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Investigation of blueshift of supercontinuum from femtosecond flament plasma grating

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Abstract

A blueshift in frequency spectrum is observed in the supercontinuum radiation from a plasma grating, which is formed by the laser feld interference in the overlap region of two noncollinear femtosecond plasma flaments. These two pump laser pulses show diferent profles in their supercontinuum radiation. Both the energy exchange between two pump laser pulses and the blueshift are caused by the plasma grating within a 200-fs relative time delay between the pump laser pulses. Thus, these two processes are induced by the propagation of the laser pulses in the plasma. In addition, this blueshift of the supercontinuum from one flament is enhanced when the other flament increases its pump laser energy. The possible mechanism of this blueshift is also discussed. This ofers a method of broadening and tailoring the supercontinuum radiation from the femtosecond plasma flament.

1 Introduction

Filamentation is a nonlinear ultrafast phenomenon that is induced by the propagation of high-peak-power ultrashort laser pulses in optical transparent media, such as gases, crystals, and liquids $[1–5]$ $[1–5]$ $[1–5]$. The peak-power of the laser pulse must be higher than a critical power $P_{cr} (P_{cr} = \frac{\lambda^2}{4\pi n_0 n_2}, \lambda$ is the wavelength of the laser, n_0 is the refractive index of the medium, and $n₂$ is the nonlinear refractive index of the medium) to generate a flament. During this ultrafast process, a medium with a nonlinear Kerr coefficient n_2 induced by a strong laser beam through the Kerr effect makes itself act as a focusing lens, causing the laser beam to self-focusing. As a result, the laser feld becomes strong enough to ionize the atoms in the medium. Then, the free electrons ionized from the atoms form a plasma which contributes a defocusing effect to the laser beam. The effects of the plasma defocusing and the Kerr self-focusing on the laser pulse propagation balance against each other, causing the laser beam accompanied by a plasma core to propagate a longer distance than the typical Rayleigh length without the help of

any external mechanism. During this process, there are other efects associated with the Kerr self-focusing and the plasma defocusing, including optical field ionization, plasma absorption, self-phase modulation, four-wave mixing, and Raman effects.

After its experimental discovery, the flamentation has been shown to have many applications in areas such as terahertz (THz) wave generation [[6](#page-4-2)[–8](#page-4-3)], extreme ultraviolet (XUV) generation $[9]$ $[9]$, remote sensing $[10]$ $[10]$, pulse compression [[11,](#page-4-6) [12\]](#page-4-7), supercontinuum radiation, ultrafast gaseous "half-wave plate" $[13]$ $[13]$ $[13]$, and lightning control $[14]$ $[14]$. For all these applications, the control, manipulation, and optimization of the flamentation characteristics are becoming increasingly important. And, the interaction between two filaments might offer a simply way to change or control the characteristics of the flamentation. Stelmaszczyk et al. have experimentally investigated the interaction between the flaments in the fused silica, and found that there is a change in the radiation pattern of the supercontinuum [\[15\]](#page-4-10). Berge et al. have studied the interaction between the flaments with diferent pump power, and found that there are three distinct evolution regimes according to the pump power [\[16\]](#page-4-11). The colliding flaments or multiflament also can coalesce into one central lobe [\[16](#page-4-11), [17\]](#page-4-12).

When two flaments intersect at an angle in a gas or in air atmosphere, the laser feld in the overlap region of the plasma produces a grating [[18–](#page-4-13)[20\]](#page-4-14). This grating is induced by the interference of these two laser beams through the

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plasma in space. It changes properties of these two flaments, such as the energy exchange [\[20–](#page-4-14)[22](#page-4-15)], THz radiation [\[23](#page-4-16)], and the third-harmonic generation enhancement $[24]$ $[24]$. Thus, this plasma grating offers a simple way to change the propagation and the formation of the flaments. Here, we experimentally investigated the supercontinuum radiation generated from the flament-induced plasma grating, found that the supercontinuum radiations from these two flaments have diferent spectrum, and observed a blueshift in supercontinuum associated with the exchange of energy between the pump laser pulses. And the results show that the relative time delays for the blueshift process and the energy exchange process are similar. The mechanisms behind these phenomena are also discussed in this paper.

