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SUPERCONTINUUM SPECTRUM GENERATION IN THE LIQUIDS WITH DIFFERENT BAND GAP THRESHOLDS

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Supercontinuum spectrum generation by femtosecond laser pulses propagating in transparent liquids with different band gap thresholds is investigated. As the laser energy increases, the laser spectrum is strongly broadened and modulated, and finally a supercontinuum spectrum can be observed. The spectral broadening can be mainly attributed to the optical Kerr effect at low laser energy and the plasma created by the multiphoton excitation of electrons from the valence band to the conduction band at high laser energy. It is found that the spectral broadening and modulation strongly depend on the band gap threshold E_{gap} of the medium.

Keywords: Supercontinuum spectrum; band gap thresholds: self-focusing; self-phase modulation; plasma.

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1. Introduction

The propagation of an intense ultrashort laser pulse in a transparent medium can give rise to an optical filament and supercontinuum spectrum with a nearly white light from the UV to the near IR.[1–4] The nonlinear phenomenon has attracted considerable attention for several decades because of its extensive applications, such as tunable ultrashort pulses,[5] optical parametrical amplification,[6] lighting control,[7] and the detection and identification of pollutants in the atmosphere.[8]

Supercontinuum spectrum generation correlates with the optical Kerr effect and the plasma formation.[4] The optical Kerr effect can induce self-focusing, which

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causes the beam shrinkage and the increase of peak intensity. Plasma formation results from the multiphoton excitation from the valence band to the conduction band under intense femtosecond laser pulses. Optical filamentation is the result of the balance between the Kerr self-focusing of the laser pulses and the defocusing effect of the plasma.[1,9,10] The filamentation process is usually accompanied by a broadening of the laser spectrum (supercontinuum spectrum generation) and conical emission, which has been extensively investigated in theory and experiment.[3–14] Various models have been proposed to describe this phenomenon. Self-phase modulation following self-focusing and optical breakdown is believed to be the dominant mechanism,[11] whereas self-steeping,[12] material dispersion,[13] and stimulated Raman scattering[14] have contributions to the supercontinuum spectrum generation as well. For the different transparent media, their nonlinear refractive index and band gap threshold E_{gap} are different, which results in different supercontinuum spectrum generation. The dependence of supercontinuum spectrum generation on the band gap threshold E_{gap} of extended transparent media has been investigated by Brodeur and Chin[15] using 140 fs Ti:sapphire laser pulses. However, to our knowledge, the modulation dependence of the supercontinuum spectrum was not discussed up to now. In this paper, we investigate the supercontinuum spectrum of 40 fs Ti:sapphire laser pulses propagating in transparent liquids (water, methanol, benzene, and carbon disulfide) at various input laser energies. The broadening of the laser spectrum is usually accompanied by angular emission (i.e., conical emission), our interests are focused on investigating the on-axis supercontinuum spectrum. It is found that the supercontinuum spectrum is strongly broadened and modulated, which depends on the band gap threshold E_{gap} of the medium.

2. The Experiment

The experimental arrangement for supercontinuum spectrum generation is shown in Fig. 1. A mode-locked Ti: sapphire laser (Spectra-Physics TSA-25 regenerative amplifier) is used as the excitation source with the pulse duration of about 40 fs, the repetition rate of 10 Hz and the center wavelength of 800 nm. The beam shape is near Gaussian distribution and its diameter is about 7 mm. The output beam is reflected by a flat mirror into a 120 cm long cell full of water, methanol, benzene or

Fig. 1. Experimental arrangement for the supercontinuum spectrum generation.

carbon disulfide, respectively. In the experiment, the output maximum peak intensity (about 162 GW/cm^2) far exceeds the self-focusing threshold of the liquids, so the optical filamentation in the transparent liquids, such as water, methanol, benzene, and carbon disulfide, can generate without a lens. The filamentation process is usually accompanied by a broadening of the laser spectrum and conical emission. The on-axis spectrum (supercontinuum spectrum) is measured by an Ocean Optics OOIBase32 spectrometer. At high input laser energy, a blue filter BG18 is placed before the spectrometer to filter the strong laser background around 800 nm.

3. Results and Discussion

Experiments are performed in transparent liquids including water, methanol, benzene, and carbon disulfide. Supercontinuum spectrum generation can be observed under intense femtosecond laser fields. The dependence of the supercontinuum spectrum on the laser energy in the methanol is shown in Fig. 2. As shown in Fig. 2(a), the laser spectrum is slightly broadened at the laser energies of 1.6 mJ (dashed line), and is intensely broadened toward anti-Stokes side and modulated as the laser energy is increased to 2.3 mJ (solid line). As the input laser energy is further increased (as shown in Fig. 2(b), the supercontinuum spectrum is strongly broadened and intensely modulated, and finally a strong supercontinuum spectrum is generated with a maximum broadening of about 13520 cm^{-1}.

