Temperature-Dependent Supercontinuum Generation from Femtosecond Filamentation in an Aqueous $CuSO_4$-solution

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(Received September 4, 2013)

Supercontinuum generation from laser filamentation in aqueous $CuSO_4$-solution is found to be strongly influenced by the solution temperature. The supercontinuum spectrum becomes broader as the solution temperature decreases from 65 °C to 5 °C. Correspondingly, the conversion efficiency of fundamental laser pulse to supercontinuum generation increases from 41.9% to 63.9%.

DOI: 10.6122/CJP.52.519 PACS numbers: 52.38.Hb, 42.65.Jx, 42.65.Ky

I. INTRODUCTION

The propagation of intense femtosecond pulses in liquid that results in filamentation and spectral broadening has been studied for more than ten years [1–10]. The broaden spectrum of the pulse, called supercontinuum generation, extends from ultraviolet to infrared range, which can find many applications in white light LIDAR, spectroscopy, and interferometry. The supercontinuum emission can be obtained in many kinds of media, including gases, liquid and transparent solid material. Many researchers have investigated femtosecond filamentation in those media, and the sample used is not confined to pure material. It is found that the laser interaction with mixed samples, for example water doped with scattering polystyrene microspheres [11] or noble metal nanoparticles [12], show some unique characteristics of filamentation and supercontinuum generation. Recently, an aqueous solution of $CuSO_4$ has been used as an absorptive medium, because it has a strong absorption around 800 nm of the laser central wavelength as well as a high transmission on blue side, and finally a unique supercontinuum spectrum with a flat plateau in the visible range can be obtained [13].

Refractive index of a liquid and laser parameters are important factors that determine the characteristics of supercontinuum spectrum. In the case of solution, it is known that, in addition to the concentration of the solution which affects the index of refractive, the temperature also influences its refractive index [14]. On the other hand, the temperature variation will affect stability of laser-liquid interaction, especially under high repetition rate laser condition; therefore, the temperature is an essential parameter which has to be considered besides the solution concentration and the laser parameters if one wants to

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obtain a stable and efficient white-light source. However, up to now there are no reports of temperature-dependent filamentation or supercontinuum generation in liquid samples. In this paper, we studied experimentally the temperature effect on the supercontiuum generation induced by femtosecond laser pulses from a solution of $CuSO_4$. The results show that the generated supercontinuum spectrum highly depends on solution temperature. Thus the supercontinuum generation can be optimized by means of temperature control, and it could be an easy method to get a flat and efficient supercontinuum spectrum to be more suitable for applications.

II. EXPERIMENTAL SETUP

The experimental setup for generating supercontinuum from $CuSO_4$ is illustrated in Figure 1. The laser employed in the experiment is an amplified Ti:sapphire femtosecond laser system that generates laser pulses at central wavelength of 800 nm, with the maximum pulse energy of 4 mJ, pulse duration of 50 fs, and repetition rate of 1 kHz. The linearly polarized femtosecond laser beam is loosely focused by a lens (lens L1, f = 400 mm) into a 4 cm long fused silica cuvette containing the solution of $CuSO_4$. The pulse energy is adjusted with neutral density filters. The concentration of the solution in our experiment is chosen to be 0.035 mol/L. Another lens (lens L2, f = 50.8 mm) is used to collect the laser beam into an integrating sphere. A spectrometer (USB 4000, Ocean Optics) is used to record the supercontinuum spectra. In order to control the temperature of the solution, we attached a home-made thermoelectric cooler to the cuvette. The temperature of the solution can be controlled from 5 °C to 75 °C continuously.

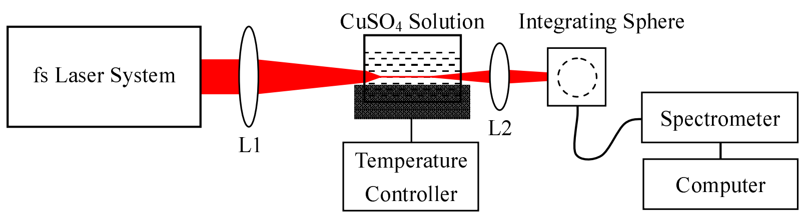

FIG. 1: Experimental setup for temperature dependent supercontinuum generation.

III. RESULTS AND DISCUSSIONS

Different from filamentation in air or solid, vapor bubbles will appear during the process of laser filamentation in liquid samples, and the bubbles greatly degrade the spectral repetitivity of supercontinuum. Laser-induced vapor bubbles are mainly influenced by laser intensity at the repetition of 1kHz we used. We found that the vapor induced by a laser pulse will interact with the followed laser pulses, and in our case the influence of the bubbles on filamentation and supercontinuum generation can be reduced to a great extent when relatively low laser pulse energy of 10 μJ is used.

