Energy transmittance of focused femtosecond pulses at different air pressures*

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Transmission of intense femtosecond laser pulses in air is accompanied by energy depletion. By measuring the transmitted spectra of the focused femtosecond pulses in air, we study the influence of air pressure and initial pulse energy on the spectra around the central wavelength (800 nm) after the interaction of the focused femtosecond laser with air. On this basis, the energy transmittance of the central wavelength of the femtosecond pulses is obtained. It is found that as the air pressure is lower than 1 kPa, the transmitted spectra of focused femtosecond pulses change with the pressure, but there is almost no energy depletion, while as the air pressure is higher than 1 kPa, femtosecond filamentation occurs and the energy transmittance of the central wavelength of the femtosecond pulses decreases with the increase of air pressure and pulse energy. According to the different regimes (i.e., nonfilamentation, and filamentation regimes), we discuss the effect of energy conversion and transfer on the energy transmittance. This work can help to understand the energy depletion during the transmission of ultrashort intense laser pulses in air and provide a guidance for the practical applications of femtosecond filamentation.

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When the femtosecond laser propagates in ambient air, its intensity becomes increasingly high due to the Kerr self-focusing/linear focusing (external focusing lens) effect, and eventually reaches the ionization threshold of air molecules, producing numerous plasmas which play a defocusing role on the laser beam. As the Kerr self-focusing and plasma defocusing effects reach a dynamic balance, a stable plasma channel is formed, which is known as the femtosecond filamentation[1]. The balance makes the laser intensity inside the femtosecond filament clamped to about 10^{13}—10^{14} W/cm^2, and maintains a long distance along the propagation direction, which can even reach thousands of meters[2,3]. The high intensity inside the filament can induce the excitation, ionization, dissociation of molecules, clusters, aerosols and other matters along the filament path, resulting in the characteristic fluorescence emission[4,5]. The plasma density inside femtosecond filament is around 10^{16} cm^{-3} and these plasmas endow the femtosecond filament electrical conductivity, which can trigger and guide electric discharges[6,7]. Due to these specific properties, femtosecond filamentation can find promising application prospects in many fields, such as rainmaking[8,9], lightning protection[6,7], material processing[10,11], light detection and ranging[12], and so on. These applications rely on the long-distance propagation of the intense femtosecond pulses. Unfortunately, energy depletion is involved in the femtosecond filamentation, bringing about difficulties to these applications. A lot of effects give rise to energy depletion in the femtosecond filamentation process, such as the excitation and ionization of molecules in air, absorption of plasmas, electromagnetic radiation such as fluorescence and terahertz, and acoustic and thermal radiation, etc. Although linear effects like the diffraction, refraction and scattering also cause energy depletion, their influence is ignorable compared to the nonlinear ones. To realize the long-distance transmission of intense femtosecond pulses, reducing the energy depletion in the transmission process has been one of research hotspots.

To produce longer filaments, researchers have conducted numerous arduous explorations. Basically, it can be realized by adjusting the laser parameters (e.g., pulse duration, beam waist, initial power, and polarization state, etc), whereas, the effect is limited. Some technological

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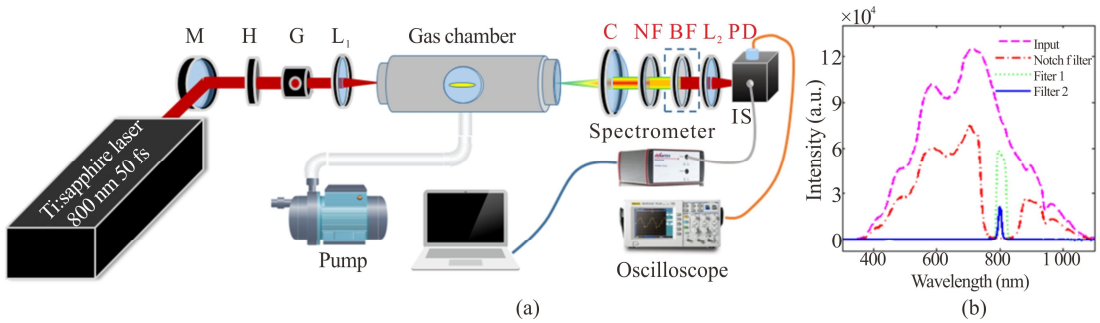
approaches like the external refuellment[13], double- or multiple-pulse technique[14,15], applying a high-voltage electric field[16], and introducing initial chirp[17] are developed to control the length, lifetime and shape of femtosecond filaments. The length of plasma channel and the value of the clamped intensity inside the filament affect the energy depletion of the femtosecond pulses. Air pressure is another key factor that affects the femtosecond filamentation as well as the accompanied dynamical nonlinear processes[18-24]. However, when the air pressure is lower than some value (usually 20 kPa), for freely propagating femtosecond pulses, femtosecond filamentation does not occur[19] (femtosecond filamentation can be initiated at lower air pressures when an external focusing lens is introduced[21]). Under this circumstance, the interaction between the laser pulse and air is weak, and little energy is depleted. With the increase of air pressure, the number of the gas molecules interacting with the laser pulses increases, and the collision is enhanced accordingly, resulting in the increase of plasma density and more energy depletion of the laser pulses. Consequently, the residual energy of the femtosecond pulses decreases. Since the pulse energy concentrates upon the central wavelength, in most fundamental researches and practical applications (e.g., atomic and molecular ionization[5], high-order harmonic generation[25], micromachining[26], etc), the researchers mainly focus on the central wavelength components of the driving laser. For this reason, the energy transmittance of the central wavelength of the femtosecond pulses in transparent media should be studied, whereas, few studies focus on this issue.

