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Invited Paper

Influence of laser polarization on plasma fluorescence emission during the femtosecond filamentation in air



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ABSTRACT

The laser polarization state has a great influence on the plasma fluorescence emission during femtoseond filamentation in air. For the spectral lines from N₂, in the case of focusing lens with longer focal length (f=100 cm), due to the impact excitation, circular polarization leads to stronger fluorescence emission when the laser energy is higher than the 'energy threshold' (2.0 mJ). As a lens with shorter focal length (f=40 cm) is used, a similar phenomenon can be observed, however, the 'energy threshold' is much lower, which is lower than 0.8 mJ. For the lines from N[±]₂, especially for the 391 nm one, their emission is stronger in the linear polarization state. The mechanism of plasma fluorescence emission during femtosecond filamentation is discussed based on the analysis of these phenomena, which will be helpful to the remote sensing and spectrum analysis.

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1. Introduction

As the femtosecond laser beam propagates in air, due to the self-focusing action resulting from Kerr effect, whose intensity becomes increasingly higher which eventually reaches the ionization threshold of air molecules, thereby generating plasmas which play the defocusing role. When the Kerr self-focusing effect, diffraction, and plasma defocusing effect reach the dynamical balance, a stable plasma channel will be formed, which is called filament. Since the first experimental observation of filamenation, it has gained great attention due to its promising applications in terahertz emission [1–3], remote sensing of atmospheric constituents [4–6], generation of air laser [7–9], *etc.*

After the filamentation of femtosecond pulse in air, the plasma left behind will undergo complex transitions, emitting characteristic fingerprint fluorescence [8]. Measuring the spectroscopy along the propagation path, we can not only extract the plasma density, electron temperature as well as laser intensity inside filaments [10–13], but also get insight into the excitation and ionization process during the filamentation [14]. What is more, due to the high sensitivity, non-intrusiveness, and real-time analysis of the laser-induced fluorescence, it has been extensively employed for sensing atmospheric trace species [4–6]. More importantly, femtosecond filamentation can be well controlled to occur at a distance as far as several kilometers in the atmosphere [15,16],

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making the remote sensing of atmospheric constituents possible.

It is known that during femtosecond filamentation in air, the fluorescence mainly comes from the first negative band system $(B^2\Sigma_u^+ - X^2\Sigma_g^+ \text{ transition})$ of N₂ and second positive band system $(C^3\Pi_u^+ - B^3\Pi_g^+ \text{ transition})$ of N₂ [17–21]. In the practical applications like air laser, nitrogen molecules are often used as the working matter [7–9], and the most direct application of air laser is remote sensing, therefore, investigating the fluorescence emission from the nitrogen molecules will be helpful to its practical applications. It has been found that by applying the circularly polarized femtosecond pulses, the UV fluorescence emission can be greatly enhanced [14,22]. However, the evolution of the spectral lines under different laser polarizations along the filament axis is less investigated, and the mechanism of plasma fluorescence emission during filamentation is still an open question.

In the present work, by measuring the nitrogen characteristic spectra, the influence of laser polarization on the propagation of femtosecond laser pulses in air is investigated. We study the fluorescence emission in the linear and nonlinear focusing regimes under different polarizations and attempt to discuss the mechanism of plasma fluorescence emission during femtosecond filamentation in air.

2. Experimental setup

Fig. 1 presents the schematic of experimental setup. We conduct the experiment by using an one-box ultrafast Ti: Sapphire amplifier (Coherent Libra) with the wavelength of 800 nm, the



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Fig. 1. Schematic of experimental setup to measure the fluorescence spectra during femtosecond filamentation in air. F: optical fiber; G: Glan laser polarizer; H: half-wave plate; L: focusing lens; Q: quarter-wave plate; R: rectangular diaphragm.

width of 50 fs at the repetition rate of 1 kHz. The combination of a half-wave plate and a Glan laser polarizer adjusts laser energy to the desired value. The laser beam is focused by focusing lens L (f=40 cm and 100 cm) to generated filament in air. The produced plasma fluorescence spectra are firstly collected by two lenses (BK7 f=75 mm) fixed on a moving stage along the propagation path, and then guided to the spectrometer (Spectra Pro 500i, PI Acton, and the grating is 150 grooves/mm) through a fiber. The light is detected using an intensified charge coupled device (ICCD, PI-MAX4, Princeton Instruments) with 1024 × 1024 pixels. A rectangular diaphragm R (1 mm × 10 mm) is placed 10 mm away from the filament axis to resolve longitudinally the fluorescence signal along the propagation path. To improve the signal-to-noise ratio, each data point throughout this paper is typically an average of 10 groups of 500 shots' accumulation.