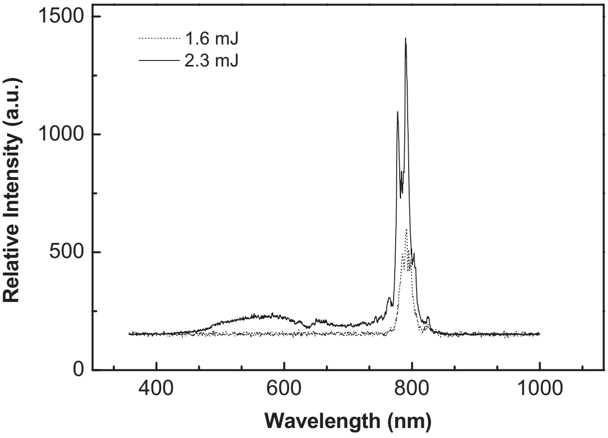
Supercontinuum spectrum generation correlates with optical Kerr effect and plasma formation.[4] Both the effects give rise to the nonlinear contributions, the refractive index in the medium, and is given by[16]

$$\Delta n = n_2 I(r,t) - \frac{2\pi e^2 N_e(I(r,t))}{m_e \omega_0^2}, \tag{1}$$

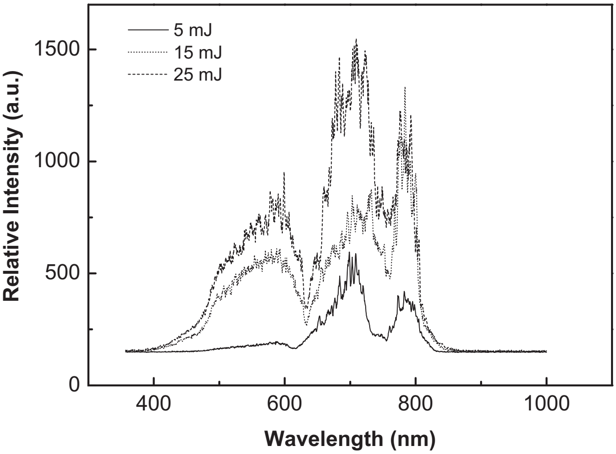
where n_2 is the nonlinear refractive index of the medium, $\mathrm{N}_e(I(r,t))$ is the intensity-dependent free electron density, ω_0 is the optical carrier frequency, and m_e is the mass of the electron. According to the nonlinear phase $\Delta\phi = \Delta n k z$, the spectral broadening can be written as

$$\Delta\omega = -\frac{\partial \Delta\phi}{\partial t} = -\frac{\omega_0 n_2 z}{c}\frac{\partial I(r,t)}{\partial t} + \frac{2\pi e^2 z}{m_e \omega_0 c}\frac{\partial N_e(I(r,t))}{\partial t}, \tag{2}$$

where z is the propagation distance in the medium. The first term in Eq. (2) represents the contributions of the nonlinear refractive index n_2 to the spectral broadening. If n_2 is positive, lower frequencies (Stokes shift) can be created by the leading edge of a laser pulse and higher frequencies (anti-Stokes shift) can be created by its trailing edge. If n_2 is negative, lower frequencies (Stokes shift) can be created by the trailing edge of a laser pulse and higher frequencies (anti-Stokes shift) can be created by its leading edge. The second term shows that the spectral broadening arises from the plasma created by the multiphoton excitation of electrons from the valence band to the conduction band. The plasma causes a sudden drop in nonlinear phase during the intensity spike, which translates into a large anti-Stokes



(a)



(b)

Fig. 2. Supercontinuum spectra obtained from the femtosecond laser pulses propagating in methanol at the different laser energies: (a) at low input laser energies, (b) at high input laser energies (using BG18 filter).

broadening by SPM. The above two processes have a different dependence on the laser peak intensity. The spectral broadening is dominated by the optical Kerr effect at low energies and by the plasma contributions at high energies. At laser energy of 1.6 mJ, the slightly asymmetrical broadening of laser spectrum can arise from the optical Kerr effect, since the laser energy is too low for the plasma to be formed. By increasing the laser energy, the plasma is formed and the laser pulses come into being split,[17–20] and thus the laser spectrum is widely broadened and strongly modulated. Since the intensity of the laser pulses can be clamped by multiphoton ionization,[4,14,21–23] the maximum intensity will limit the spectral broadening, and so finally the spectral broadening will remain unchanged even if the laser pulse energy is continually increased (as shown in Fig. 2(b)). Moreover, the wavelengths greater than 850 nm are not observed in the experiment, different from that reported in Ref. 11, which can be attributed to the absorption of water in the range of Stokes wavelength in the long cell, while a significant Stokes broadening is observed for the short water cell (1 cm).

