Original Article

Supercontinuum generation in water doped with Homeopathic medicines

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Abstract

We have carried out systematic studies to investigate the effect on supercontinuum generation in water using 40 fs laser pulses when doped with Homeopathic medicines. We perform these studies using five series of medications with different levels of dilution (10-30 to 10-100000). We measure supercontinuum spectra that span from 400-1050 nm. We monitor the area under the curve in the range 450-750 nm for each sample at a fixed incident laser energy. Our observations indicate that the yield of supercontinuum generation, in water containing Homeopathic medicine is significantly different from that obtained in water containing plain ethanol. The measurement for different dilutions shows up to 7 times standard deviation variation in the yield of supercontinuum generation. Even though linear absorption in the UV-visible region does not show any significant difference for different Homeopathic medicines, the supercontinuum yield which depends on the effective nonlinear refractive index changes with different samples.

Keywords: Supercontinuum generation, water, ultradilution, homeopathic medicines

Introduction

Ultra-high dilution, which is the basis for the preparation of homeopathic medicines, may achieve dilutions level that surpasses the Avogadro's limit. After each dilution, forceful striking or agitation is performed. The dissolved air in a liquid such as water may give rise to nanobubble formation [1] during the force striking stage and tend to form a water cluster. Formation of stable water-stable clusters has been proposed, even without the dissolved air [2-7]. There have been earlier studies that have shown that despite ultra dilutions beyond Avogadro's number, there is a memory effect that is retained in the solvent [8]. The memory effect was observed in ultra-high dilution of lithium chloride and sodium chloride-based on the thermo-luminescence study when irradiated by gamma-ray, and x-ray [9]. Anagnostakos *et al.* [10] took chamomile extract and diluted solutions far beyond the Avogadro number. They employed dielectric relaxation spectroscopy and differential scanning calorimetry techniques to verify water clusters formation. Ultraviolet-visible absorption measurements have been reported in the ultra-dilute samples that showed significant changes compared to the control solvent [11]. For nearly three decades, Demangeat and his group have carried out a systematic study on Homeopathic medicines using a nuclear magnetic resonance

technique [12-13]. It should be noted that similar dilutions are performed in the preparations of Homeopathic medicines. The cluster formation or molecular organization of water [8] have been proposed in Homeopathic doses, based in water or alcohol [14-15], along with the presumptions that clusters remain stable over a long period. Very recently, density, viscosity, speed of ultrasonic waves, and refractive index are measured for samples of *Sulphur* in ethanol wherein the signature of *Sulphur* was found under the condition of extreme dilution [16].

If water cluster formation or molecular organization takes place during the dilution and succussions process, then the nonlinear optical effect such as supercontinuum generation might be different as compared to undiluted samples. The nonlinear optical effects are readily observed when an intense pulsed laser is utilized. In general, an intense ultrashort laser beam after propagation through a transparent medium, experiences self-focusing due to the optical Kerr effect, thereby increasing the intensities locally to very high values to induce nonlinear optical effects like the generation of supercontinuum and filamentation. The propagation of femtosecond duration laser pulses through transparent media [17] like water has attracted a lot of research attention owing to its application in the generation of acoustic waves [18], generation of free electrons and radicals for cancer therapy [19-22], bubble formation [23], and broadband supercontinuum source [24-25]. The supercontinuum that is generated occurs due to self-phase modulation (SPM), which is symmetric. The asymmetry is introduced predominantly in the blue side of the spectrum by processes such as space-time focusing, self-steepening, and plasma [26]. Earlier, supercontinuum generation (SCG) in water was observed to enhance due to the presence of metal ions such as Zn^{+} and K^{+} [27]. Recently, metal nanoparticles enhanced SCG when doped in water [28-30].

On the contrary to these observations, the addition of dopants such as proteins [31-32] and lactose [33] showed significant suppression of the blue side of SCG. The reduction in SCG was ascribed to the scavenging action of free electrons [31-32] that reduce the plasma mediated SPM. In the case of lactose, scattering was also found to be responsible for the reduction in SCG. In our earlier study, we have observed an indication of the difference in supercontinuum generation from the addition of diluted and undiluted samples in water [34]. Moreover, the linear chromatic dispersion of water also plays a vital role in suppressing SCG [35].