In this paper, by measuring the transmitted spectra of the femtosecond pulses around the central wavelength, we study the effect of air pressure and pulse energy on the energy transmittance of the central wavelength of the femtosecond pulses, and attempt to have a deeper understanding of the energy conversion and transfer effect during the transmission of intense ultrashort pulses in ambient air, which may pave the way for the practical applications of femtosecond filametation, like remote sensing, and optical communication in free space.

The experiment is carried out by using a regenerative amplified Ti: sapphire laser system (Cohernent Libra), which generates the femtosecond pulses with a central wavelength of 800 nm and a duration of 50 fs at a repetition rate of 1 kHz. Fig.1(a) shows the schematic diagram of the experimental setup. The pulse energy is adjusted by an energy attenuator composed of a half-wave plate (H) and a Glan prism (G). A focusing lens (L_1) (f=400 mm) is used to focus the femtosecond pulses in a cylindrical gas chamber whose length and diameter are 750 mm and 100 mm. To overview the air pressure effect on the energy transmittance of the central wavelength of the femtosecond pulses, the experiment is conducted from the vacuum to one atmospheric pressure. The pressure inside the gas chamber is adjusted by a mechanical pump, and its value ranges from 0.1 Pa to 100 kPa. A capacitance diaphragm gauge linked to the gas chamber is used to monitor the air pressure and a digital panel connected to the gauge displays the value of the air pressure. The optical signals emitted from the chamber outlet are collimated by a convex lens (C) (f=400 mm), then pass through a notch filter (NF) (740—865 nm), a band-pass filter (BF) (two kinds of BFs are used in the experiment: BF_1: 790—820 nm, self-manufactured; BF_2: 795—805 nm, Thorlabs FB800-10), and converged into an integrating sphere (IS) by a focusing lens (L_2) (f=100 mm). The dashed box in the figure suggests the BF can be replaced or removed. In Fig.1(b), the dashed pink curve shows the spectra of the light emitted by a tungsten lamp, and the dash-dotted red the dotted green, and solid blue curves denote the spectra measured when the NF, BF1 and BF2 are used, respectively. It should be noted that the NF has two functions in the experiment: one is to attenuate the laser energy to avoid damage to optical elements such as the BF and IS, and the other one is to detect spectral signals whose wavelength is lower than 740 nm or higher than 865 nm. The optical signals in the IS are guided to a spectrometer (Avantes-avaspec-uls2048l) through an optical fiber to obtain the transmitted spectrum of the femtosecond pulses. A silicon photodiode (PD) (Thorlabs SM05PD1B) which converts the received optical signals into the electrical ones connected to the IS, and the electrical signals are guided to an oscilloscope through an electrical wire. The integration time of the spectrometer is set as 5 ms. To reduce the error, each group of data is measured 200 times and averaged.



Laser: femtosecond laser; M: mirror; H: half-wave plate; G: Glan prism; L_1, L_2: focusing lenses; C: collimation lens; NF: notch filter; BF: band-pass filter; IS: integrating sphere; PD: silicon photodiode

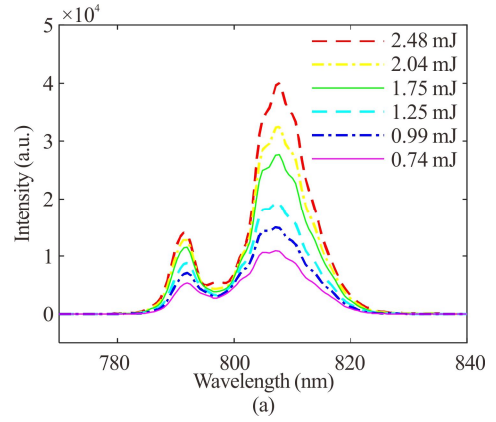
Fig.1 (a) Schematic diagram of the experimental setup to measure the transmitted spectra of the focused femtosecond pulses; (b) Spectra of the light emitted by a tungsten lamp measured when the NF, BF_1 and BF_2 are used

