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ABSTRACT

Spectral broadening is an important nonlinear effect during the interaction of intense femtosecond laser pulses with air. In this paper, we experimentally study the dependence of spectral broadening on pressure and pulse energy. It is found that increasing both pressure and pulse energy can enhance the spectral intensity, while only increasing pressure leads to obvious blue shift of spectra, and the pulse energy has little effect on it. To get a better insight of the mechanism of spectral broadening, the numerical simulation relating the instantaneous frequency to the time dependent pulse intensity and time dependent plasma density in air is carried out. The results indicate that the plasma generation induced self-phase modulation which is related to the pressure plays an important role in blue shift of spectrum. Besides the transmission of the laser pulse is measured and decreases with pressure. It proves that the energy transfer of the laser pulse is promoted by pressure. This study will be helpful to understand a deeper physical mechanism of femtosecond filament induced supercontinnum generation in air.

I. INTRODUCTION

Propagation of an ultrafast laser pulse in Kerr media produces a stable plasma channel when Kerr self-focusing and plasma defocusing effects reach a dynamic balance, which is also named as filamentation.[1] This procedure is accompanied by lots of nonlinear optical effects, whose inherent features are spectral broadening and producing new frequency components.[2–5] Self-phase modulation (SPM) is one of the consequences of nonlinear interaction with the medium during filamentation, which is caused by the temporal variation of the refractive index in an optical medium. It can make the spectra broaden toward both the red and the blue sides.[6] Consequently, the white light supercontinuum (SC) generated during filamentation in air generally consists of a white central part surrounded by a rainbow-like conical emission due to these nonlinear optical effects when the ultrafast laser pulse propagates in Kerr media, which is called conical radiation.[6–8] The radial order of the spectral components is inverse of diffraction with bluer frequencies appearing on the outside rings. SC can be generated not only in gaseous media,[9] but also in liquid[10] and solid media.[11] The wavelength of SC ranges from the ultraviolet region to the infrared region.[5] Due to these superior properties of SC, it has attracted numerous attention and has been successfully employed in many applications, including femtosecond time-resolved spectroscopy,[12,13] generation of ultrafast laser pulse,[14–16] seed pulse of optical parametric amplification,[17] light detection and ranging,[18] remote sensing,[19] biomedical imaging,[20] molecular fingerprint spectroscopy,[21] and so on.

SC generated in air has its advantages over that generated in other media: first, it can be generated in desirable spatial positions where the femtosecond laser pulse can reach, second, the air can recover rapidly after being damaged by the high power laser pulses compared to other materials, therefore, significantly high pulse energy can be used to generate SC.[22–28] However, due to the complicated spatiotemporal dynamical process and environment condition, getting controllable SC generation in air still faces some challenges. To solve this problem, it is a crucial issue to understand the physical mechanism of SC in air. In 1999, Chessa *et al.* temporally and angularly resolved the ionization-induced refraction and the ionization-induced blue shifted spectra in helium gas.[29] To make it be better used, the performance of SC in air at different pressures is also required. Therefore, we conduct the experiment in air at various pressures from 490 Pa to 100000 Pa which correspond to an extremely high vertical attitude. We also investigate the physical mechanism of spectral broadening in air about how SPM induced by plasma effect affects the spectral broadening by combining the experiment and the simulation. The transmission of the pulse energy is also recorded to make the experiment substantial.

In this paper, we investigate the influence of the pressure on the spectral broadening effect of femtosecond laser pulses by conducting experiments in a gas chamber, where the gas pressure is controllable. In addition, the simple simulation relating the instantaneous frequency to the time dependent pulse intensity and time dependent plasma density in air, and the simulation of the temporal evolution of plasma density are conducted to study the relationship between the pressures and the blue shift of the spectrum. By comparing the simulated results with experimental observations, we find that the SPM induced by plasma generation related to the pressure is an important factor of blue shift of the spectrum. Besides, we also measure the transmission of the incident laser energy at different pressures.

II. EXPERIMENTAL SETUP

To study the influence of pressure on spectral broadening induced by femtosecond filament and collect spectra at different pressures, a stable and controllable pressure environment is required. Therefore, we assemble a cylindrical chamber whose length is 750 mm and radius is 50 mm. The laser beam can travel through the front quartz window to enter the gas chamber and depart through the back window. There are also two quartz windows on the middle of the cylindrical surface of the gas chamber for two obvious reasons: first, the information of the initial position of filament generated in the chamber can be seen through the window, then it is easy to adjust the position of the filament, second, the transverse fluorescence can be collected through the quartz window on the middle of the cylindrical surface of the gas chamber. The pressure in the gas chamber is controllable by a mechanical pump linked to the chamber. A capacitance diaphragm gauge lined to the chamber is used to supervise the vacuum degree, and a digital panel connected to the gauge is used to display the value of pressure.