2 Experimental results and discussion

The experimental schematic used in our study is similar to that used by Bernstein et al. [[20\]](#page-4-14). In the experiment, a commercial Ti:sapphire amplifer laser system produces 35-fs, horizontally polarized laser pulses with a central wavelength of 800 nm, a 1-kHz repetition rate, and a pulse energy of 3 mJ. The laser beam is split into two beams (beam 1 and beam 2) with a 40/60 beam splitter. Then, these two beams are focused to produce flaments by two achromatic lenses, each with a focal length of $F = 15$ cm. The crossing angle *θ* between these two flaments is 25°. A motorized stage in beam 2 is used to control the time delay between the laser pulses. A laser power meter and a fber spectrometer (Ocean Optics, USB4000) are used to measure the laser pulse energy and the spectra from the flaments, respectively. The whole system is set in the air atmosphere. In the actual measurement, beam 1 has a pulse energy of 1.36 mJ, and beam 2 has a pulse energy of 0.9 mJ.

Figure [1](#page-1-0) shows the exchange of energy between the laser pulses induced by the plasma grating. Note that beam 1 and beam 2 have diferent ordinate labels. It has been reported that the frequency diference between the laser pulses causes the energy exchange, and this diference dictates the direction of energy flow $[21]$ $[21]$ $[21]$. Although the two pump laser pulses have the same carrier frequency and identical chirp before the plasma grating, the nonlinear response of the plasma formation will result in diferent phase modulations when there is a relative time delay between the two pump laser pulses [[22](#page-4-15)]. This frequency diference of laser pulses during the propagation in the flaments causes the laser energy transfer from one pump beam to the other. The zero time delay between the two pump laser pulses is also obtained according to this energy exchange process.

After measuring the laser pulse energies, we measured the frequency spectra of pump laser beam after the

Fig. 1 The exchange of energy between the two pump laser pulses induced by the plasma grating

flamentation. By changing the time delay between these two pump laser pulses, the temporal evolution of the spectrum from supercontinuum is obtained. The time delay, including the zero time delay, is the same to that used in Fig. [1.](#page-1-0) Figure [2](#page-2-0)a shows the temporal evolution of the spectra from the beam-1 flament, and Fig. [2](#page-2-0)b presents several distinct spectra with diferent time delays between two pump laser pulses. Note that Fig. [2](#page-2-0)b also shows a supercontinuum (black line) obtained from a single flament (in the absence of beam 2) to show a comparison. It is obvious that the spectrum have become narrower around the zero time delay. When the time delay is far from zero, the spectra are same to that emitted from single flament. Figure [2c](#page-2-0), d are the corresponding results from the beam-2 flament. Similarly, a supercontinuum (black line) from a single flament is also given in Fig. [2d](#page-2-0) as a comparison. It is obvious that the spectra are diferent for beam-1 flament and beam-2 flament. In the beam-2 flament, the supercontinuum has a strong blueshift at around the zero time delay. This blueshift is only produced in the plasma grating, but disperses in the single flament. Thus, this ultrafast plasma grating generates the strong blueshift of the supercontinuum radiation from beam-2 flament.

After the plasma grating is formed, the electron density will decrease slowly because of electron–ion recombination and electron difusion. The plasma entirely decays away in the air atmosphere over several tens nanosecond [[1\]](#page-4-0). It can be seen that the spectrum evolution occurs over about 200 fs, which corresponds to the time of the propagation of the laser pulses through the plasma grating. Depending on the pump power and the wavelength of the laser, the diameter of the plasma usually varies from several 10 μm to several 100 μm. For a typical flament generated by a femtosecond pulse with an energy of 1 mJ, its plasma core diameter is around 100 μ m [\[1](#page-4-0)]. The refractive