Fig.2 shows the transmitted spectrum of the focused femtosecond pulses with different energies at 0.1 Pa (vacuum) and 100 kPa (1 atmospheric pressure). In our work, six sets of pulse energies are chosen, i.e., 0.74 mJ, 0.99 mJ, 1.25 mJ, 1.75 mJ, 2.04 mJ and 2.48 mJ. It can be seen that the higher the pulse energy, the stronger the transmitted spectrum is. The NF can filter out the spectral signals in the range from 740 nm to 865 nm, and its extinction index around 800 nm is about 98%, while the spectrum of the incident laser pulse ranges from 770 nm to 820 nm, which is within the action range of the NF. The transmitted spectrum measured at 0.1 Pa when only using NF is the spectrum of the focused laser pulse after it passes through the gas chamber, as shown in Fig.2(e). Under this circumstance, the NF attenuates the pulse energy. Though the air in the gas chamber is thin, when the intense laser pulse interacts with it, the laser spectrum also changes with the increase of air pressure. The variation of the spectral broadening induced by the self-phase modulation (SPM) can be described by[1,18]

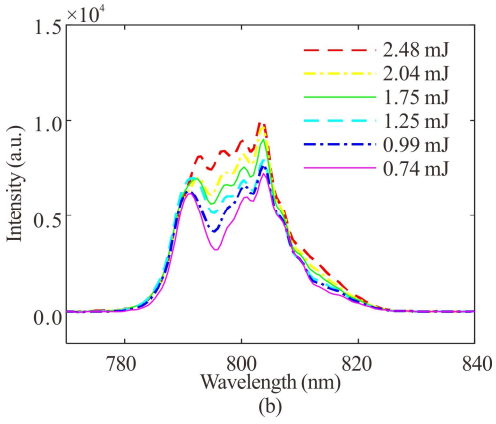
$$\Delta\omega = \frac{\omega_0}{c}\left(-n_2\frac{\partial I(r,t)}{\partial t}+\frac{1}{2n_0\rho_c}\frac{\partial\rho_e(r,t)}{\partial t}\right), \tag{1}$$

where ω_0, z and n_2 are the central wavelength of the incident femtosecond pulse, propagation distance in media and second-order nonlinear refractive index, respectively. $\rho_c=\varepsilon_0 m_e\omega_0^2/e^2$ denotes the plasma critical density, ε_0, e, $\rho_e(r, t)$ and m_e are permittivity of vacuum, electron charge, electron density and electron mass, respectively. The laser spectrum is greatly broadened due to the intense SPM effect, and consequently the spectral components whose wavelength is lower than 740 nm can be observed, as shown in Fig.2(f). Under the combined action of the NF and BF, the change of the spectra around the central wavelength (800 nm) can be observed, as shown in Fig.2(a)—(d).

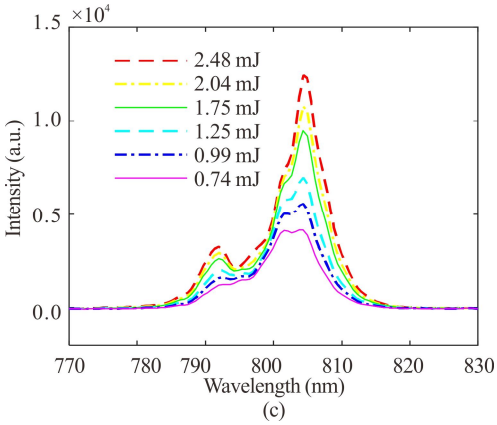
The change of air pressure directly affects the number density of molecules and the collision probability, thereby affecting the dynamic processes occurring during the femtosecond filamentation, such as the molecular excitation and ionization, plasma absorption[21,23,24]. These dynamic processes affect the electromagnetic radiation like fluorescence, terahertz, and the thermal and acoustic radiation. As a result, the transmitted spectra after the femtosecond filamentation are inevitably affected by the change of air pressure. For example, in our previous work, we systematically investigated femtosecond laser-induced nitrogen fluorescence emission at different air pressures, and found that the N_2^+ and N_2 fluorescence signals show opposite variation in intensity as a function of air pressure[21]. Fig.3 shows the transmitted spectra of the focused femtosecond pulses with energy of 2.48 mJ and 0.74 mJ at different air pressures. The intensity of the transmitted spectrum decreases with increasing air pressure, as shown in Fig.3(a) and (b). Fig.3(e) and (f) show that with the increase of air pressure, the intensity of the spectrum at the red side of the central wavelength becomes stronger, indicating that the spectral broadening is enhanced, which is consistent with our previous work[18]. We can also see that the spectrum around the central wavelength seems to undergo a blue shift: at 0.1 Pa, the maximum of the spectrum appears at 810 nm, while it moves gradually to 800 nm with the increase of air pressure, as shown in Fig.3(a) and (b). We speculate that the SPM leads to the change of laser spectrum, which is obvious when the air pressure ranges from 0.1 Pa to 1 kPa, as shown in Fig.4(a). As the air pressure is lower than 1 kPa, the maximum intensity of laser spectrum shifts from 810 nm towards 800 nm. Since BF_2 works in the range from 795 nm to 805 nm, which is narrower than the linewidth of the laser pulse, and consequently parts of the laser spectra are filtered out by it, as shown in Fig.2(c) and (d), Fig.3(c) and (d), Fig.4(c) and (d). In this paper, the data obtained when using BF_2 are taken as a reference.