Figure 2 shows the spectra of the supercontinuum generated from laser filamentation in the aqueous $CuSO_4$ solution under conditions of different temperatures. We can see from the Fig. 2 that the temperature results in spectra with different degrees of spectral broadening. The supercontinuum spectrum mainly covers a spectral range of from 720 nm to 850 nm at temperature of 65 °C, and it spreads from about 450 nm to 950 nm which covers more than 500 nm at temperature of 5 °C. Lower solution temperature leads to a relatively wider spectral broadening and a stronger emission especially in the visible range, i.e. a higher conversion efficiency of supercontinuum generation.

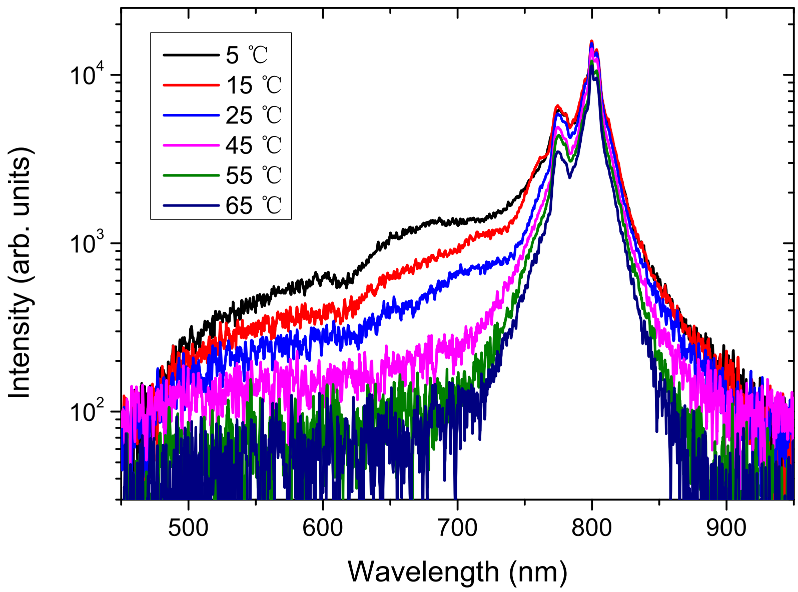

FIG. 2: Supercontinuum spectra generated in the solution of $CuSO_4$ at different temperatures varying from 5 °C to 65 °C.

In order to investigate the effects of temperature quantitatively, we define the conversion efficiency as the energy ratio of the continuum part (spectral range of $<$ 785 nm and $>$ 815 nm) except the fundamental part (spectral range of 785–815 nm) to the whole. As shown in Fig. 3, the supercontinuum conversion efficiency decreases linearly with the increase of the solution temperature. Under the condition of our experiments, at the temperature of 5 °C we got the highest conversion efficiency of 63.9%, and a broadened spectrum of supercontinuum which covers at lease from 450 nm to 950 nm. The spectral intensity in the visible range at wavelength of 600 nm for example in the case of 5 °C is two orders of magnitudes higher than that in the case of 65 °C, and the corresponding conversion efficiency is more than 1.5 times higher than that at temperature of 65 °C.

It is known that the decrease of the temperature will result in an increase of the linear and nonlinear refractive index of the liquid sample, which would lead to a lower critical power for self-focusing, hence to a lower clamped intensity of filament. This would

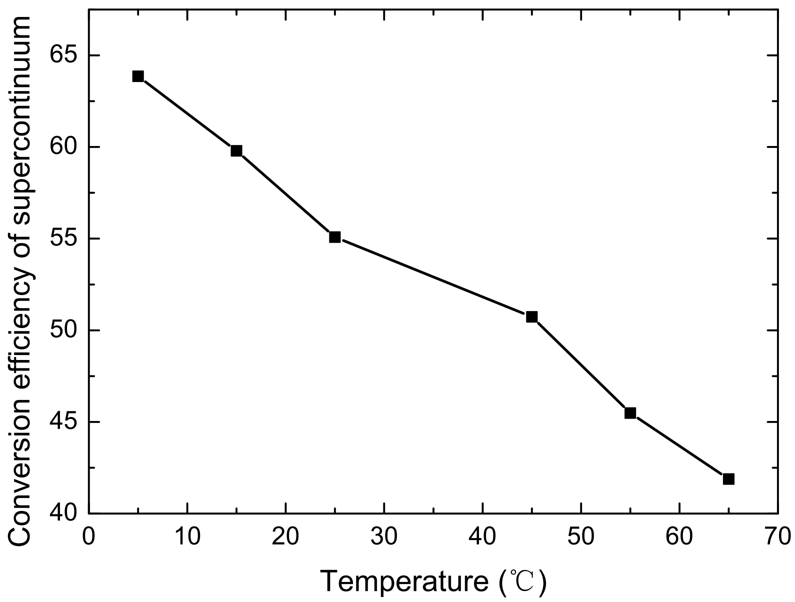

FIG. 3: Supercontinuum conversion efficiency as a function of temperature.