A schematic of experimental setup is shown in Fig. 1. A Ti: Sapphire femtosecond Laser amplifier (Coherent Libra) is used to provide laser pulses whose duration, central wavelength, and repetition rate are 50 fs, 800 nm, and 1 kHz, respectively. The diameter of a laser beam is measured to be 10 mm. The maximal energy of an output laser pulse is 3.0 mJ, and the output energy can be adjusted by an energy controller composed of a half wave plate and a Glan prism. In our experiment, we measure the spectra when the input pulse energy E_{in} is 1.0, 1.5, 2.0, and 2.5 mJ, respectively, to explore the influence of pulse energy on spectral broadening. The filament induced by a femtosecond laser is generated in the center of the chamber focused by a lens with a focal length of $f = 400$ mm which is placed just in front of the front quartz window. Intense and stable broadened pulses generated by the filament propagate through the back window and a shortpass filter, whose cutoff wavelength is 780 nm which is used to avoid the disturbance of central wavelength. Then it is collected by an integrating sphere and guided to the spectrometer (Avantes-AvaSpec-ULS2048L) through an optical fiber. The exposure time is set as 2 ms, and spectra are accumulated 1000 times to reduce errors.

III. RESULTS AND DISCUSSION

The laser spectra and the pictures of filaments induced by femtosecond pulses at different pressures in the case of E_{in}= 2.0 mJ are shown in Fig. 2. Considering that it is a tremendous span from 490 Pa to 100000 Pa, in which the spectral intensity varies a lot, to study the spectra at different pressures specifically, the collected laser spectra are manually divided into two parts: spectra obtained when the pressure is lower than 1000 Pa and higher than 1000 Pa, which are shown in Figs. 2(a) and 2(b), respectively. It can be seen from Fig. 2(a) that the spectrum measured at 490 Pa (solid red curve) is almost identical to that at 680 Pa (dashed green curve) and 900 Pa (dotted blue curve), which indicates that the laser pulse does not experience obvious spectral broadening when pressure is lower than 1000 Pa. As we further increase the pressure, the spectral peak intensity increases drastically and the spectra experience obvious broadening, as shown in Figs. 2(b) and 2(c). The pictures of filaments at different pressures are present in Fig. 2(d). It can be seen in Fig. 2(d) that the lengths of filaments decrease with increasing pressure. In the following part, analysis of influence of pressure on spectral broadening will be conducted.

As is known, filamentation is accompanied by a strong spectral broadening, which depends on the strong reorganization of the temporal shape of the pulse. Spectral broadening is caused by several effects: self-phase-modulation, self-compression, pulse splitting, self-steepening, ionization of the medium, etc.[6] However, SPM plays a key role in spectral broadening accompanied by filamentation in air,[30]

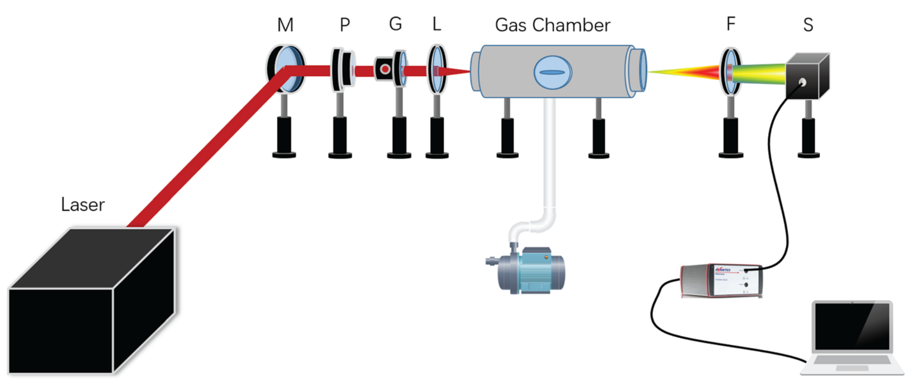

FIG. 1. Schematic of experimental setup: Laser: femtosecond laser, M: planar mirror, P: half wave plate, G: Glan prism, L: focusing lens ($f = 400$ mm), F: 800 nm filter, and S: integrating sphere.