(a)



(b)



(c)

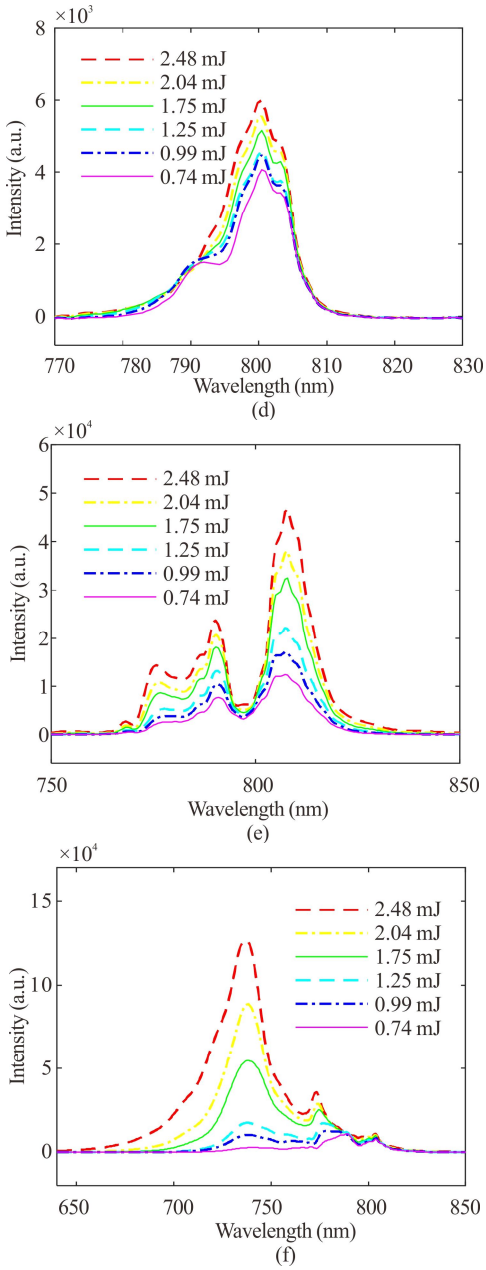
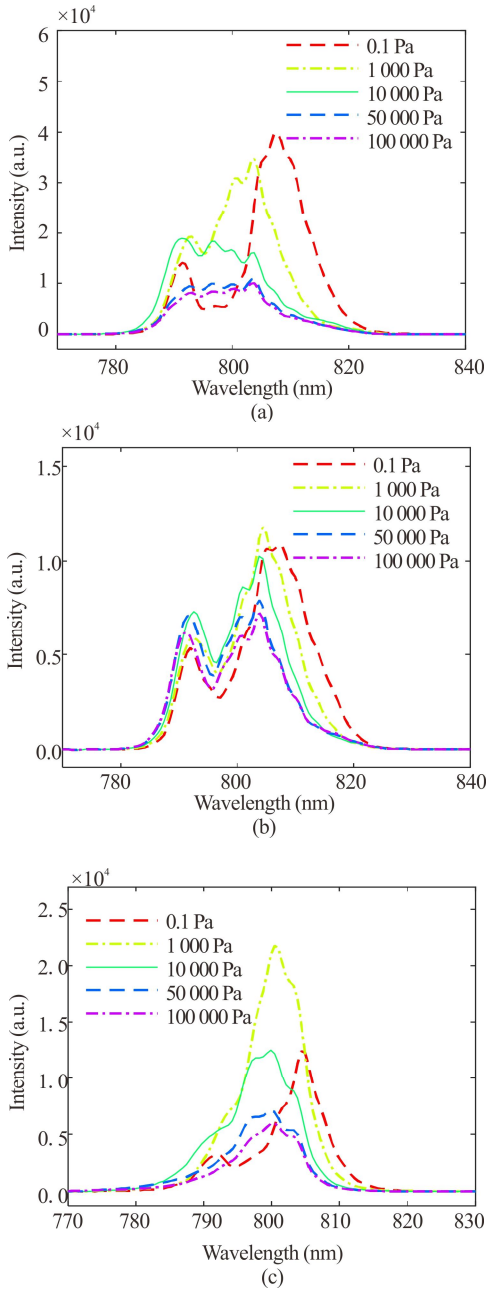


