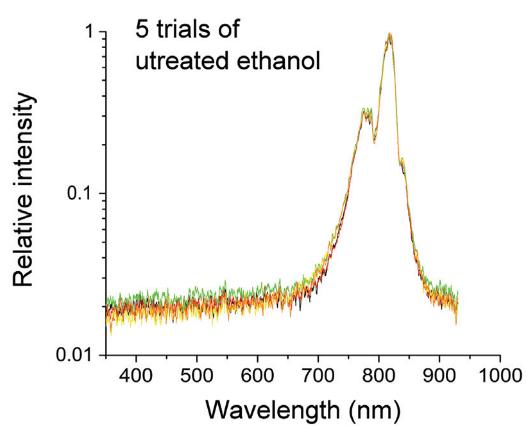

Figure 2: The spectrum generated by repeating for the untreated ethanol (4 times), and the intensity of the standard spectrum (black line), plotted as a function of wavelength

We also investigated the effect of quantity of ethanol added, on the spectrum; the quantity of untreated ethanol was increased from 20 µl to 100 µl in steps of 20 µl. The five spectra thus generated can be seen in Figure 3.

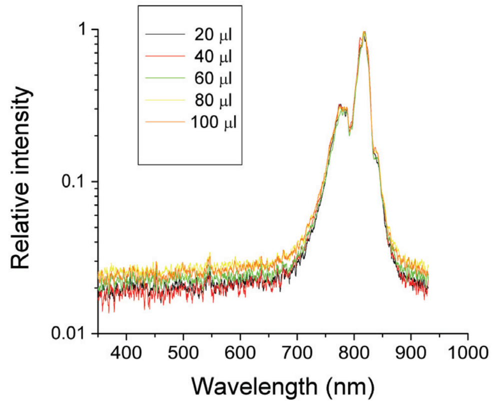

Figure 3: The comparison of the spectra generated with varying quantities of ethanol, and the standard spectrum, plotted as a function of wavelength

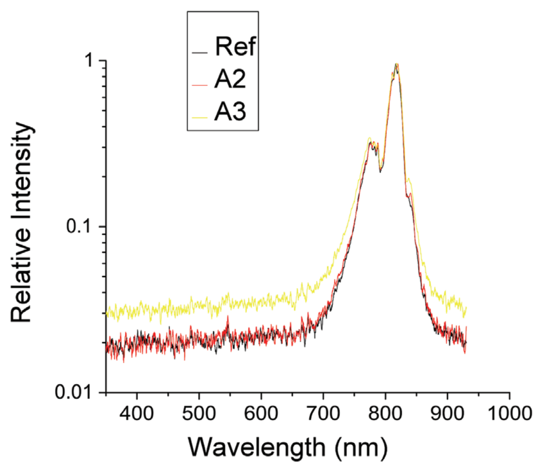

Figure 4: The spectrum measured for samples of series A plotted in comparison with spectrum of plain ethanol as a function of wavelength

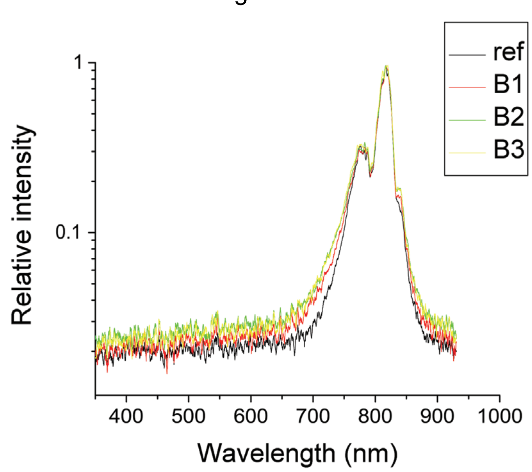

Figure 5: The spectrum measured for samples of series B plotted in comparison with spectrum of plain ethanol as a function of wavelength

It can be seen that there is an insignificant change in the spectra, from the standard spectrum, while increasing the quantity of ethanol in water by factor of 5. The effect of dilution on supercontinuum generation is measured in 5 different series of treated ethanol with varying dilutions. The ultra-dilute alcohol, their series names and potency (dilution) are tabulated in Table1. The potency 30 corresponds to dilution by factor of 30 and M denotes dilutions by factor of 1000.

In the subsequent section only series names will be referred to. Spectrum with each of the treated sample (20 µl of ethanol in 3ml of water) was collected and was compared with standard spectrum

After rinsing the cuvette with distilled water, 20 µl of treated ethanol, picked at random, was added to 3 ml of water. The spectra corresponding to various series along with the reference spectra of untreated (plain) can also be seen in the following Figures 4-8.