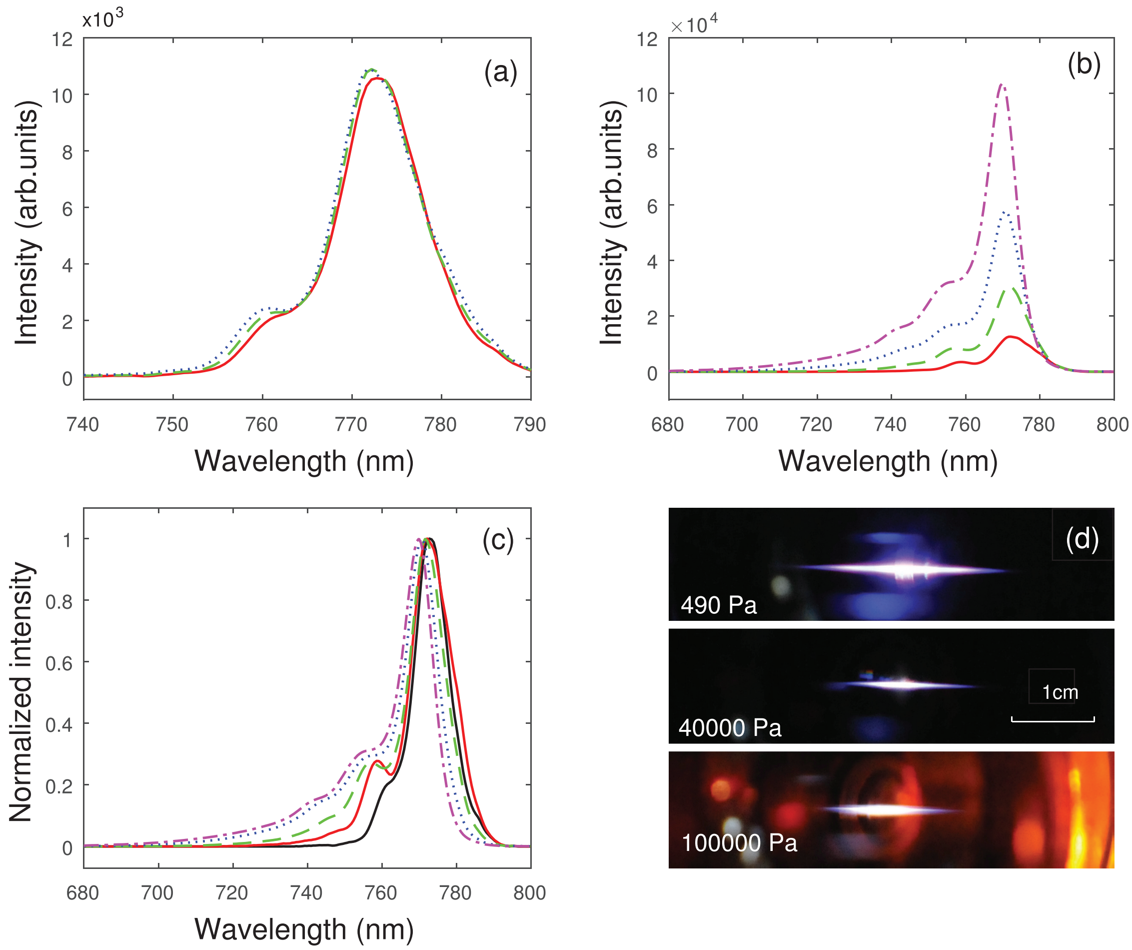

FIG. 2. Spectra measured at E_{in}= 2.0 mJ when the pressure is (a) lower and (b) higher than 1000 Pa, (c) normalized spectra. (d) Pictures of the luminous region taken from the middle quartz window when the pressure in the gas chamber is 490, 40 000, and 100 000 Pa, respectively.

which includes the SPM induced by the Kerr effect and plasma generation. The simulation relying on only SPM was conducted by several authors.[31,32] Whereafter, the simple simulation relating the instantaneous frequency to the time dependent pulse intensity and time dependent plasma density in air can be described as following equation:[6]

$$\Delta\omega = -\frac{\partial\Delta\varphi}{\partial t} = \frac{\omega_0 Z}{c}\left(-n_2\frac{\partial I(r,t)}{\partial t} + \frac{1}{2n_0\rho_c}\frac{\partial\rho_e(r,t)}{\partial t}\right), \quad (1)$$

where $\Delta\varphi$ is the variation of phase, ω_0 is the central frequency of a laser pulse, Z is the propagation distance in media, n_2 is the second-order nonlinear refractive coefficient, ρ_c denotes the value of the critical plasma density above which the plasma becomes opaque and satisfies $\rho_c = \varepsilon_0 m_e\omega_0^2/e^2$, $\rho_e(r,t)$ is the plasma density, and m_e is the mass of an electron. The right-hand side of Eq. (1) contains two terms: the first term describes the contribution of Kerr effect which leads to both the Stokes broadening and the anti-Stokes broadening of the spectra; the second term describes the contribution of plasma generation resulting in the anti-Stokes broadening. It should be mentioned that in Eq. (1), the duration of ultra-short laser pulse is on the femtosecond scale which means that the evolution of filament in a single shot is prompt. The model in this simulation takes into account the main physical effects proposed to be responsible for the self-channeling of ultrashort laser pulses in air. The temporal evolution of plasma density (ρ_e) satisfies the rate equation[33]

$$\frac{\partial\rho_e}{\partial t} = W(I)(\rho_{at} - \rho_e) + \frac{\sigma}{n_0^2 U}\rho_e I - a\rho_e^2. \quad (2)$$