Fig.2 Transmitted spectra of the focused femtosecond pulses with different initial energies at (a, c, e) 0.1 Pa and (b, d, f) 100 kPa when (a, b) BF_1 (790—820 nm) + NF (740—865 nm), (c, d) BF_2 (795—805 nm) + NF and (e, f) NF are used

The decrease in the intensity of transmitted spectrum means that the energy of transmitted laser pulse decreases, undergoing energy depletion. Nonlinear effects in the femtosecond filamentation process are the main factors that lead to reduction in transmitted energy of the femtosecond pulses. We can divide them into two categories: (1) molecular excitation and ionization, plasma absorption, fluorescence, terahertz and other electromagnetic radiation, acoustic and thermal radiation, etc consume the pulse energy; (2) SPM leads to spectral broadening[1,18], transferring the energy of the central wavelength to other wavelengths. Among them, the former converts pulse energy into other kind of energy (for example, intramolecular energy and mechanical energy, etc), which consume the pulse energy, and the latter is an energy transfer process, which redistributes the energy of the laser pulse over the wavelength. Besides, the latter is a nonlinear optical parametric process, which does not change the coherence of the femtosecond pulses, while the former brings incoherent components to the transmitted spectrum. To study the energy conversion and transfer in the interaction of the focused femtosecond laser with air, we can quantitatively analyze the energy loss of the femtosecond pulses by measuring the energy transmittance of the central wavelength of the focused femtosecond pulses. In this work, the energy transmittance is measured by the following two methods simultaneously.



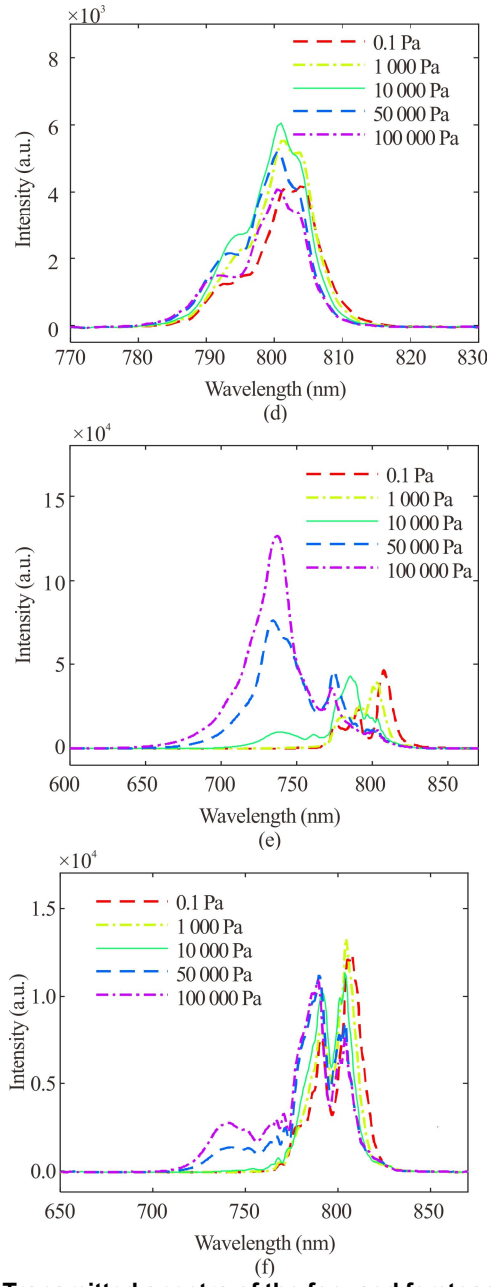
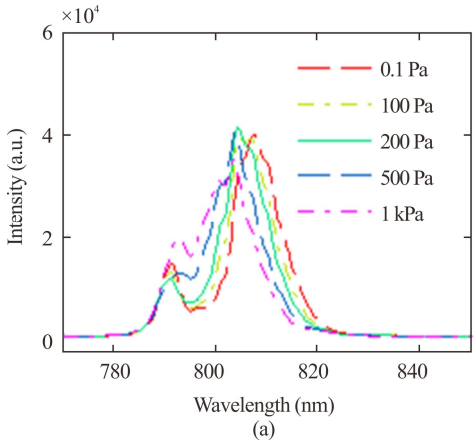