Experimental setup

In this work, we report systematic measurements of SCG in water doped with Homeopathic medicines and compare it with SCG in water doped with plain ethanol. If there is any change in the physical property of water post addition of the dopant, the nonlinear optical effect might be different as compared to reference plain alcohol sample. Since self-phase modulation is the dominant mechanism for SCG that occurs due to time-varying refractive index change, the addition of a small quantity of Homeopathic medicine to water may be a simple test to observe the changes in the nonlinear optical effects, if any. We describe in the following our measurements based on SCG in water doped with minute quantities of different samples purchased from commercial pharmacies. We perform measurements using these samples with different levels of dilutions. The water used in all our measurements is deionized, to which we add small quantities (20 μ l) of the sample. We compare the changes in SCG from water doped with plain alcohol and Homeopathic medicines. In this article, plain alcohol refers to unpotentized ethanol which includes neither any dilution nor any succussion.

Figure 1 shows a schematic of the experimental setup. The laser used was a Ti: Sapphire system at 800 nm with 40 fs pulse duration, 1 kHz repetition rate [24]. The energy of the laser used in the present set of experiments was $\sim 60 \mu\text{J}$. The laser beam was focused using a 20 cm lens inside a 1 cm long cuvette of fused silica containing the sample. The spectra were recorded using a spectrometer (Ocean-optics HR 4000). The water used in all our measurements was deionized water obtained from the Millipore system (Model MilliQ). The ultra-dilute ethanol samples are procured from the homeopathic pharmaceutical company (SBL India or Pekana Naturheilmittel GmbH), five series of homeopathic medicines (different levels of dilution) are used. Five sets of samples with different levels of dilution (10^{-30} to $10^{-100000}$) were used. The sample with commercial names as *Nat Mur*, *Lachesis*, *Ruta* (Pekana Naturheilmittel, GmbH) *Nat Mur* and *Pulsatilla* indicates (SBL India) are designated as sample A, B, C, D, and E respectively throughout this work. The prefix number 1 to 6 corresponds to dilution power of 30, 200, 1000, 10000, 50000 and 100000 respectively. It should be noted that at the time of our measurements the pharmacy could not supply us with SBL Brand *Nat Mur* and *Pulsatilla* of power 50000 and 100000. The reference sample used for SCG was always 20 μl of ethanol added to 3 ml of deionized water (0.67 % by volume).

Figure 1- Schematic of the experimental set up to measure supercontinuum generation. L_1 and L_2 are lenses; S is a cuvette filled with the sample.

Throughout the paper dilution is always followed by the process of succussion, (even though not stated explicitly), performed by the pharmaceutical companies from whom the medicines were obtained. The word “dilution” is used instead of the samples to stress the level of dilution and its impact on the spectral results. The powers of the medicines used are in centesimal or C scale. Plants, Animals and Minerals are three major sources of Homeopathic medicines. Hence, we have selected medicines representing three different sources.

Chemical Analysis

We also examined the possibility of the presence of any impurities in deionized water that might affect the supercontinuum generation. The deionized water used in all our measurements was tested using Inductively Coupled Plasma - Atomic Emission Spectrometry (ICP- AES), an emission spectrophotometric technique. The deionized water was found to contain only four trace elements B, Ca, Cu, and Si. Randomly picked samples of deionized water (3 ml) along with Homeopathic medicine also had four trace elements B, Ca, Cu, Si exactly same as 3ml of deionized water indicating that impurities in water did not affect our measurements.

We monitor the area under the spectra from 450-750 nm region for any changes due to doping. The spectra are average of ~ 300 laser shots and normalized with respect to the maximum intensity of the peak. The areas reported are in arbitrary units. The samples were prepared by one of the co-authors and supplied for blind studies to another co-author with code names, and the same was made in random order.

Results and Discussion

The femtosecond pulse-based supercontinuum generation is a nonlinear optical effect observed during its propagation in a transparent medium. The physical mechanism mainly responsible is self-phase modulation. Self-phase modulation occurs because of the intensity-dependent refractive index, which is defined as $n = n_o + n_2 I$, where I is the intensity of laser (I is proportional to E^2 (E is the electric field)), n_o is the linear refractive index. n_2 is the nonlinear refractive index that has a positive sign in the transparency range of dielectric media. In the temporal domain, the time-varying refractive index imparts a change in the phase of the pulse [25]