give rise to a weaker interaction, and consequently a narrower SC emission is expected. However, our results show that the SC from $CuSO_4$ becomes broader as the temperature decreases, this behavior can be explained as the following. When filamentation forms in liquid with a high repetition rate laser as the case of our experiment, the laser would heat up the filament zone very efficiently, especially when an absorbing medium is used. This heating would induce some nonuniform heat flow in the filament region. This turbulence will perturb the focusing, thus the laser intensity in the filament zone, and this would reduce the intensity in the zone. The larger this perturbation is, the lower the intensity in the filament zone will be. A colder medium would dissipate heat more efficiently than a warmer one. Thus, when the liquid is colder, heat dissipation is more efficient. This leads to weaker turbulence, resulting in higher laser intensity in the filament zone. A higher intensity would give rise to more ionization, hence, more self-steepening, and consequently a broader SC spectrum is obtained.

Compared with supercontinuum from pure water, the overall intensity of the SC from $CuSO_4$ solution is lower, especially near the pump wavelength. This is due to that the SC is modulated by the transmission of $CuSO_4$ solution, whose transmission at shorter wavelength (400–600 nm) is high (80–90%), while its value sharply decreases to below 10% near the pumping wavelength [13]. Even though the efficiency of SC generation decreases in the case of $CuSO_4$, the use of the solution is favorable for a flat-plateau SC. Furthermore, the cooling of the $CuSO_4$ solution will, to some extent, compensate the loss of SC conversion efficiency.

IV. CONCLUSIONS

The effect of temperature on supercontinuum generation in the aqueous solution of $CuSO_4$ has been experimentally investigated. We have found that a lower solution temperature is favorable for a broader spectrum and higher conversion efficiency of supercontinuum, i.e. lower temperature enables more efficient supercontinuum generation. The underlying mechanism is attributed to that the lower temperature of the solution gives rise to a less turbulence in the filament zone. As a result, a lower temperature leads to a stronger nonlinear laser-sample interaction and consequently a stronger supercontinuum emission. The method of supertinuum generation control via solution temperature as shown in this work offers another degree of freedom to get a flat and much more efficient supercontinuum emission besides the one via concentration control [13].

Acknowledgements

We would like to thank Prof. S. L. Chin from Laval University, Canada for fruitful discussions. This work was supported by 973 program under Grant No. 2013CB922404, the National Natural Science Foundation of China under Grant Nos. 11274053, 11074027, 61178022 and 11211120156, the Doctoral Program of Higher Education under Grant Nos. 20122216120009, 20122216110007 and 20112216120006, and Funds from Sci. & Tech. Dept of Jilin Province under Grant No. 20130522149JH, 20111812.

References

[1] A. Brodeur, F. A. Ilkov, and S. L. Chin, Opt. Commun. **129**, 193 (1996).
[2] W. Liu *et al.*, Appl. Phys. B **75,** 595 (2002).
[3] W. Liu, S. L. Chin, O. Kosareva, I. S. Golubtsov, and V. P. Kandidov, Opt. Commun. **225**, 193 (2003).
[4] A. Dubietis, G. Tamosauskas, I. Diomin, and A. Varanavicius, Opt. Lett. **28**, 1269 (2003).
[5] V. P. Kandidov, I. S. Golubtsov, and O. G. Kosareva, Quantum Electron. **34**, 348 (2004).
[6] J. S. Liu, H. Schroeder, S. L. Chin, R. X. Li, and Z. Z. Xu, Opt. Express **13**, 10248 (2005).
[7] J. Yu, H. B. Jiang, J. Wen, H. Yang, and Q. H. Gong, Opt. Express **18**, 12581 (2010).
[8] N. Kaya *et al.*, Opt. Express **20**, 13337 (2012).
[9] Q. N. Cui, J. P. Yao, J. L. Ni, and S. Zhang, J. Mod. Opt. **59**, 1569 (2012).
[10] S. Suntsov *et al.*, Appl. Phys. Lett. **103**, 021106 (2013).
[11] V. Jukna *et al.*, Appl. Phys. B, **94**, 175 (2009).
[12] C. Wang, Y. X. Fu, Z. H. Zhou, Y. Cheng, and Z. Z. Xu, Appl. Phys. Lett. **90**, 181119 (2007).
[13] L. Wang, Y. X. Fan, Z. D. Yan, H. T. Wang, and Z. L. Wang, Opt. Lett. **35**, 2925 (2010).
[14] H. M. Dobbins and E. R. Peck, J. Opt. Soc. Am. **63**, 318 (1973).