Fig.3 Transmitted spectra of the focused femtosecond pulses whose energy is (a, c, e) 2.48 mJ and (b, d, f) 0.74 mJ at different air pressures measured when (a, b) BF_1 + NF, (c, d) BF_2 + NF, and (e, f) NF are used



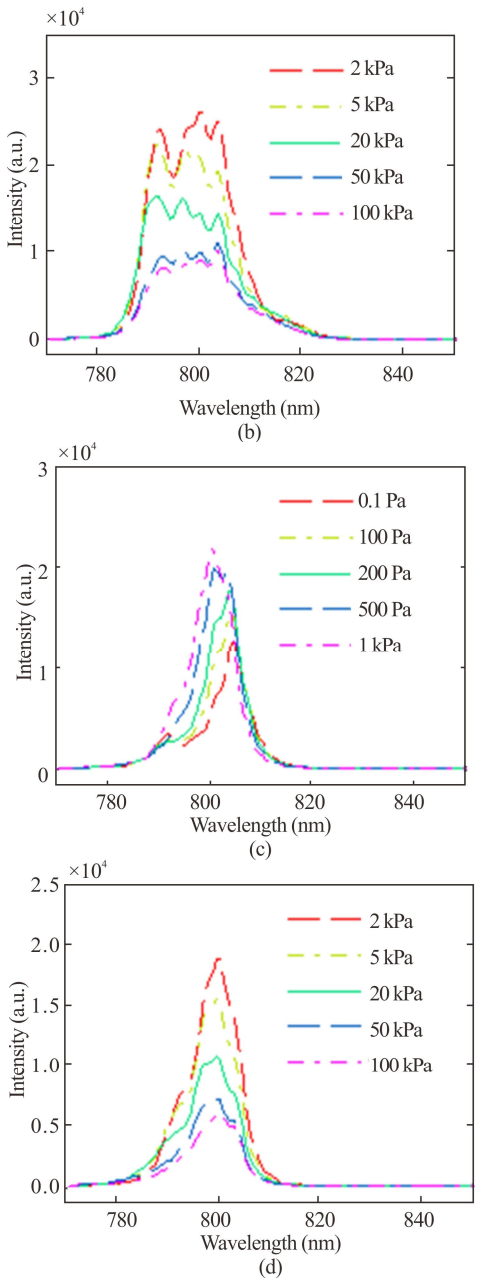


Fig.4 Transmitted spectra of the focused femtosecond pulses at different air pressures when (a, b) BF_1 + NF and (c, d) BF_2 + NF are used (The pulse energy is 2.48 mJ)

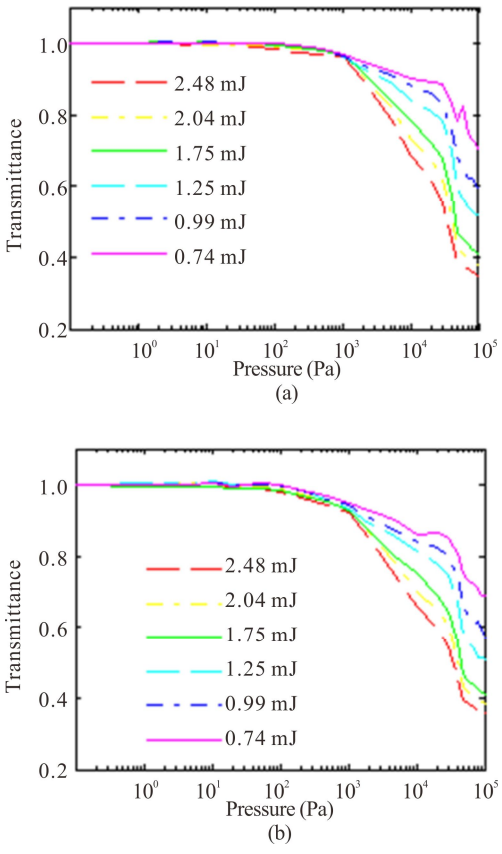
Spectrographic method: Firstly, the transmitted spectrum is integrated over the wavelength to obtain the integral intensity I, which is directly proportional to the transmitted pulse energy E. When the laser pulse propagates in vacuum, no energy depletion occurs. In this paper, the integral intensity measured at 0.1 Pa is set as I_0, and the corresponding transmitted pulse energy is set as E_0; the integral intensity measured at other air pressures is set as $I(p)$ and the corresponding transmitted pulse energy is set as $E(p)$. Consequently, the energy transmittance measured by the spectrographic method is given by

$$T_S(p)=\frac{E(p)}{E_0}=\frac{I(p)}{I_0}. \tag{2}$$

Photoelectric method: Firstly, the received optical signals are converted into electrical ones by using a silicon photodiode, and then the electrical signals are guided to the oscilloscope through an electrical wire. The transmitted pulse energy is proportional to the voltage of the electrical signals. In this paper, the voltage measured at 0.1 Pa is set as V_0 and the corresponding transmitted pulse energy is set as E_0; the voltage measured at other air pressures is set as $V(p)$, and the corresponding transmitted pulse energy is set as $E(p)$. Consequently, the energy transmittance measured by the photoelectric method is given by