$$\Delta\phi(t) = (\omega_o/c) n_2 I(t) L,$$

where ω_o is the central frequency, I is the intensity of the laser, L is the length of the medium, this induces a change in frequency given by $\delta\omega(t) = -d\Delta\phi(t)/dt$ giving rise to an instantaneous change in frequency that is given by $\omega(t) = \omega_o + \delta\omega(t)$ causing broadening in the incident spectrum. During the rising part of the laser pulse, red-shifted frequency components are generated; on the other hand, during the trailing part, blue-shifted frequency components are generated. In general, self-phase modulation gives rise to symmetric broadening. The asymmetry in spectral broadening occurs due to self-steepening and plasma mediated self-phase modulation [26]. The spectral broadening is influenced by other factors that include band-gap of the material, chromatic dispersion, group velocity dispersion, multi-photon absorption, and ionization induced free electrons [17,26, 35]. In the spatial domain, the optical Kerr effect overcomes the diffraction of the beam, giving rise to self-focusing that increases intensity within the medium limited by free electrons that defocus the beam. The optical energy propagates in a series of focusing-defocusing sequence known as a filament, over a distance much longer than Rayleigh range of a lens. The intensity within a filament is clamped by plasma. The extent of spectral broadening in the blue side depends on the band-gap of the medium and intensity clamping.

The supercontinuum generation spectrum was measured using deionized water, followed by the measurement of spectrum using 20 μl of plain ethanol added to 3 ml of deionized water. This spectrum is called the *base spectrum*, with which we compare all sample spectrum.

The measurements are performed in the following manner; one randomly selected Homeopathic medicine was tested four times, in no particular order, and their spectra were compared to quantify random fluctuations. SCG spectrum for different series of samples and their dilutions were measured. The area under the curve in the wavelength range of 450 nm to 750 nm is analyzed. We initially carried out experiments at two different incident laser energies of 20 μJ and 60 μJ . The sample used in this measurement was 20 μl of sample A (different dilution) added to 3 ml of deionized water. Figure 2 shows the SCG spectra at two incident energies keeping the focal length fixed. The incident laser is centered near 800 nm; we observe an asymmetric broadening in the SCG spectrum, particularly in the blue side. Earlier [26], it was found that such asymmetry depends on incident energy, beyond a threshold value of 2 μJ asymmetric supercontinuum was observed in water. In fig. 2, both the incident energies are beyond this threshold value, and we observe asymmetrically broadened SCG.

Figure 2- Supercontinuum spectra are shown for sample A for dilutions at two different incident energies. Base spectra correspond to water doped with undiluted alcohol.

It is clear that with 20 μJ of incident energy, SCG for different dilutions of Sample A is nearly identical. On the other hand, at 60 μJ of incident energy, SCG for different dilutions of Sample A shows a detectable variation in the spectra. It should be noted that in our earlier work [34], UV-Visible absorption measurement did not show any detectable difference for diluted samples.

In figure 3 (a) and 3 (b), we present the area under the curve in the spectral range 450-750 nm corresponding to data shown in fig. 2. At 60 μJ , we observe a detectable change. Increasing the incident energy beyond 60 μJ , we see the formation of bubbles. Thus, all our measurements were performed at an incident energy of 60 μJ . We also carried out careful measurements to check whether the orientation of the cuvette (containing water sample) with respect to the incidence of the laser beam has any effect on supercontinuum generation. The cuvette was oriented at angles of 0° , 1° , and 15° . Again, we quantify this effect in terms of the area under the curve of spectrum in the wavelength range 450-750 nm region. In the case of 0° the area under the curve is measured to be 13.1 (arb.units), and for 1 degree, it is 13.6 (arb.units). At a large angle of 15° the area under the curve reduces to 10.8 (arb.units). Thus, our measurement shows that the variation in the SCG is significantly less mainly for the orientation of cuvette at lower angles. In this work, we ensured that the cuvette was oriented at 0° . We now discuss the supercontinuum generation in water doped with various samples.

Figure 3- The graph shows the area under the curve of supercontinuum spectra for different dilutions of sample A at two incident energies.

It should be noted that the trace levels of metals are of the same order in water to which either 20 μl of plain alcohol or 20 μl of medicine samples were added. Thus, comparing the SCG of water with medicated samples and plain alcohol will result in similar background conditions in both samples.

1) Sample A

The area under the curve for sample A for different dilution samples are shown in figure 4(a). In this case, sample A2 showed a higher area under the curve compared to plain alcohol (undiluted). The sample A2 was repeated four times randomly during the experiment. The areas are shown in Fig. 4(b). The area under the spectrum in the range of 450 – 750 nm is 30.7 ± 0.4 (arb units). From figure 4(b), it can be seen that the area differs from plain alcohol by at least 1σ .