Fig. 2 a The temporal evolution of the spectra from the beam-1 flament, and **b** its several supercontinuum spectra with diferent time delays between two pump pulses. The black line in **b** shows

the supercontinuum in the absence of beam 2 as a comparison. **c**, **d** Respectively, the corresponding results from the beam-2 flament

index in the plasma is $n = n_0$ $\sqrt{1-\frac{\omega_{\rm p}^2}{\omega^2}}$, where $\omega_{\rm p} = \sqrt{\rho_{\rm e}e^2/m_{\rm e}\epsilon_0}$ is the plasma angular frequency, ω is the laser angular frequency, ρ_e is the plasma electron density, ε_0 is the vacuum permittivity, and *e* and m_e are the electron charge and mass, respectively. When the ratio is very small, $\frac{\omega_p}{\omega} \ll 1$, the index can be estimated from the expression $n \approx n_0(1 - \frac{\omega_p^2}{2\omega^2}) = n_0(1 - \frac{\rho_e}{2\rho_c})$ based on the firstorder series expansion. Here, $\rho_c = \frac{m_e \omega^2 \epsilon_0}{e^2}$ is the plasma critical density (which is 1.7×10^{21} /cm³ for 800-nm laser pulses). The plasma electron density in an air flament is estimated to be around 10^{19} /cm³ [[19,](#page-4-19) [25\]](#page-4-20). In air, n_0 is 1.0002905 [[26\]](#page-4-21). Thus, the refractive index *n* in the plasma is estimated to be 0.997349. Therefore, the time that the laser pulses propagate in the plasma grating is around 333 fs. In actuality, the diameter of the plasma core, 100 μm, is not a precise value, so this time for the laser propagation in the plasma grating is not a precise value. However, the times for (1) the propagation of laser pulses in the plasma grating and (2) energy exchange between the laser pulses are nearly in the same time scale. Thus, the coupling of these two pump pulses in the plasma induces

the energy exchange and the blueshift of supercontinuum simultaneously.

The density distribution of electrons in the flament is shaped like a long elliptical cylinder, which means that the electrons is denser in the middle, while a small electron density exists in the front and in the tail. During the experiment, we adjusted the beam-2 flament and made its front or tail cross the middle of the beam-1 flament. Then we performed the measurements again. The phenomena shown in Fig. [2](#page-2-0) became severely degraded. Thus, the electron density plays a key role in the blueshift of the supercontinuum from the beam-2 flament. The typical lifetime of the free electrons in the flament is on the order of several tens nanoseconds [\[1](#page-4-0)], while the range of time delays for the energy exchange and the blueshift processes are within about 200 fs (as shown in Figs. [1](#page-1-0)b, [2](#page-2-0)a, c). It has been experimentally reported that the plasma grating is mainly formed by the interference between the laser pulses in the plasma [\[27](#page-4-22)]. The blueshift of spectrum is also caused by the propagation of the second laser pulse in the plasma grating. After the second laser pulse propagates off the plasma, the energy exchange and the blueshift disappear.

When the pulse energy of beam 1 varies, its infuence on the blueshift of supercontinuum of beam-2 flament varies.

Fig. 3 a Several supercontinuum spectra from beam-2 flament pumped by beam 1 with diferent pulse energies. **b** Several supercontinuum spectra from a single flament with diferent laser pulse energies