Whether or not supercontinuum spectrum is identical for different transparent liquids, the on-axis spectra in the transparent liquids with different band gap thresholds are compared at the laser energy of 25 mJ (as shown in Fig. 3). All the spectra show a strongly asymmetrical broadening toward higher frequency and their anti-Stokes shift $\Delta\omega_+$ are about 13270, 12120, 7730, and 4920 cm^{-1} for water, methanol, benzene, and carbon disulfide, respectively. Table 1 shows the anti-Stokes shift $\Delta\omega_+$, the band gap threshold E_{gap}, and self-focusing threshold P_{th}.[15] As shown in Table 1 and Fig. 2, it can be indicated that the anti-Stokes shift $\Delta\omega_+$ increases with increasing the band gap threshold E_{gap} and the self-focusing threshold P_{th}. The larger band gap threshold E_{gap} requires higher intense laser pulse achieved in media for enough free electrons generated by the multiphoton ionization from the valence band to the conduction band, and thus it is sufficient to accumulate enough nonlinear phase for supercontinuum generation. So, the larger band gap threshold E_{gap} implies a larger nonlinear phase and thus larger anti-Stokes broadening.

As shown in Fig. 3, another striking observation is that the supercontinuum spectrum is modulated except for carbon disulfide. It is well known that group-velocity dispersion can play important role in self-focusing and could induce the splitting of pulse into two or more pulses, which can result in the modulation of

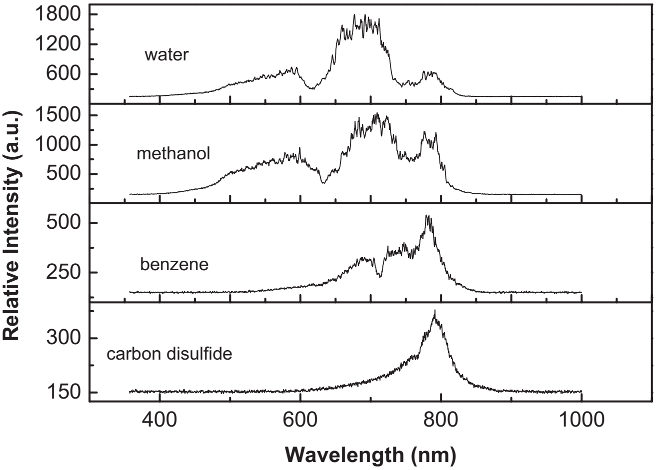


Fig. 3. Measured spectra in water, methanol, benzene, and carbon disulfide at the laser energy of 25 mJ (using BG18 filter).

Table 1. The anti-Stokes shift $\Delta\omega_+$ of the laser pulses propagating in water, methanol, benzene, and carbon disulfide at laser energy of 23 mJ, and the band gap threshold $E_{\rm gap}$ and the self-focusing threshold $P_{\rm th}$.

	Water	Methanol	Benzene	Carbon Disulfide
$E_{\rm gap}$ (eV)	7.5	6.2	4.5	3.3
$P_{\rm th}$ (MW)	4.4	3.9	0.9	0.23
$\Delta\omega_+$ (cm^{-1})	13270	12120	7730	4920

supercontinuum spectrum. The larger band gap threshold $E_{\rm gap}$ implies larger anti-Stokes broadening, and it can induce the split pulses with different wavelength in the propagating process, which results in different modulation period of generated supercontinuum spectrum. So, the larger band gap threshold $E_{\rm gap}$ suggests the larger modulation period of the generated supercontinuum spectrum, which accords with the experimental results. Compared with the other liquids, carbon disulfide has lower band gap threshold $E_{\rm gap}$, and the induced maximum self-focusing peak intensity is lower, and so the generated supercontinuum spectrum has lower anti-Stokes broadening. Accordingly, it is reasonably believed that the pulse splitting in carbon disulfide is inconspicuous and therefore no modulation in the generated supercontinuum spectrum is observed.

4. Conclusion

In conclusion, the supercontinuum spectra of femtosecond laser pulses propagating in transparent liquids with different band gap threshold (water, methanol, benzene, and carbon disulfide) have been investigated. As the laser energy increases, the laser spectrum is broadened and strongly modulated, and the supercontinuum spectrum generation strongly depends on the band gap threshold $E_{\rm gap}$ of the medium. The spectral broadening is mainly attributed to the optical Kerr effects at low laser energy and to the plasma created by multiphoton excitation of electrons from the valence band to the conduction band at high laser energy. The spectral modulation can be attributed to the laser pulse splitting, which depends on the band gap threshold $E_{\rm gap}$. The experimental results indicate larger band gap threshold $E_{\rm gap}$ which implies larger spectral modulation.

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