Where the value of 1σ is 0.4 in arb. Units. However, for different dilutions, the variation in the area is much more significant than 1σ to 7σ . In figure 4(a), the error bar, obtained from sample A2 is taken to be same (percentage) for all the dilutions of sample A. Similar is the depiction in subsequent samples.

Figure 4- The graph shows a histogram of the area under the curve of supercontinuum spectra for (a) different dilutions of sample A (b) sample A2 repeated four times randomly.

2) Sample B

The supercontinuum spectra are measured in sample B, and the area under the spectrum is plotted in figure 5. The sample B1 to B6 indicates different dilutions.

Four measurements are performed for Sample B5 (of all the B samples, B5 was chosen randomly) measurements, and the average area for the B5 sample was found to be 22.0 ± 1.2 arb units. For different dilutions, the difference from plain alcohol (undiluted) is either 1σ or 2σ in most of the cases (Fig.5) except B2.

Figure 5- The graph shows a histogram of the area under the curve of supercontinuum spectra for (a) different dilutions of sample B (b) sample B5 repeated four times randomly

3) Sample C

Figure 6 shows the supercontinuum spectra measured for different dilutions of sample C. We observe a dip in the spectra around ~ 630 nm corresponding to an inverse Raman effect [31]. In the case of sample A we did not observe a dip in the spectrum (see fig.2 graph pertaining to $60 \mu\text{J}$). Moreover, in sample C; we visibly observed the plasma filaments when the laser beam is focused inside the sample. Nevertheless, we did not visibly observe the plasma filaments for all other samples. When sample C6 was repeated four times, the average area obtained was 28.1 ± 1.5 . However, the difference between the samples of the C series and the reference spectrum was around 2σ .

Figure 6- The graph shows supercontinuum spectra for different dilutions of sample C.

4) Sample D

The area under the curve for Sample D for different dilution samples are shown in figure 7(a) the sample D3 was repeated four times randomly during the experiment. The areas are shown in Fig. 7 (b). The average area under the wavelength range of 400-750 nm was 19.0 ± 0.3 . For different dilutions of Sample D, the areas differ from that of plain alcohol by 1σ to 7σ , as shown in fig 7 (a).

5) Sample E

The SCG in Sample E for different dilution is carried out; the area under the curves are shown in figure 8(a). The sample E3 was repeated four times randomly during the experiment. The area is shown in Fig. 8 (b), and the average area under the spectra was observed to be 34.8 ± 1.1 (arb units). In the case of different dilutions of Sample E, it can be seen that area under the curve for the region 450 nm -750 nm of SCG differs from plain alcohol by more than 1σ .

Figure 7- The graph shows a histogram of the area under the curve of supercontinuum spectra for (a) different dilutions of sample D (b) sample D3 repeated four times randomly.

Figure 8- The graph shows a histogram of the area under the curve of supercontinuum spectra for (a) different dilutions of sample E (b) sample E3 repeated four times randomly.

In earlier theoretical work, it was predicted that water doped with silver nanoparticles showed a lower threshold for SCG and one octave of broadening in the spectrum. The observed broadening was attributed to an enhancement in the effective nonlinear index coefficient due to the silver nanoparticles. [28]. Recently, the addition of a minute quantity of gold and CdS nanoparticle was shown to affect supercontinuum generation in water [29]. We have earlier demonstrated that doping water with gold nanoparticles enhance SCG in the region 620-855 nm due to surface plasmon resonance compared to the neat water [30]. Our experimental observation of changes in the SCG yield can be rationalized based on changes in the effective nonlinear refractive index due to the addition of minute quantity (20 μ l) of **diluted alcohol (sample)**. Even though we have observed changes in SCG yield, it is difficult to *a priori* predict, enhancement or suppression of the SCG.

Summary

SC generation in water doped with Homeopathic medicine is measured using 40 fs laser pulses. By comparing SC spectra generated when water is doped with plain alcohol to that doped with **diluted alcohol**, we observe differences in its yield. However, the difference (beyond the effects of random variations in the measurements) in the generation of SC indicates that the two conditions of doping of water are physically different, pointing to changes in the optical property *vis-a-vis* nonlinear refractive index, which affect the generation of SC.

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