In Eq. (2), the photoionization rate $W(I)$ describes the probability of ionization of an atom with potential U which obeys the results of the Perelomov-Popov-Terent'ev (PPT) ionization model.[34] In process of femtosecond filamentation in air, multiphoton ionization (MPI) is dominating at the moderate intensities which is less than 10^{14} W/cm^2 due to the intensity clamping, which means the tunnel ionization can be neglected.[6] Therefore, the first term at the right-hand side of Eq. (2) describes MPI in the filamentation dynamics and the parameter, $W(I)$, can be approximately expressed by $W(I) = \sigma_K I^K$, and $K = mod(U/\hbar\omega_0) + 1$ is the minimum number of photons needed to ionize the gas medium whose value depends on the gas species. In consideration of the gas species in the chamber, U in the second term represents the mean ionization potential of air $U = 14.6\,\text{eV}$.[6] The minimum number of photons and the ionization cross section of MPI corresponding to the mean ionization potential of air are $K = 10$ and $\sigma_K = 1.38 \times 10^{-128}$ cm^{20}/W, respectively.[35] Furthermore, the inverse bremsstrahlung cross section $\sigma = 5.1 \times 10^{-20}$ cm^2 in a standard atmospheric pressure and the last term describes the electron recombination with coefficient $a = 5 \times 10^{-7}$ cm^3/s which can be attained in Ref. 36. Some of the parameters in Eqs. (1) and (2) are related to the pressure: $n_2 = n_2(p_0)\tilde{p}$, $\rho_{\text{at}}(p) = \rho_{\text{at}}(p_0)\tilde{p}$ and $\sigma(p) = \sigma(p_0)\frac{\tilde{p}[1+\omega_0^2\tau_0^2(p_0)]}{\tilde{p}^2+\omega_0^2\tau_0^2(p_0)}$. Here, we set $n_2(p_0) = 3 \times 10^{-19}\text{cm}^2/\text{W}$, $\tilde{p} = p/p_0$, where p_0 denotes a standard atmospheric pressure and p stands for the practical pressure of gas.[36] What is more, the second term and third term at the right-hand side of Eq. (2) describe the contribution of the cascade ionization and radiative electron recombination which assures a saturation in time of the electron plasma. The cascade ionization is of secondary importance for the femtosecond pulse, and the contribution of radiative electron recombination to the overall phenomenon is negligibly small on the femtosecond timescale.[34,35,37,38]

To investigate the influence of SPM to blue shift of spectrum conveniently and qualitatively, and analyze the tendency of the temporal evolution of plasma density briefly, some assumptions are required. First, the profile of the laser pulse in the time domain is assumed to be Gaussian type invariably which is $I(t) = I_0 \exp(-2t^2/\tau_p^2)$ (where I_0 is set as the clamped intensity 8×10^{13} W/cm^{-2} and τ_p is the pulse duration whose value is 50 fs) during its propagation in the interaction region, neglecting the dispersion, self-steepening effect, self-compression, and self-splitting effect which can influence the pulse profile in the time domain dramatically. The dispersion effect can be neglected due to the long dispersive length of the laser pulse we use in the experiment at standard atmosphere pressure. (The dispersive length is longer at low pressure.) It should be noted that for pulse self-compression, changing the gas species, the gas pressure, or the initial pulse energy leads to similar pulse compression. However, the peak intensity of the maximally compressed pulse varies which is depending on the ionization potential of the gas species. Therefore, the self-compression dose not influence the clamped intensity in our experiment.[39] What is more, we also neglect the diffraction effect which influences the spatial profile of the laser pulses. As the pressure increases, all the nonlinear indices in the nonlinear terms, such as the Kerr effect, plasma defocusing, and MPI, scale with the pressure, and the corresponding balance between them should be unaffected by the change of pressure.[36,40] Second, in our work, only the pulse intensity along the propagating axis is taken into consideration for we do not focus on the spatial distribution of the pulse after propagating in air. Besides that, it should be noted that the clamped intensities for various pressures are comparable for the same incident pulse energy, but slight differences are obtained when the geometrical focusing condition is changed.[41] Therefore, the clamped intensity of the filament changes little with the pressure whose regime is between 20 000 and 100 000 Pa, and we also neglect the contribution of electron temperature to the nonlinearity which results in distortion of the laser pulse so that we set it as a constant.[36,41,42] Therefore, the temporal evolution of plasma density is only the function of time. The filament length Z can be got from Figs. 2(d) and supplementary material, Figs. S1(g) and S1(h), which also varies with pressure. As is seen from Figs. 2(d) and supplementary material, Figs. S1(g) and S1(h), the filament length decreases with increasing pressure, and in the region from 20 000 and 80 000 Pa, the filament length changes little, which is around 20 mm. Based on these approximations, the simulation is conducted simply and the results are depicted in Fig. 3. Since the pulse duration is 50 fs in our experiment, the simulation is conducted on the timescale from −150 fs to 150 fs to guarantee the precision of calculation.

