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DYNAMICS OF IONIZATION-ENHANCED SPECTRAL EXPANSION IN WATER INDUCED BY AN INTENSE FEMTOSECOND LASER BEAM*

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The dynamic process of white-continuum generation in water was investigated by the pump–probe technique with a femtosecond intense laser at 805nm. The spectrum width of the probe beam was broadened at the blue side and varied with different delay times. This blueshift was attributed to the ionization-enhanced optical nonlinearity, in which both the multi-photon ionization and avalanche ionization had an effect.

Keywords: supercontinuum, ionization, ultrafast
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I. INTRODUCTION

With the development of the chirped pulse amplification (CPA) technique[1,2] and broadband amplification media such as Ti:sapphire,[3] a high peak power laser source became available at the table top. These developments led the former expensive researches of interaction between high-intensity laser fields and materials (bulk media, cluster, atom and plasma etc) into laboratories. Many important phenomena, such as white-light generation (supercontinuum) and high-order harmonic generation, could then be widely studied. The response of materials under such an intense laser field would exhibit new features, which cannot be described by a single nonlinear process and perturbation theory.

Usually, under the radiation of a high-power laser beam, a nonlinear optical (NLO) phenomenon, such as white-light continuum generation, is mostly accompanied by strong self-focusing and ionization. White-light continuum generation has been observed in many condensed matters[4−7] and gases.[8,9] It is shown as bright white spots, most of which are accompanied by a rainbow-like pattern (cone emission). One of the most remarkable characters of this phenomenon is the ultra-broadband spectrum. This spectrum is extensively broadened in the blue side, with a relatively small broadening in the red side of laser wavelength. This supercontinuum beam was widely used as a continuum light source for spectrum applications.[10,11] However, there has not been a thorough understanding of its generation mechanism, even though it has been investigated for over two decades. The experimental results have been explained in a complicated way by several different nonlinear optical processes, including self-phase modulation (SPM),[6,12] ionization enhanced SPM,[5,13] four-wave mixing (FWM)[4,14,15] and Cerenkov-type processes.[16] Usually, the central part of white-continuum light, which is in the same direction or within a small angle to the fundamental laser beam, is interpreted as SPM[17] or ionization-enhanced SPM.[7] The accompanying conical emission is attributed to four-photon parametric generation or Cerenkov radiation.[16,18] More recently, a two-colour pump–probe experiment has been performed on the plasma-induced spectral broadening of rare gases.[19] Two processes, self-focusing and optical break-through, were commonly accepted as important experimental facts. In this paper, we report on a pump–probe experiment performed on a water sample. The temporal characteristics of white-light continuum generation in the liquid sample were investigated. Only the blue side extension of the probe pulse was clearly observed, which revealed the contribution from the ionization-enhanced nonlinearity. The decay

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process was also observed in the experiment.

Ⅱ. EXPERIMENTAL SET-UP

An ultrafast laser source from a Ti:sapphire CPA system at 805nm (TSA-10, Spectra Physics, USA) was used for the experiment. The pulse energy was 10mJ and the pulse duration was set as 280fs determined by a single-shot autocorrelator (Positive Light, USA) to obtain the best Gaussian-like spectrum output. The laser output was split into two parts by a beam splitter (BS). These two pulse trains with an intensity ratio of 10:1 were separately used as pump pulses and probe pulses. The optical length of the probe beam could be varied by an optical delay line, which was driven by a step motor. The probe pulse crossed the pump pulse with a small angle of 5° at the sample position, where a glass cell of 4.8cm in length, filled with pure water, was placed carefully to ensure that most of the cross section was enclosed. To determine the pulse duration *in situ*, the sample cell could be replaced by a thin BBO crystal for cross-correlation measurements. During the experiment, the probe beam was finally collected and detected by one channel of a dual-channel optical fibre spectrometer (PC2000, Ocean Optics, USA). The other channel of the spectrometer monitored the original laser spectrum as a reference in real time. Both the spectra were recorded by a computer program for further analysis. The transient behaviour was observed by driving the optical delay line. In Fig.1, the detailed experimental set-up is shown. The special position of P1 is designed for the purpose to be discussed below.

