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Lasing and fluorescence of air plasma in presence of an external electric field

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Cavity-free lasing of nitrogen molecules pumped by intense femtosecond laser pulses holds the potential for remote sensing of electric fields. Here we compared the influence of an external direct current (DC) electric field on both the directional lasing radiation and omnidirectional fluorescence emission of neutral nitrogen molecules. It was found that the forward lasing radiation in both pure nitrogen gas and ambient air shows a sensitive dependence on the direction and strength of the DC field, while the fluorescence is not influenced. The effect of pump laser polarization was also investigated. The distinct behavior of the lasing and fluorescence in response to the DC field was attributed to their different dependences on the population distribution of excited nitrogen molecules. This study consolidates the method for standoff detection of electric field with an air lasing effect in the atmosphere.

Keywords: femtosecond filamentation, air plasma, remote sensing

PACS: 31.70.–f, 34.50.Gb, 34.90.+q, 52.38.–r

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1. Introduction

Intense femtosecond laser pulses experience extremely nonlinear propagation, termed "filamentation" or "femtosecond filaments", when they are launched in ambient air.[1–5] Filamentation features a variety of spectacular phenomena, including generation of a long (centimeters to hundreds of meters) and bright plasma channel in air, a supercontinuum spanning from ultraviolet to visible light and near infrared, as well as a characteristic sound wave for the human ear.[1,5–8] The underlying physics of filamentation has been attributed to the working together of a few linear and nonlinear effects including dispersion and diffraction, the optical Kerr effect, ionization and plasma generation, absorption and loss, etc.[1,5–12] The filamentation process has found various applications in pulse compression,[13–15] remote sensing,[16,17] triggering and guiding of discharge and lightning[18–20] and micromachining,[21,22] to name just a few. Among the various applications, the fluorescence emission of filamentary air plasma has been utilized as a sensor for electric fields. Sugiyama *et al.* reported that air plasma fluorescence can be enhanced in the presence of high voltage of a few MV.[23] Inspired by this observation, Liu and coworkers demonstrated that the fluorescence signal of nitrogen molecules can be employed for the detection of THz fields with strengths of several hundred kV/cm.[24,25] The fluorescence emissions peaking at 337.4 nm, 357.2 nm and 380.2 nm correspond to the optical transition between the third excited state $C^3\Pi_u\,(v=0,1,2,\ldots)$ (hereinafter referred to as "C") and the second excited state $B^3\Pi_g\,(v'=0,1,2,\ldots)$ (hereinafter referred to as "B") of neutral nitrogen molecules, with different vibrational quantum numbers v and v'.

It has also been shown that stimulated emission at 337.4 nm and 357.2 nm in the forward and backward directions can be obtained in pure nitrogen gas or ambient air pumped by powerful circularly polarized 800 nm pulses.[26–29] With circularly polarized intense 800 nm femtosecond laser pulses, free electrons due to photoionization can be significantly accelerated in the laser field and acquire substantial kinetic energy around $2U_p$, with $U_p = e^2I/(2\omega_0^2 c\varepsilon_0 m_e)$ being the ponderomotive energy of electrons in the laser field with an intensity I and frequency ω_0.[30,31] Inelastic collision of such high-energy electrons with N_2 molecules in the ground state can lead to population inversion between the C and B states, giving rise to stimulated emission at 337.4 nm in both the forward and backward directions of pump laser propagation.

Recently, some of the current authors showed that cavity-free lasing emission of neutral nitrogen molecules can be used for standoff sensing of external electric fields. In fact, significant modulation of both the backward and forward 337.4 nm stimulated emissions was observed for an external electric field of a few kV/cm,[32] providing a more sensitive technique for remote sensing of electric fields with filamentary air plasma. In the proof-of-principle work by Zhang *et al.*,[32] lasing emission was obtained in pure nitrogen gas and the external DC field was applied in the vacuum chamber, hindering the practical application of this method in the atmosphere. More-

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over, the response of air plasma fluorescence under such a DC field remains unexplored, leaving a comparison between the behaviors of lasing and fluorescence highly necessary in view of future applications.