Figure 3(a) depicts the variation of pulse intensity with time. According to Eq. (2), the variation of plasma density with time and the partial derivative of plasma density at different pressures can be calculated by the second order Runge-Kutta method. In the simulation, the step width is set to 0.3 fs. The partial derivative of time dependent pulse intensity is also simulated, and then the change of frequency can be calculated according to Eq. (1). Figure 3(e) shows the change of frequency at 100 000 Pa, and the simulation at other pressures are also conducted which shows the same tendency with Fig. 3(e), as shown in supplementary material, Figs. S1(a)–S1(f). The result indicates that the spectral broadening, specifically, blue shift of spectrum is mainly caused by the contribution of the SPM induced by plasma generation, while the SPM induced by Kerr effect has little influence on spectral broadening. In addition, according to Eq. (1), the spectral blue shift comes from two parts: $-\frac{\omega_0 Z}{c} n_2 \frac{\partial I(\text{back part of the pulse})}{\partial t}$ and $\frac{\omega_0 Z}{2cn_0\rho_c}\frac{\partial \rho_e}{\partial t}$. The first part and the second part which represent the SPM induced by Kerr effect and plasma effect are both positively proportional to the pressure, because the variation of pulse intensity is negative and the parameter n_2 and the variation of plasma density with time are positively proportional to pressure. Therefore, the spectral broadening related to the partial derivative of time dependent plasma density is positively proportional to the pressure, which is demonstrated in Fig. 3(f) and has similar tendency to the experimental results in Fig. 2. It means that the spectra broaden toward blue side by increasing the pressure. However, there are some differences between results got from experiment and simulation by contrasting Fig. 2(c) and Fig. 3(f). The reason for this nuance is discussed below. First, the assumption we make is neglecting some nonlinearity which can distort the pulse profile. Besides, spectral broadening is calculated by the equation $\lambda = \frac{2\pi c}{\Delta\omega + \omega_0}$, in which the laser pulse is assumed as monochromatic light whose central frequency is $\omega_0 = 2.36 \times 10^{15}$ Rad/s. However, the laser pulses used in the experiment are not monochromatic light strictly, and its linewidth is about 40 nm. Therefore, the zero line of blue shift of the spectrum is not the central wavelength ($\lambda_0 = 800$ nm) and the linewidth should be considered.

The measurement of spectra under different incident pulse energies in the case of constant pressure is also carried out. Here, the spectra measured at 40 000 Pa and spectra measured at 100 000 Pa in Fig. 4 are presented, respectively. It can be clearly seen from Figs. 4(a) and 4(a′) that the intensity of both spectra measured at 40 000 Pa and those at 100 000 Pa increase with laser energy. The increase in the pulse energy will surely enhance the intensity of every frequency component of spectrum. What is more, comparing the variation of the peak intensity with pulse energy shown in Figs. 4(c) and 4(c′), the peak spectral intensity in Fig. 4(c′) is higher under the same pulse energy. However, no obvious spectral broadening can be observed with increasing pulse energy and the spectra are not totally the same, as shown in Figs. 4(b) and 4(b’). In effect, the spectra measured at other pressures show the