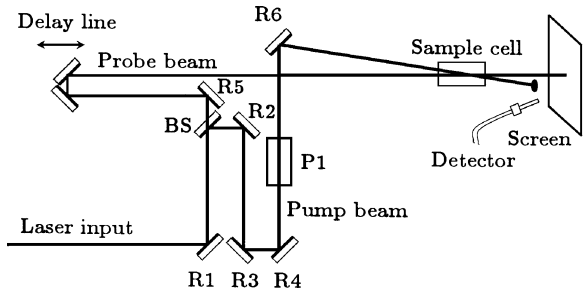


Fig.1. Experimental set-up.

Ⅲ. EXPERIMENTAL RESULTS AND DISCUSSION

To eliminate any spectral change of self-interaction of the probe beam, we checked the spectrum of the probe beam before and behind the sample cell in the absence of the pump beam. No distinguishable difference was found for the two spectra. This demonstrates that the spectral change of the probe beam during the experiment would be the effect of the pump pulse. The supercontinuum from the sample cell was generated by the pump pulse. Scattered white light could be observed along the optical path in the medium. The central part of the outgoing pulse was a small bright spot with a diameter comparable to the original pump beam. Coloured rings appeared around the central axis on the screen. The red ring had the smallest radius while the other rings had larger radii according to their wavelengths. Weak blue light appeared in the outermost ring. No self-focusing filaments were observed in this non-focusing configuration. In the measurement, the spectrum of the probe beam was detected at different time delays relative to the pump pulse. Figure 2 shows a typical broadened spectrum, which is located at the maximum broadening. It is clear that the broadening is on the blue side while no Stocks shift is observed on the red side. This broadening was found to vary with the time delay. Figure 3 shows the dependence of the broadening on the time delay. We define the broadening width as the difference between half of the maximum spectral intensities of the signals detected from the two channels of the probe beam passing through the sample cell and the reference laser beam. The maximum expansion is found to be 5.5nm. Due to the cross configuration of the two beams, the spectrum of the probe beam is broadened by the average effect of whole cross section, which will be explained in detail later.

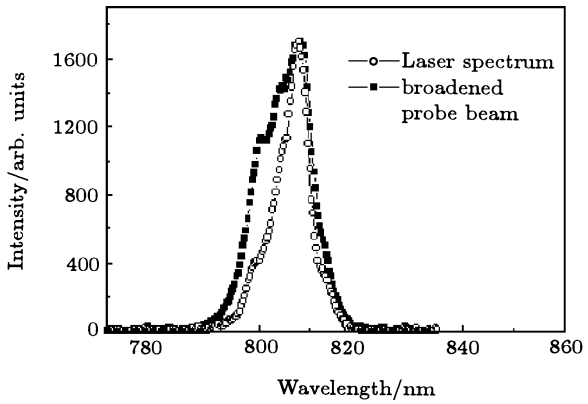


Fig.2. Spectrum of the laser and probe beams when the pump beam is presented. The spectrum expansion is clearly shown.

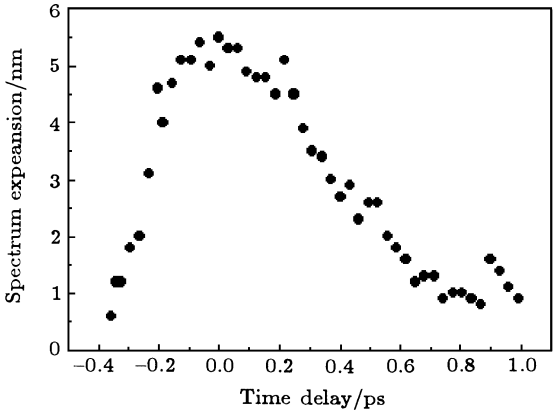


Fig.3. The decay dynamics of spectral broadening for the probe beam.