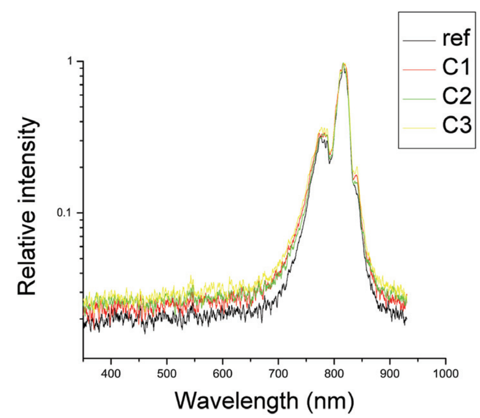

Figure 6: The spectrum measured for samples of series C plotted in comparison with spectrum of plain ethanol as a function of wavelength

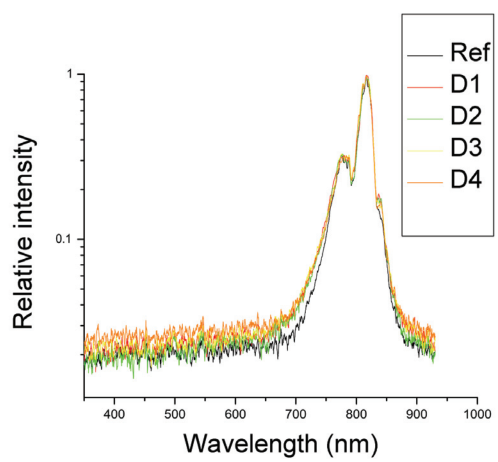

Figure 7: The spectrum measured for samples of series D plotted in comparison with spectrum of plain ethanol as a function of wavelength

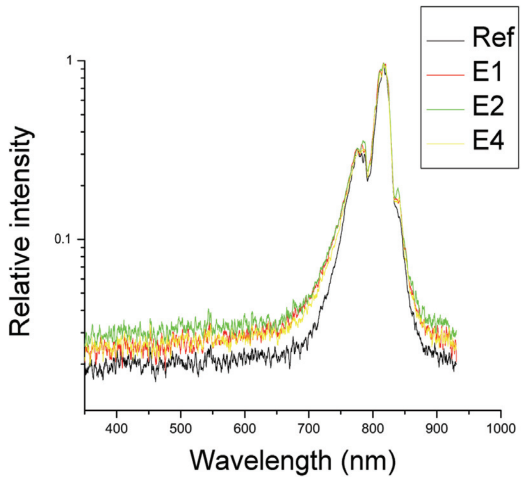

Figure 8: The spectrum measured for samples of series E plotted in comparison with spectrum of plain ethanol as a function of wavelength

In case of E2, the trial was repeated 5 times and the 5 different spectra generated were identical indicating that by repeated trials we eliminated the possibility of introduction of any random effects and the broadened spectrum with same treated ethanol was reproducible.

It is clear from Figures 4-8 that supercontinuum generation in water is enhanced after addition of treated alcohol compared to that with addition of plain alcohol. In order to quantify the differences in the spectra of treated sample compared to the reference sample wherein we observe an increase in white light generation in the blue and red side of spectra we measured the area under the curve for spectral range 550 nm to 750 nm. The area under the curve, from 550 to 750 nm region in the repeated trials of untreated ethanol fluctuated between values of 31 and 35 (arb.units), with an average of 33±2 (arb.units). In the case of ethanol for the concentration of 20, 40, 60, 80 and 100µ1 the area under the curve are 31, 32, 34, 39 and 38 (arb.units) respectively, giving an average of 35±4 (arb.units). We find a marginal increase of area under the curve when we increase the amount of untreated ethanol in water by factor of five. However, in the 550 to 750 nm range, the area under the curve measured for the treated ethanol, showed a significant increase, indicating an increase in the white light generation. The values are presented in **Table 2.** We also observe similar trend in the supercontinuum generation in wavelength regions greater than laser wavelength 800 nm (up to 930 nm, red region). Detailed investigation of supercontinuum generation in various media has been recently reported.[9]

The gas chromatography tests carried out indicated that all the samples contained water and ethanol only, and in similar portions. We had changed the alcohol content from 20 µl to 100 µl in steps of 20 µl. The data in figure 3 indicates no significant change (within error bar) in the area under the curve. This supports our claim that difference in alcohol content between treated and untreated samples, which is less than 0.1 %, has no effect on change in supercontinuum spectra between treated and untreated alcohol. This small difference cannot account for the significant increase in the supercontinuum generation. The physical factor like nonlinear refractive index n_2 might be playing a role in enhancing supercontinuum generation. It is known that supercontinuum generation[1] depends on the material parameter i.e. the nonlinear refractive index n_2, we are setting upexperiments to measure the n_2 value for treated and untreated sample.