In the experiment, we attempt to investigate the influence of the polarization of laser pulse on its propagation. The polarization state of laser pulse is changed by rotating the quarter-wave plate, as its rotation angle is $\varphi = 45^{\circ} + n \times 90^{\circ}$, it corresponds to the circular polarization state; which as it is $\varphi = n \times 90^{\circ}$, it corresponds to the linear one. The rectangular diaphragm, lenses and optical fiber fixed on the moving stage move back and forth so as to measure the spectra along the propagation path, where the nearest distance from it to the focusing lens is d=35 cm and d=87 cm as its focal length is 40 cm and 100 cm, respectively. For the purpose of facilitating the record of experimental data, these positions are defined as z=0 mm, therefore, the actual propagation distance is Z=z+d.

3. Results and discussion

Fig. 2 shows the evolution of the 337 nm spectral line along the propagation axis as the air is irradiated by the circularly, elliptically and linearly polarized pulses with different energies. The focal length of the focusing lens is f = 100 cm. When the energy is 0.8 mJ, the intensity of 337 nm signal in the linear polarization case is much higher than that in the elliptic and circular polarization cases, as shown in Fig. 2(a). As we increase the laser energy, the maximum values of 337 nm signal for different polarizations become equal to each other when the energy is 2.0 mJ (we call it 'energy threshold'), as shown in Fig. 2(b). If the laser energy is increased further, it can be seen there exists a reversal in intensity of fluorescence between linear and circular pump laser polarization, as shown in Fig. 2(c), which has already been reported by Mitryukovskiy et al. [14]. They attributed this phenomena to the onset of impact excitation in the case of circularly polarized pulses:



Fig. 2. Spatial evolution of the 337 nm signal generated by the linearly, elliptically (ellipticity e=0.5) and circularly polarized pulses as the laser energy is (a) 0.8, (b) 2.0 and (c) 3.1 mJ, respectively. The focal length is f=100 cm.

$$N_2(X^1\Sigma_g^+) + e \to N_2(C^3\Pi_u^+) + e \tag{1}$$

$$N_2(X^1\Sigma_g^+) + e \to N_2^+(B^2\Sigma_u^+) + 2e \tag{2}$$

For the processes (1) and (2), only when the electron energy exceeds a relative high threshold energy [11 eV for process (1) and 18.75 eV for process (2)], can they occur [14,23]. Irradiated by the linearly polarized laser pulses, free electrons generated experience alternative acceleration and deceleration by the laser field during each optical cycle of the pulse, as a result, their kinetic energies are low; in contrast, irradiated by the circularly polarized ones, electrons are always accelerated away from the molecular ion, and thus their energies can be relative high, which can open up the impact excitation channel. Mitryukovskiy et al. calculated electron energy distribution in the case of linearly, elliptically and circularly polarized laser pulses (see Fig. 3 in [14]), and which supports the above analysis. It should be noted that the cross-sections for impact excitation depend on the energy of electrons driven by the laser field or their temperature. Along with the increase of the electron energy, the cross-sections firstly increase and then decrease, and for process (1) and (2), they peak at 14.5 and 110 eV, respectively (see Tables 11 and 19 in Ref. [23]).

In addition, no matter how high the laser energy is, the filament length is the largest in the linear polarization case. In general, there exists a 'critical position' on the propagation axis where the fluorescence intensity is almost independent of laser polarizations. For instance, as the laser energy is 2.0 mJ, the 'critical position' is at z=103 mm [see Fig. 2(b)], before this position, the fluorescence emission in the linear polarization case is more intense than that in the circular polarization case, and vice versa. It can be also seen from Fig. 2 that along with the increase of pulse energy, the plasma string moves towards the laser source, which is a signature of filamentation.

It can be also seen from the Fig. 2 that, the position where the 337 nm signal reaches its maximum is different for different

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