$$T_V(p)=\frac{E(p)}{E_0}=\frac{V(p)}{V_0}. \tag{3}$$

Using the two methods, we obtain the energy transmittance of the focused femtosecond pulses at different air pressures, as shown in Fig.5. It can be seen from the figure that the energy transmittances measured by the two methods are in good agreement. Due to the use of the BF, the spectral range of the transmitted light signals is confined to the vicinity of central wavelength of the incident femtosecond pulses. Though the spectral response of the spectrometer and silicon photodiode differs from each other, the spectral and electrical measurement after the BFs agree very well. In effect, if we attempt to characterize the supercontinuum, a detector with flat spectral response should be used to measure the complete signal[27].



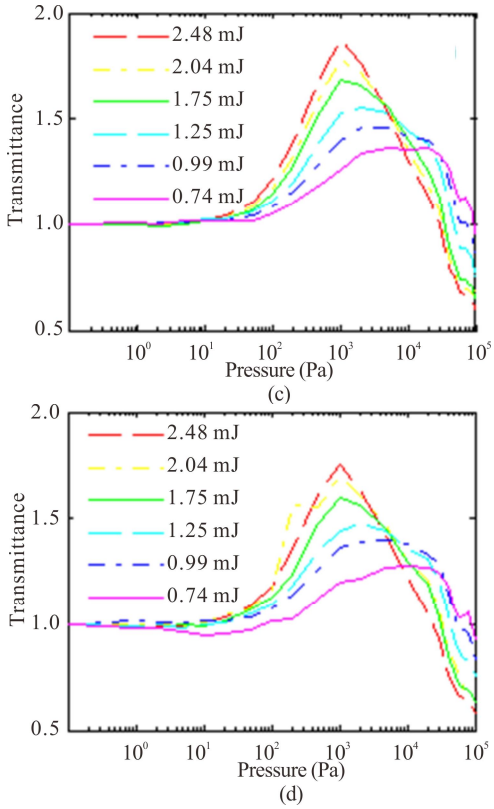


Fig.5 Energy transmittance of the 800 nm femtosecond pulses with air pressure when (a, b) BF_1 and (c, d) BF_2 are used ((a, c) and (b, d) are measured by the spectrographic and photoelectric methods, respectively)

It can be seen from Fig.5(a) and (b) that the energy transmittance of the femtosecond pulses remains almost unchanged when the air pressure is below 1 kPa, which is close to 1. Under this circumstance, the pulse energy is hardly depleted. Though the spectrum changes below 1 kPa, it only occurs around the central wavelength and no significant spectral broadening occurs, as shown in Fig.4(a). In this case, the transmitted spectra remain coherent. In this paper, we call this air pressure region (p<1 kPa) as the nonfilamentation regime. When the air pressure is higher than 1 kPa, the energy transmittance of the femtosecond pulses decreases with increasing air pressure, as shown in Fig.5(a) and (b). It comes to the filamentation regime. p=20 kPa is the typical starting point for filamentation of femtosecond pulses[19] for the self-focusing effect can be obvious when the air pressure is higher than 20 kPa. In our experiment, the femtosecond laser beams are focused by the lens, and the laser intensity is enhanced by the linear focusing, generating weak plasma channel, thereby greatly lowering the starting point for femtosecond filamentation in air[28]. Some difference can be also observed. In the range from 1 kPa to 20 kPa, the energy transmittance decreases slowly with increasing air pressure. When the air pressure is higher than 20 kPa, the self-focusing effect starts playing

a role, and it works with the linear focusing effect, enhancing the molecular excitation and ionization, etc, and thus consuming more energy. Consequently, the energy transmittance decreases more drastically with increasing air pressure in the case of p >20 kPa.

In the filamentation regime, the laser intensity and plasma density change drastically in time, giving rise to intense SPM effect, and therefore the spectrum is greatly broadened. Consequently, more energy is transferred from the central wavelength to other wavelengths, resulting in the decrease of spectral intensity around the central wavelength and the decrease of energy transmittance. In effect, by using a set of NF and BF with the same action range, the contribution of spectral broadening induced by the SPM to the decrease of energy transmittance can be quantitatively determined. By this way, one can weight spectral broadening on the energy transmittance, and further determine the dominant factor leading to the decrease of energy transmittance, which will be studied in the further work. Here, we can only say that the transmitted spectra contain bot coherent and incoherent components, and the proportion of incoherent components increases with increasing air pressure.