In this study, we compared the forward stimulated emission and the lateral fluorescence at 337.4 nm from femtosecond laser filaments under the influence of an external DC electric field. Utilizing high-intensity circularly polarized 800 nm femtosecond laser pulses, we found that the forward 337.4 nm signal exhibits a pronounced modulation in response to the external electric field. As the electric field strength varies between -1 kV/cm and 1 kV/cm, the intensity of the forward 337.4 nm laser signal exhibits a monotonic increase. This behavior was also confirmed in ambient air. In contrast, it was observed that the lateral 337.4 nm fluorescence signal remains invariant with respect to the magnitude and orientation of the electric field, regardless of the polarization state of the pump laser. Consequently, our observations manifest the advantage of bi-directional air lasing for remote sensing of an electric field with respect to the fluorescence. We therefore believe that the 337.4 nm signal excited by the strong circularly polarized femtosecond laser can serve as a robust detector for external electric fields, which has potential application in atmospheric environmental detection and weather forecasting.

2. Experimental setup

In the experiment, we used a Ti:sapphire laser system (Coherent Legend DUO) that provides femtosecond laser pulses with a central wavelength of 796 nm, pulse width of 35 fs, repetition rate of 1000 Hz and a maximum output energy of 12 mJ. The laser pulse passed through a quartz quarter-wave plate to convert the linearly polarized pulse into circular polarization. The pulses were focused by a focusing lens with a focal length f_1 of 75 cm to form a plasma filament with a length of about 10 cm in a gas chamber or ambient air, as shown in Fig. 1. A pair of parallel electrode plates (10 cm$\times$10 cm) were installed around the plasma to ensure that the entire plasma is immersed in the electric field and a DC electric field is applied in the vertical direction. The forward 337.4 nm laser signal and the lateral 337.4 nm fluorescence signal generated from the plasma were coupled into a fiber spectrometer (Avantes, Avaspec-3648) for observation. The forward laser signal was filtered through a dichroic mirror (DM) that reflects 800 nm and transmits 337.4 nm, as well as a narrowband filter at 337 nm. During the experiment, the changes in the forward 337.4 nm laser signal and the lateral 337.4 nm fluorescence signal were observed by varying the magnitude and direction of the electric field, as well as the polarization state of the pump laser. For 337.4 nm emission in the forward direction, we found that a slight rotation of the quarter-wave plate led to a significant decrease or disappearance of the 337.4 nm signal, confirming that the signal was due to stimulated emission, not fluorescence, as reported before.[26,27]

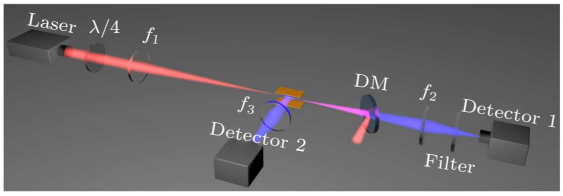


Fig. 1. Experimental setup. We observed both the forward stimulated emission and the lateral fluorescence from the femtosecond filaments in the presence of an external electric field. Here, detector 1 and detector 2 are spectrometers. The focusing lens f_1 has a focal length of 75 cm, while f_2 and f_3 are collection lenses with a focal length of 10 cm.

3. Results and discussion

Figure 2 presents the characteristic forward laser emission spectra and fluorescence spectra recorded in the vicinity of 337.4 nm. The sharp peak observed at 337.4 nm is attributed to the radiative transition of nitrogen molecules from the excited state $C^3\Pi_u^+(v=0)$ to the ground state $B^3\Pi_g^+(v=0)$. In the fluorescence spectrum shown in Fig. 2(b), multiple sharp peaks are discernible, which are ascribed to transitions between various vibronic energy levels. The corresponding transition channels are annotated in the figure. We also present the spectra obtained with a linearly polarized pump laser for comparison. It is seen that the forward lasing signal almost disappears while the fluorescence signal presents a slight decrease, which has been reported previously.[30,31]

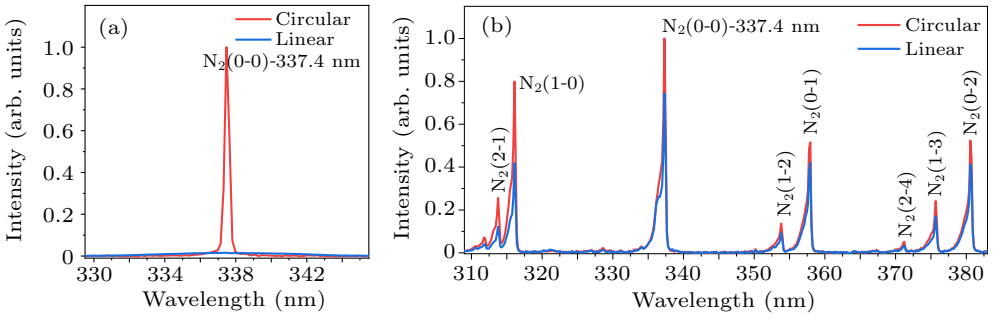


Fig. 2. Typical spectra of forward lasing at 337.4 nm (a) and the fluorescence signal (b). The signal was normalized separately in (a) and (b). The results were obtained in pure nitrogen gas with a pump laser energy of 12 mJ.