Using SPM theory, the spectrum broadenings, on the Stocks side ($\Delta\omega_-$) and anti-Stocks side ($\Delta\omega_+$), come from the largest nonlinear phase changing rate:

$$\Delta\omega_- = \left(- \frac{\mathrm{d}\Phi_{NL}}{\mathrm{d}\tau} \right)_{\min}$$

$$\text{and } \Delta\omega_+ = \left(- \frac{\mathrm{d}\Phi_{NL}}{\mathrm{d}\tau} \right)_{\max} \tag{1}$$

Φ_{NL} is the nonlinear phase from the path integration, which is described as:

$$\Phi_{NL}(\delta) = \int^{L} n_2 I(z,\delta) \, \frac{\omega_0}{c} \, \mathrm{d}z \tag{2}$$

where n_2 is the NLO refractive index and δ is the relative position along the pulse. By this means, the maximum Stocks or anti-Stocks broadening depends on the maximum change rate of the NLO phase. This is also applied to the probe beam, which is in a cross phase modulation configuration in this experiment. In the explanation of ionization-enhanced SPM, the fast process of multi-photon ionization (MPI), which can be regarded as an instant response, induces the production of free electrons of n_e. The free electrons show negative n_2, which can cancel the bulk nonlinear refractive index. This fast change in the overall spatial nonlinear refractive index induces the fast phase changing along the optical path. Thus a strong blueshift (anti-Stocks) occurred. The anti-Stocks shift is much larger than the Stocks shift in many experiments and proves to be the reality of this model. This fast change of the NLO refractive index can also affect the phase of the probe beam. By observing the spectrum-change process of the probe beam, the temporal resolution of the nonlinear refractive index change can be demonstrated. The probe beam exhibits a much smaller spectral extension on the blue side than the supercontinuum generation of the pump beam does. This is due to the cross overlap configuration, in which the probe beam experienced only a small ionized area induced by the pump beam. The corresponding phase difference integration and coupling are quite small. In contrast, the broadening on the red side is quite small for the probe beam and can even be neglected. This may also be due to the original small redshift of the pump beam.

For the ionization processes in media under high-intensity laser fields, two main mechanisms, MPI and avalanche ionization, will contribute to the generation of free electrons. The former process occurs instantaneously to the electrical field, while the latter needs a building time and is mostly considered immaterial[20] for the first few tens of femtoseconds. In our experiment, the laser pulse is 280 fs. Therefore, avalanche ionization may play a considerable role in the decaying part of the spectrum broadening. Because of the asymmetric rising and falling edges of signal, these two processes may be separated in the time sequence. MPI is the main mechanism for the fast appearance of the supercontinuum, and the avalanche ionization is attributed to the decay phenomenon of the blueshift of the probe beam. The MPI process is a transient phenomenon. If the MPI only is considered, the symmetric rise and fall of the shapes of the original laser pulse will lead to the symmetric blueshift on the rising and falling sides of the final observation. This is contradictory to the observation shown in Fig.3. Therefore, the conclusion could be made that different processes are present in the experiment. The MPI and avalanche ionization processes may both contribute to the amplitude of decay processes, and the latter breaks the temporal symmetry.

To confirm our conclusion, we must distinguish the two different origins of the delayed signal. One is the self-modulation of the pump pulse in water, which would make the pump pulse shape change and produce a 'tail' for itself. If this happens, the delayed process in Fig.3 could be explained as a non-delay response, but only instant response, of media. The other origin is the delayed nonlinear optical response of media. We placed the sample cell in position one (P1 in Fig.1) and then determined the cross-correlation signal of the probe and pump beams. Meanwhile, the cross-correlation signal of the probe and pump beams without a water sample cell is also presented in Fig.4.