Summary

The linear absorption spectrum does not show any variation in the plain and treated alcohol. The comparison of the supercontinuum generation spectra of water containing plain ethanol and the treated ethanol i.e. Homoeopathic medicines show increase in supercontinuum generation in water containing treated alcohol. Further experiments are required to identify the mechanisms that cause these variations.

Acknowledgements

We would like to thank Prof. Deepak Mathur for his support in carrying out the experiment and fruitful

Table 2: The value of super continuum generated in the range of 550 to 750 nm, in arbitrary units.

Sample	A	B	C	D	E
1	A	-	40	39	45
2	-	38	39	37	48
3	32	45	46	40	-
4	49	44	-	41	42

discussions. We would also like to thank Prof. M.S.Valiathan and Dr. S.K. Rohida for their useful discussions and suggestions. We are also thankful to Kritika Dota and K. Pradnya for their help during the experiment.

References

1. Alfano R R Ed, The Supercontinuum Laser Source, Springer, New York (1989).
2. Couairon A and Mysyrowicz A. Phys. Rep. 441, 2007: 47
3. Dharmadhikari A K, Rajgara FA, Reddy NCS, Sandhu A S, and Mathur D. Opt. Exp. 12, 2004: 695.
4. Dharmadhikari A K, Rajgara F.A. and Mathur D. App. Phy. B. 80, 2005: 61.
5. Dharmadhikari A K, Rajgara F A and Mathur D. Appl. Phys. B 82, 2006: 575.
6. Santhosh C, Dharmadhikari A K, Alti K M, Dharmadhikari J A and Mathur D. J BioMed. Opt.Lett. 12, 2007: 020510.
7. Lo SY. Anomalous State of Ice, Mod. Phys. Lett. B10, 1996: 909
8. Lo SY et. al. Physical Properties of Water with IE Structures Mod. Phys Lett. B10, 1996: 921.
9. Dharmadhikari Aditya K and Mathur Deepak. Propagation of Ultrashort Pulses in Condensed Media. Progress in Ultrafast Intense Laser Science *V* 98:81-108. Eds. Kaoru Yamanouchi, Antonio Giulietti and Kenneth Ledingham, Springer Berlin Heidelberg.

ERRATUM

1. K.C. Muraleedharan et al. Effectiveness of Homoeopathic Medicines in HIV patients - A Clinical Trial. Indian Journal of Research in Homoeopathy Vol. 4, No.4, Oct.- Dec. 2010: 29-35.

   In table 4, on page 32, the heading of the 1st column has been printed incorrectly. The heading in the 1st column should be WHO QOL Domain.

2. Girish Gupta et al. Evidence Based Clinical Study to Assess the Usefulness of Homoeopathic Medicines in Patients of Benign Prostatic Hyperplasia. Indian Journal of Research in Homoeopathy Vol. 4, No.4, Oct. - Dec. 2010: 49-56.

   On page 49, under Results: 10 medicines prescribed has been mentioned where as it should be 20 trial medicines.

   In table 3, on page 53, the numerical values given in the 3rd column have been printed incorrectly. The table is being reproduced as below:

Table 3 : Pre and post treatment status (Mean ± SE, n=43) of different parameters

Outcome measures	Pre treatment	Post treatment	Difference(%)
IPSS (score)	19.63 ± 0.78	9.77 ± 0.74**	50.29
Prostate weight (gm)	36.34 ± 2.08	32.78 ± 1.74**	9.80
PVRU (cc)	77.37 ± 11.75	67.22 ± 9.16ns	13.10
PSA (ng/ml)	1.74 ± 0.17	2.75 ± 0.40*	36.80
Maximum flow (ml/sec)	13.63 ± 1.59	13.79 ± 1.64ns	1.20
Average flow rate (ml/sec)	5.03 ± 0.51	6.02 ± 0.49**	16.45

ns – (p>0.05); * - (p<0.05); ** - (p<0.01)

The publisher apologizes for the inconvenience caused to the readers.