Even for the energy conversion, the present methods are unable to quantitatively analyze the contribution of each effect to the energy depletion of the focused femtosecond pulses, because it is difficult to measure the energy consumed by each effect. Besides, some effects are coupled with each other in the femtosecond filamentation process, which can not be separately considered. More importantly, apart from the excited N_2 and O_2 molecules, some other particles like N_2^+, O_2^+, O_4 and N_2O_2, etc are formed during the femtosecond filamentation[5,16,21,23,24,29-32]. When the air pressure changes, the composition of air changes accordingly. For example, the molecular excitation and ionization are greatly affected by the air pressure. At higher air pressures, both the photo-ionization and impact ionization contributes to the ionization of molecules, while at lower air pressures, the role of impact ionization can be neglected due to the low collision probability[21,23,24]. Moreover, the photo-ionization regime can be divided into multiphoton ionization (MPI) and tunneling ionization (TI), which can be judged according to the Keldysh theory[33]. The photo-ionization regime differs at different air pressures. At higher air pressures (close to one atmospheric pressure), the laser intensity is clamped to about 5×10^{13} W/cm^2 due to the intense plasma defocusing effect, and thus the MPI dominates. At lower air pressures (close to vacuum), the laser intensity can exceed 1×10^{14} W/cm^2 since few plasmas are generated (weak defocusing), and thus the TI dominates. At the intermediate air pressures, the MPI and TI coexist. As a result, it is difficult to quantitatively discuss the contribution of these effects to energy depletion separately. In the paper, we can only consider their collective effect on energy depletion.

It can also be seen from Fig.5(a) and (b) that the energy transmittance decreases more drastically with increasing air pressure at higher pulse energy. In other words, at the same air pressure, the energy transmittance decreases with increasing pulse energy. Since the critical power of the self-focusing for femtosecond pulses P_{cr} is inversely proportional to the air pressure[22,28], the femtosecond laser pulses with lower energy can only generate filament at higher air pressures, i.e., no filament is formed at lower air pressures, and therefore less energy is consumed. As a result, at the same air pressure, more pulse energy is consumed for the higher pulse energy cases in which filamantation is more likely to occur.

When BF_2 is used, it is found that the energy transmittance begins to increase at 10 Pa, and reaches its maximum at about 1 kPa, as shown in Fig.5(c) and (d). The phenomenon that the energy transmittance is larger than 1 seems to violate the energy conservation law. In effect, the action range of BF_2 is narrower than the linewidth of the femtosecond laser, making it impossible to collect the complete spectral signals around the central wavelength. When the air pressure is lower than 1 kPa, the spectrum around the central wavelength of the femtosecond pulses changes (see Fig.4), resulting in the spectrum collected at 1 kPa being more intense than that collected at 0.1 Pa. When the air pressure is higher than 1 kPa, the cause for the decrease in energy transmittance is the same as the cases when BF_1 is used. To obtain the correct energy transmittance of central wavelength of the femtosecond pulses, a BF with proper action wavelength range is required.

In this paper, the transmitted spectrum of the focused femtosecond pulse around its central wavelength is measured, and the influence of air pressure and initial pulse energy on it is studied. By integrating the transmitted spectrum, we deduce the transmitted pulse energy and further obtain the energy transmittance of the central wavelength of the focused femtosecond pulse. The air pressure affects the nonlinear effects occurring in the interaction between femtosecond laser and air, which inevitably consumes pulse energy thus lowering the energy transmittance. Due to the linear focusing effect, the starting point for the filamentation of focused femtosecond pulses is greatly lowered (to about 1 kPa). At lower air pressures (below 1 kPa), no filament is formed, the pulse energy is hardly depleted, and the energy transmittance remains at about 100%. At higher air pressures, femtosecond filamentation occurs, a lot of energy is consumed due to the strong interaction between the femtosecond pulses and air molecules, leading to the decrease in the energy transmittance. It is also found that the higher the pulse energy, the lower the energy transmittance is. Besides, the SPM effect transfers the laser energy of the central wavelength to other wavelength, thus also affecting the energy transmittance. Though it is difficult to weight the influence of the nonlinear processes on the femtosecond filamentation and energy

transmittance, we can say that the transmitted spectra contain bot coherent and incoherent components. At lower air pressures (below 1 kPa), the transmitted spectra mainly contain coherent components due to the weak interaction between femtosecond laser and air, while at higher air pressures, the proportion of incoherent components increases with increasing pressure. The energy transmittance is qualitatively analyzed through the energy conversion and transfer effect, which may be helpful to the understanding of energy depletion during the transmission of intense ultrashort pulses in ambient air. We hope this work can provide a guidance for the practical applications of femtosecond filamentation, like remote sensing, and optical communication in free space.

Ethics declarations

Conflicts of interest

The authors declare no conflict of interest.

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