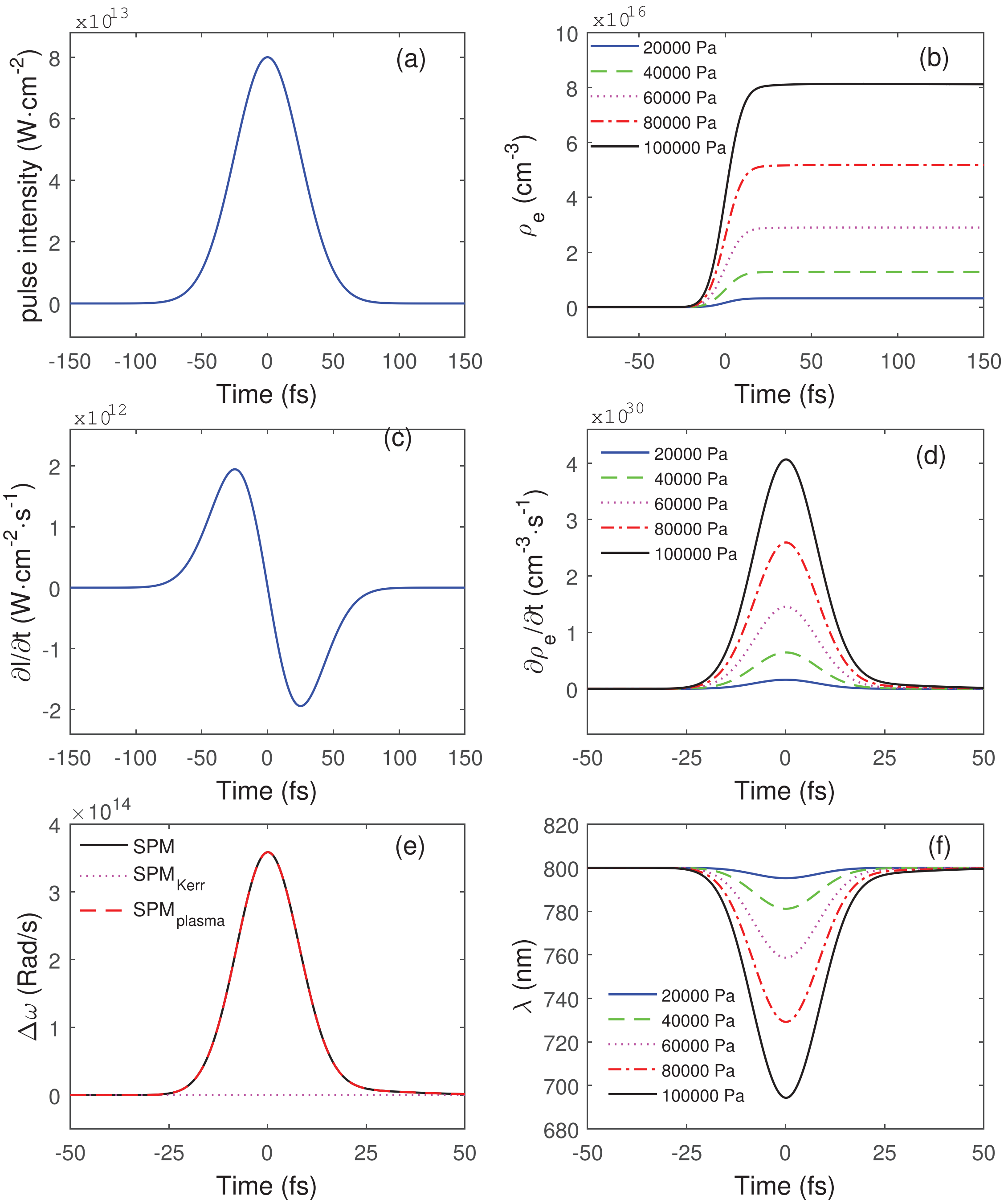

FIG. 3. Time-evolution of (a) Gaussian pulse intensity and (b) plasma density (c) $\partial I/\partial t$ and (d) $\partial \rho_e/\partial t$ at different pressures. (e) Change of frequency caused by SPM induced by the Kerr effect only, plasma only, and both of them at 1 atm. (f) Spectral broadening at different pressures.

similar tendency in Fig. 4: with increasing pulse energy, the spectral intensity is enhanced while the spectral broadening is not obvious. Since the intensity in a filament is clamped no matter how much the pulse energy changes. As a result, the second term at the right-hand side of Eq. (1) representing the contribution of SPM induced by plasma generation varies little with the increasing pulse energy at the same pressure. In our work, according to Eq. (1), the filament length can only affect the magnification of the contribution to SPM, which means that it has the same influence on the SPM induced by plasma generation and Kerr effect. The increasing pulse energy surely extends