Figure [3a](#page-3-0) shows several supercontinuum spectra from the beam-2 flament resulting from the plasma grating where beam 1 has diferent pulse energies. During the measurement, the pulse energy of beam 2 remains stable at 0.9 mJ, while only the pulse energy of beam 1 is changed by a neutral attenuator. Then, we obtained the spectra with the largest bandwidth by changing the time delay between the two pump laser pulses. As shown in Fig. [3a](#page-3-0), the spectra become broader and have wider plateaus in the high-frequency regime when the energy of beam 1 increases. The energy increase of beam 1 increases the length and the electron density of its plasma core, and as a result, the nonlinear efect for the blueshift of supercontinuum of beam-1 flament becomes stronger. This relation is similar to that between the energy exchange and the pump energy, as reported previously [[21](#page-4-18), [22](#page-4-15)]. Figure [3](#page-3-0)b gives several supercontinuum spectra from a single flament with diferent pump energies. During the process of plasma flament generated by the femtosecond laser pulses, the self-steeping and the space–time focusing afect the profle of pump laser pulses, and then the self-phase modulation generates the supercontinuum radiation [[2\]](#page-4-23). As shown in Fig. [3](#page-3-0)b, the spectra of supercontinuum become broader when the pump energy increases. However, the spectrum (black) in Fig. [3](#page-3-0)a with the lowest pump energy is much broader than that (red) of the single flament with the highest pump energy in Fig. [3b](#page-3-0). Thus, the plasma grating can broaden the bandwidth of the supercontinuum of the flament. Although only one crossing angle between these two flaments is used in our experiment, it is possible to produce similar phenomena with diferent angles. Here, the fber spectrometer limits the measured data in the high-frequency regime, but the trend shows that the supercontinuum is extended greatly to the high-frequency regime by the plasma grating.

The supercontinuum radiation of the ultrafast flament is generated mainly by the self-phase modulation (SPM) of laser pulses [\[28](#page-4-24)]. However, for the plasma grating, both selfphase modulation (SPM) and cross-phase modulation (XPM) contribute to the supercontinuum radiation [[22](#page-4-15)]. Thus, during the two-beam coupling, the frequency shift of one beam induced by both SPM and XPM is given by $\frac{\Delta \omega_{\text{1SPM+XPM}}}{\Delta \Omega} = 1 + 2 \frac{|E_2|^2}{\ln^2}$ $\Delta \omega_{\rm 1SPM}$ $\frac{|P_2|}{|E_1|^2}$ [[29\]](#page-4-25), where E_1 and E_2 are the laser

amplitudes of the two beams, respectively. The occurrence of the observed blueshift in the plasma grating is the result of these two mechanisms. Here, beam 2 is weaker than beam 1. Thus, the contribution of SPM and XPM to the frequency shift of beam 2 is more obvious than the corresponding contribution to the frequency shift of beam 1. In addition, beam 2 obtains energy from beam 1 initially, and thus its blueshift is enhanced. At the same time, beam 1 loses energy during the energy exchange, which makes its supercontinuum a little narrower. Then, beam 2 loses energy for the same reason, and this weakens its blueshift, while beam 1 obtains the energy that allows its supercontinuum to recover, as before. The second possible reason is that the coupling of the two flaments increases the electron density in the region of overlap between the flaments [\[30](#page-4-26)]. Therefore, the increasing of the electrons density enhances the nonlinear interaction between the laser pulses and the plasma. As a result, the supercontinuum radiation from the plasma grating has a strong blueshift during this process, as shown in Fig. [3.](#page-3-0) Although this is just a phenomenological explanation, our experimental results show that the shapes of the supercontinuum of the flaments have been changed, as reported that in the fused silica $[15]$ $[15]$ $[15]$. Thus, this might offer a method to tailor the supercontinuum radiation from the flament by the interaction of flament-flament.

3 Conclusions

In summary, we have experimentally investigated the energy exchange between two pump laser pulses and the supercontinuum blueshift in a plasma grating, which is produced by two noncollinear femtosecond flaments with a crossing

angle of 25°. It is found that both the energy exchange and the blueshift happen within a 200-fs relative time delay between the two pump laser pulses. The blueshift of the supercontinuum is also a function of the pump energy of the other flament. When the pump energy increases, the supercontinuum blueshift becomes fatter in the high-frequency regime. The blueshift of the supercontinuum might be caused by the contribution of the SPM and the XPM of the laser pulses. The other reason might be that the increased electron density in the plasma grating also enhances the interaction between the laser pulse and the plasma, and consequently, induces the blueshift of the supercontinuum radiation. It is hoped that such blueshift can be used to improve the bandwidth of the supercontinuum generation from the flament and the self-compression of laser pulse in the flament.

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