3.1. Effect of an electric field on the forward lasing signal

We first investigated the influence of a DC electric field on the forward 337.4 nm lasing emission in pure nitrogen gas and ambient air. The results are presented in Fig. 3 for negative electric fields (the electric field direction is oriented from top to bottom) of three different amplitudes. These spectral signals were captured by the fiber of the spectrometer positioned 1.4 m from the plasma in both pure nitrogen and air. In both Figs. 3(a) and 3(b) it is evident that the intensity of the 337.4 nm lasing signal exhibits a biphasic response to varying electric field amplitudes of 0 kV/cm, −0.3 kV/cm and −1.5 kV/cm, initially diminishing with subsequent augmentation.

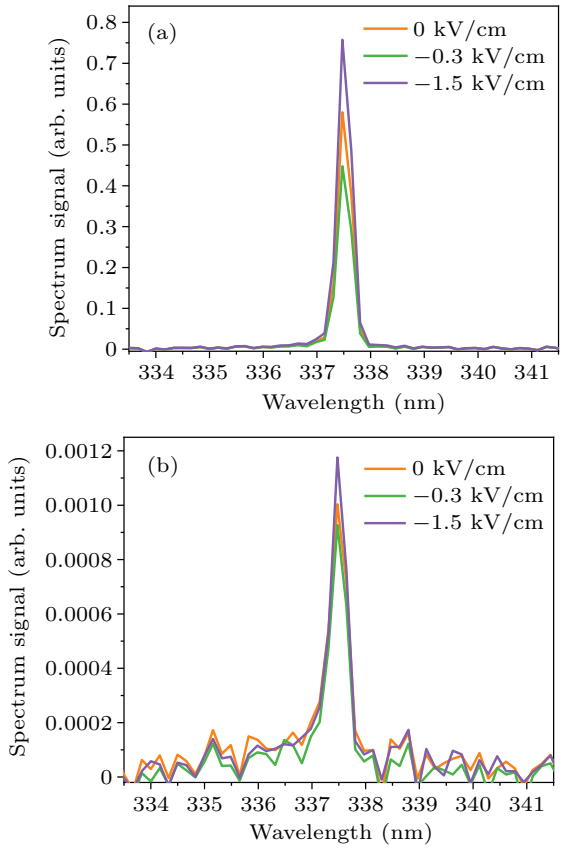


Fig. 3. Influence of the electric field on the 337.4 nm signal measured 1.4 m away from the plasma: panels (a) and (b) show the 337.4 nm spectral signals for different strengths of electric field in nitrogen and air, respectively.

Figure 4 summarizes the variation of the 337.4 nm lasing signal with the direction and strength of the external DC electric field. Figures 4(a) and 4(b) show that, in both pure nitrogen and air environments, the intensity of the 337.4 nm signal undergoes significant alterations in accordance with variations of the external electric field. Moreover, the 337.4 nm lasing signals in both nitrogen and air exhibit a strikingly similar behavior. For $E > 0$ kV/cm (the electric field direction is from bottom to top), the intensity of the 337.4 nm lasing signal increases with the increase of the electric field and subsequently exhibits saturation. In contrast, for $E < 0$ kV/cm (the electric field direction is from top to bottom), two distinct stages emerge: for $E > -0.3$ kV/cm the intensity of the 337.4 nm signal diminishes as the electric field amplitude augments, while for an electric field going beyond −0.3 kV/cm the signal increases with the electric field amplitude and then displays a saturation trend. These observations confirmed our previous report on forward and backward lasing emissions in pure nitrogen gas with a different focal condition of $f = 750$ mm.[32] The different dependence of the lasing signal for positive and negative electric fields has been attributed to asymmetric electron motion in the x–y plane driven by the intense 800 nm field together with the residual 400 nm second harmonic pulse, which is found to be generated in the quarter-wave plate.[32] The saturation behavior of the forward signal may be attributed to the lasing signal entering the regime of saturation amplification.