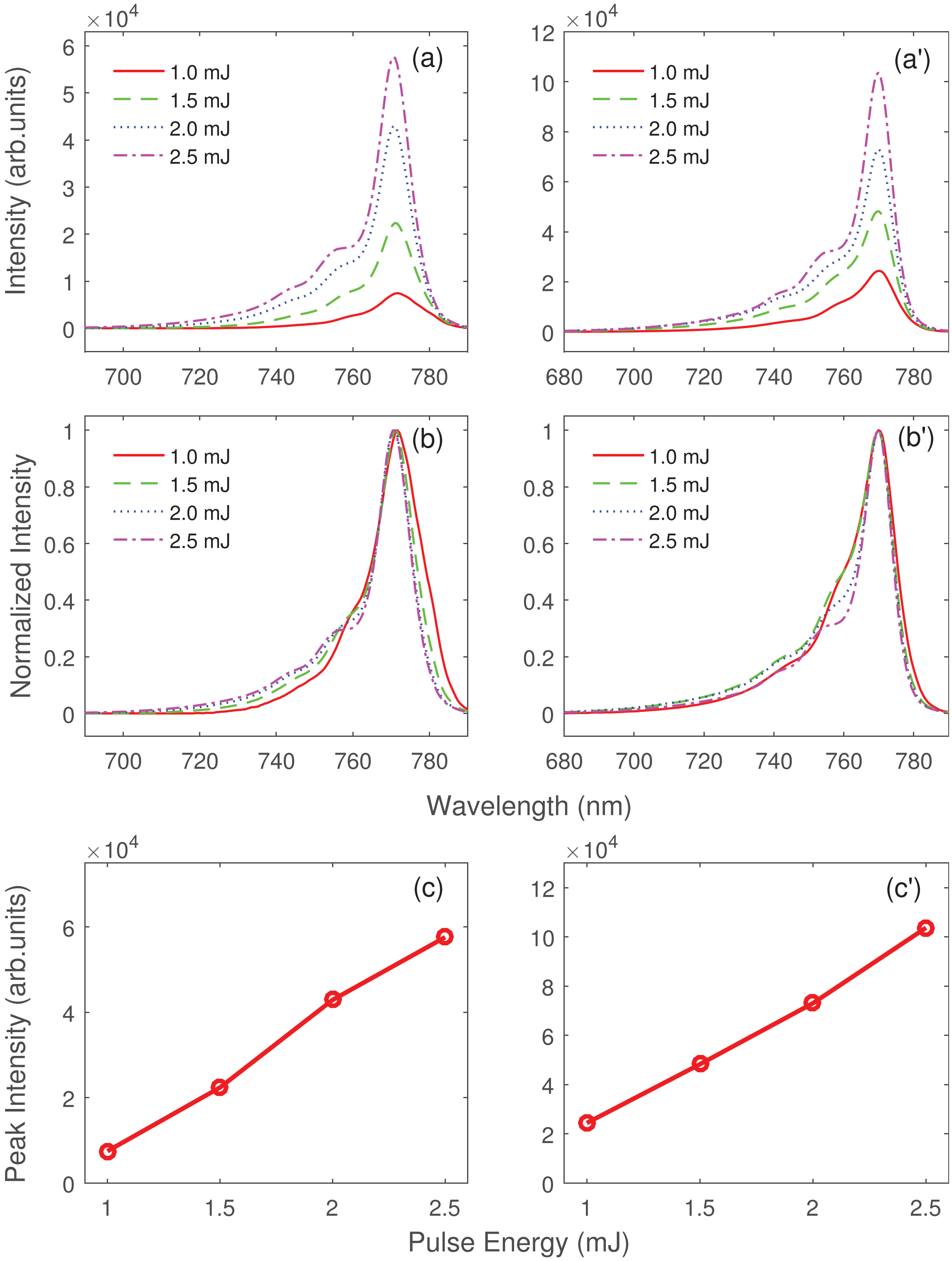

FIG. 4. (a) Spectra measured at 40 000 Pa and (b) normalized spectra measured at 40 000 Pa under different pulse energies, and (c) variation of the peak spectral intensity with pulse energy. (a′) Spectra and (b′) normalized spectra under different pulse energies at a fixed pressure of 100 000 Pa, and (c′) variation of the peak spectral intensity with pulse energy.