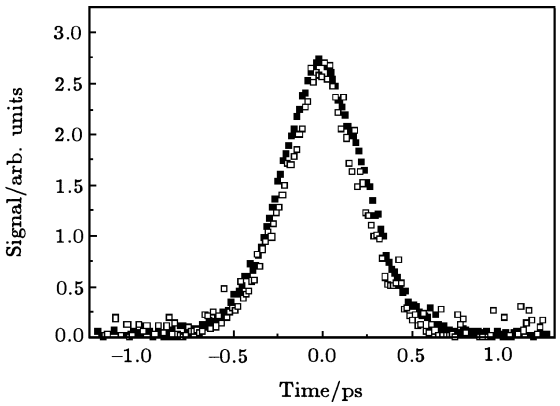


Fig.4. Cross-correlation signal (filled rectangle) of pump–probe beams. The open rectangle represents the signal obtained when the pump beam passes through the sample cell in position 1 (P1).

It is clear that the two cross-correlation signals are identical. They are symmetric and show no observable delay. This is very different from what is shown in Fig.2. This result proves that the observed delay in the spectral broadening of the probe beam is caused by the nonlinear response of water. Therefore, this temporal response represented the excitation dynamics of the condensed media. It is different from the previous work reported by Fork *et al*,[6] where they observed the dynamic response of liquids including water, and found that the response time is comparable to, or faster than, the width of the pump pulse, using an 80fs laser centred at 627nm. However, the intensity of their reference beam was not explained in their experiments.

IV. SUMMARY

In conclusion, an intense pump–probe technique was applied to investigate the dynamics process of white-continuum generation in water by monitoring the temporal and spectral change of the probe beam. The probe beam showed a clear anti-Stocks extension of 5.5nm in maximum while the redshift could be neglected. A delay process was observed and attributed to the ionization-enhanced nonlinearity in water. Two ionization processes, MPI and avalanche ionization, were employed to explain the experimental results.

References

[1] Strickland D and Mourou G 1985 *Opt. Commun.* **56** 219
[2] Martinez O E 1987 *IEEE J. Quantum Electron.* **23** 1385
[3] Perry M D and Mourou G 1994 *Science* **264** 917
[4] Alfano R R and Shapiro S L 1970 *Phys. Rev. Lett.* **24** 11 584
[5] Bloembergen N 1973 *Opt. Commun.* **8** 285
[6] Fork R L, Shank C V, Hirlimann C and Yen R 1983 *Opt. Lett.* **8** 11
[7] Brodeur A and Chin S L 1999 *J. Opt. Soc. Am.* B **16** 4637
[8] Francois V, Ilkov F A and Chin S L 1992 *J. Phys. B: At. Mol. Opt. Phys.* **25** 2709
[9] Ilkov F A, Ilkova L S and Chin S L 1993 *Opt. Lett.* **18** 681
[10] Wilson K R and Yakovlev V V 1997 *J. Opt. Soc. Am.* B **14** 444
[11] Glezer E N, Siegal Y, Huang L and Mazur E 1995 *Phys. Rev.* B **51** 6959
[12] Yang G Y and Shen Y R 1984 *Opt. Lett.* **9** 510
[13] Smith W L, Liu P and Bloembergen N 1977 *Phys. Rev.* A **15** 2396
[14] Wittman M and Penzkofer A 1996 *Opt. Commun.* **126** 308
[15] Luther G G, Newell A C, Moloney J V and Wright E M 1994 *Opt. Lett.* **19** 11 789
[16] Golub I 1990 *Opt. Lett.* **15** 6 305
[17] Xing Q R, Yoo K M and Alfano R R 1993 *Appl. Opt.* **32** 12 2087
[18] Nibbering E T J, Curley P F, Grillon G, Prade B S, Franco M A, Salin F and Mysyrowicz A 1996 *Opt. Lett.* **21** 1 62
[19] Le Blanc S P and Sauerbrey R 1996 *J. Opt. Soc. Am.* B **13** 1 72
[20] Feng Q, Moloney J V, Newell A C, Wright E M, Cook K, Kennedy P K, Hammer D X, Rockwell B A and Thompson C R 1997 *IEEE J. Quantum Electron* **33** 127