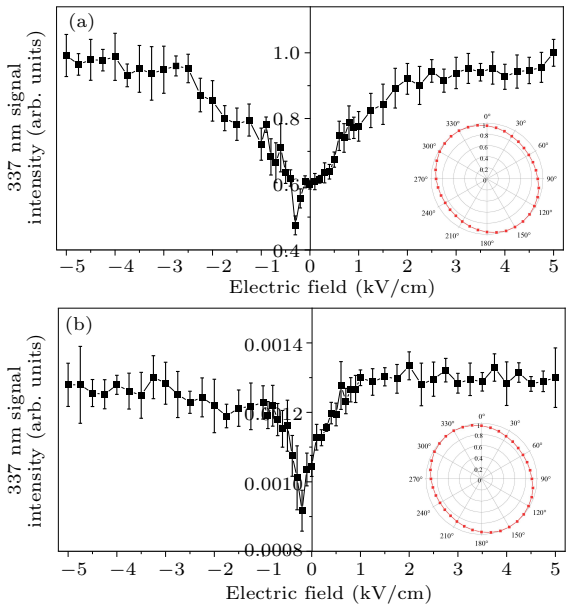


Fig. 4. Effect of an electric field on the 337.4 nm forward lasing emission signal (a) in nitrogen and (b) in air. The inset presents polarization measurement of the pump laser.

3.2. Effect of electric field on the fluorescence signal

Next, we investigated the plasma fluorescence signals subjected to an external electric field in pure nitrogen gas. The effect of the electric field for pump pulses with different polarization states on the nitrogen plasma fluorescence signals was measured, and the results are depicted in Fig. 5. The experimental results show that the 337.4 nm fluorescence signal intensity generated by pump light of different polarization states is almost independent of the applied electric field. Note that the signal intensity in Fig. 5(a) is about 1.2 times stronger than that of the two linearly polarized cases. This finding aligns with the observations reported previously.[30] Additionally, it is notable that irrespective of the polarization state of the pump light, linear or circular, the 337.4 nm fluorescence signal does not show significant changes with the magnitude and direc-

tion of the electric field. Consequently, the fluorescence signal demonstrates a lack of sensitivity to external electric fields, which stands in marked contrast to the behavior of the forward lasing signal.

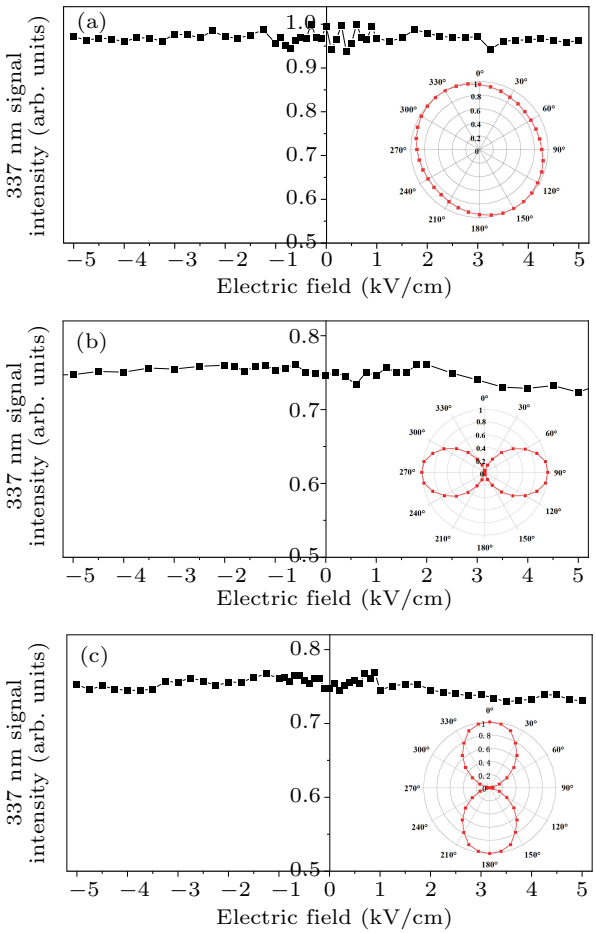


Fig. 5. Effect of an electric field on the 337.4 nm fluorescence signal. The incident laser pulse is (a) circular polarized, (b) linearly polarized in the horizontal direction and (c) linearly polarized in the vertical direction. The inset presents the polarization measurement of the pump laser.