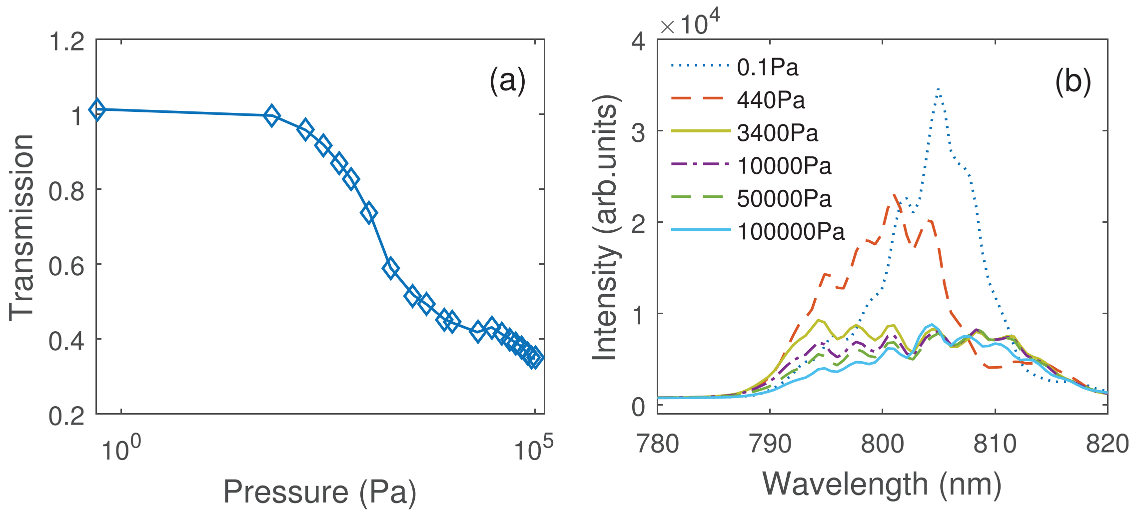

FIG. 5. (a) Variation of the transmission (blue diamond line) for 800 nm with pressure when the incident laser energy is 2.5 mJ. (b) Spectra measured at different pressures after the transmitted laser energy is detected.

the filament length, however, the change of the pulse energy in our work is so small that the filament length will not change dramatically which can be seen in Figs. 2(d) and supplementary material, Figs. S1(g) and S1(h). It should be noted that higher order perturbations may affect the spectral broadening with increasing the laser energy such as Raman instability. The first Stokes shift due to the rotational Raman effect from N_2 (12 cm^{-1}) and O_2 (8.4 cm^{-1}) corresponds approximately to a wavelength shift of 1.5 nm and 1.1 nm, respectively. It is too hard to tell, because the spectral bandwidth of the newly generated IR pulse is much wider than that of the first Stokes shift due to rotational Raman effects from N_2 and O_2. In our work, we focus the blue shift of the spectrum and may neglect the influence of Raman instability.[43,44] As a result, the spectral broadening is not obvious when the pulse energy increases.

By contrasting Figs. 2(c), 4(b), and 4(b′), we can see that the normalized spectra above 780 nm do not overlap with each other. It can be attributed to the fact that for the wavelength larger than 780 nm, the transmission of shortpass filter decreases to zero gradually, therefore the spectra whose wavelength is larger than 780 nm cannot be filtered completely, as a result, they look more like slopes rather than steep edges, and do not overlap with each other after normalization.

We also measure the transmitted laser energy after the propagation in the gas chamber to investigate the relationship between the transmission of laser pulse and the pressure. First, we replace the shortpass filter to the narrow band filter, whose central wavelength and bandwidth are 800 and 40 nm corresponding to the central wavelength and the linewidth of the laser used in our experiment. Then, we insert an energy meter between the filter and the integrating sphere to measure the laser energy after propagation in the gas chamber. Subsequently, we move out the energy meter and collect the spectra which are shown in Fig. 5(b). It is clear to see in Fig. 5(a) that the transmission decreases with the increasing pressure. When the pressure increases, the density of the neutral molecule increases which means more pulse energy is absorbed to ionize the molecules in air. Besides the MPI effect, there are other phenomena accompanying the filament which can consume the incident laser energy, such as fluorescence, supercontinuum, terahertz generation, high harmonic generation, and acoustic wave generation. It is depicted in Fig. 5(b) that the spectral intensity decreases with pressure, which means the pulse energy transforms to other forms corresponding the results shown in Fig. 5(a).

IV. CONCLUSION

In this paper, we investigate the influence of pressure on the spectral intensity and blue shift of spectrum during the plasma generation induced by femtosecond laser pulses experimentally and theoretically. We find that increasing pressure can enhance spectral intensity and blue shift of spectrum at the same time. With the aid of numerical simulation, we analyze the relationship between spectral broadening and SPM induced by plasma generation and Kerr effect, and find that the blue shift of the spectrum generated by the SPM induced by plasma generation increases with pressure. The experimental and simulated results have a similar tendency and prove that SPM induced by the plasma effect is an important factor of blue shift of the spectrum. In addition, the influence of variation of pulse energy on spectra is also discussed. Increasing the incident pulse energy can strengthen the spectral intensity; however, it has little influence on blue shift of spectrum due to the intensity clamping. Besides, we analyze the transmission of the incident laser energy and find that the transmission decreases with pressure, which can be attributed to the fact that the plasma density increases with pressure and make other phenomenon accompanying the filament more violent which can consume the incident laser energy such as fluorescence, supercontinuum, terahertz generation, high harmonic generation, and acoustic wave. The results may contribute to the practical uses of controllable white light.

SUPPLEMENTARY MATERIAL

See the supplementary material for the complete simulated results of the change in frequency caused by the SPM and the pictures of the luminous region at different pressures.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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