In the above experiments one can notice a sharp contrast between the fluorescence and the lasing signal at 337.4 nm. The fluorescence is almost uninfluenced by the electric field (Fig. 5) while the lasing signal is significantly modulated (Fig. 4). Moreover, electric fields of different polarities lead to either an increase or a reduction of the 337.4 nm lasing signal for $|E| < 4$ kV/cm (Fig. 4(a)). We speculate that the different behaviors are rooted in the distinct dependence of two signals on the population distribution of the excited N_2 molecules. As mentioned above, the 337.4 nm emission corresponds to the transition $C^3\Pi_u(v=0) \rightarrow B^3\Pi_g(v'=0)$. The fluorescence signal has the nature of spontaneous emission, and is proportional to the population density in the C state according to the Einstein relation between emission and absorption, i.e., $S_{\text{fluo}} \propto n_C$. In the meantime, the bidirectional 337.4 nm lasing signal is stimulated emission and shows an exponential dependence on the population difference between the upper and lower states, i.e., $S_{\text{lasing}} \propto \exp[(n_C - n_B)]$. Therefore, one expects that a slight variation of the population n_C will lead to a substantial change in the lasing signal while the fluorescence barely changes, in line with our observations in Figs. 4 and 5. These different behaviors show that the bidirectional lasing signal is much more sensitive to the electric field, which is an advantage for standoff optical detection.

Now we explain why the population density in the C state is modulated by the external electric field. The 337.4 nm signal of nitrogen molecules originates from inelastic collisions between the ground state nitrogen molecules and high-energy electrons, and the population transfer is established by $N_2(X^2\Sigma_g^+) + e = N_2(C^3\Pi_u) + e$. Current research shows that an externally applied electric field can change the energy distribution of free electrons in the laser-induced plasma filament.[33] Specifically, the acceleration/deceleration of electrons under the Lorentz force modulates their kinetic energy, which gives rise to the collisional ionization process. Under our experimental conditions (pump laser intensity $I \sim 10^{14}$ W/cm^2), the electron energy distribution peaks at about 16 eV, which exceeds the threshold ($\sim$ 11 eV) required for the excitation of the $C^3\Pi_u$ state. At the same time, the externally applied electric field affects the electron energy distribution and causes changes in the population distribution of the $C^3\Pi_u$ state and the $B^3\Pi_g$ states, resulting in enhancement or weakening of the 337.4 nm radiation signal.[34] Therefore, we attribute the modulation of the lasing signal by the externally applied electric field to the acceleration or deceleration of electrons inside the plasma caused by the external electric field.

Finally, one can note that a positive or negative electric field leads to either an increase or decrease of the lasing signal, as presented in Figs. 4(a) and 4(b). In our previous study we attributed this effect to asymmetrical motion of the electrons in the transverse plane due to the presence of a weak residual second-harmonic 400 nm field.[35] This residual 400 nm field was found to be generated by the quartz quarter-wave plate. Here we would like to point out another possible origin of the 400 nm field. With the application of an external electric field on the plasma formed by the femtosecond laser pulses, a third-order sum frequency generation process can lead to the production of a second harmonic field through $E_{400\text{ nm}} \sim \chi^{(3)} E_{\text{dc}} E_{800} E_{800}$.[36] This process has been employed for the detection of terahertz fields with a laser-formed air plasma.[37] In addition to the residual second harmonic of the quarter-wave plate, this 400 nm field generated inside the plasma will also impart motion of the ionized electrons and further promote the formation of an asymmetrical electron current. Consequently, electrical fields of opposite directions will accelerate or decelerate electron motion in general and lead to an increase/decrease in the average kinetic energy of the electrons.[31] This change in electron en-

ergy corresponds to variation of the C state population due to inelastic collisions. As a result, the lasing signal is enhanced/suppressed with electric fields of different directions. To quantify the role of the two 400 nm fields from different sources, a systematic investigation of the intensity and polarization of the 400 nm fields is necessary in view of practical application for standoff detection.

4. Conclusion

In summary, we investigated the influence of external DC electric fields on neutral nitrogen molecular lasing and fluorescence in both air and pure nitrogen gas. The results demonstrate that DC electric fields at the kV/cm level give rise to substantial modulation of the nitrogen molecular lasing signal while the lateral fluorescence signal is nearly untouched. We attributed these two different behaviors to their distinct dependences on the population distributions of excited states. The observed modulation of the 337.4 nm lasing signal is ascribed to electric field-induced variation of the kinetic energy of free electrons within the plasma, which subsequently alters the population dynamics of the upper laser level ($C^3\Pi_u$ state) through electron–molecule collision excitation. In addition, we proposed a new possibility for the asymmetric signal response to the polarity of the electric field. Specifically, second harmonics can be generated by third-order nonlinear effects in the presence of an electric field; this modifies ionization electron trajectories and results in the formation of asymmetric electron currents. This work advances our understanding of nitrogen molecular lasing for electric field detection and provides insights for developing air-lasing-based high-sensitivity, non-invasive electric field detection in atmospheric environments.

Acknowledgements

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