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Abstract: We report on the observation of backward lasing at 391.4 nm of nitrogen ions pumped by linearly polarized intense femtosecond pulses at 800 nm. The strongly enhanced spectral intensity at 391.4 nm, as well as the amplification of an externally injected backward seeding pulse, confirm that the backward 391.4 nm signal is due to optical amplification in the nitrogen gas plasma. Compared to the forward emission at 391.4 nm, the optimal backward emission is achieved at a lower gas pressure around 10 mbar, which is due to asymmetry of the backward and forward directions rooted in the traveling excitation geometry. Comparison of the signals in pure nitrogen and air revealed a strong quenching effect of the oxygen molecules, preventing backward lasing action in ambient air.

1. Introduction

Cavity-free lasing of the air molecules or their constituent atoms or ions in a plasma pumped by ultrafast laser is intensively studied in recent years [1-18]. In particular, lasing emission in the backward direction (opposite direction of the pump laser propagation) has attracted much attention [1, 10-18], since it holds unique potential for optical remote sensing applications. In the traditional optical remote sensing, laser pulses from the ground laser station are shoot into the sky and the backward scattered photons or fluorescence signal are detected by the ground observer [19]. Since these emissions are incoherent, emitted in 4π solid angle, the optical signal collected by the ground observer is very weak. When backward air laser is used for the detection of trace gas or pollutions in atmosphere, the backward laser beam carries the information of the target under investigation [20]. Compared to the scattered photon or the fluorescence, the backward laser beam is emitted in a very small solid angle and therefore the optical signal can be tremendously increased [10, 12-14]. More importantly, coherent nonlinear optical techniques such as Stimulated Raman Gain/Loss can be used for air pollution detection [20], while the common detection techniques based on scattered photons or fluorescence are normally incoherent.

Up to now, several methods for generation of backward lasing action with air constituents have been demonstrated. In the first report of Dogariu *et al*, both backward and forward stimulated emissions of oxygen atoms at 845 nm have been observed when ambient air was pumped by picosecond pulses at 226 nm [1]. Later, it was shown that the threshold of backward lasing can be significantly reduced by pre-disassociation of the oxygen molecules with a nanosecond pre-pulse [12]. Recently, it has been reported that nitrogen atoms can lase in a similar manner when air is pumped by intense 206 nm pulses [10]. However, applications of this technique for remote sensing are limited due to a poor transmission of the deep-UV pump pulses in atmosphere. Backward lasing emission of neutral nitrogen molecules was reported in two schemes [13-18]. Strong backward emissions at 337 and 357 nm have been observed when the mixture of a nitrogen and a high pressure argon gas was pumped by linearly polarized mid-infrared pulses at 3.9 or 1.03 μm [13]. However, this technique cannot be used in ambient air since a high-pressure argon (> 3 bar) is necessary for the realization of population inversion [13]. Another observation of backward lasing at 337 nm of neutral nitrogen molecules was reported by some of the current authors when nitrogen gas was pumped with circularly polarized 800 nm femtosecond pulses [14-16]. However, the presence of oxygen molecules with the percentage above 13% quenches the

backward emission at 337 nm [14]. Therefore, there exists a strong demand for other methods for backward lasing based on air molecules pumped by optical pulses in a cavity-free manner.

In this paper, we report on backward lasing at 391.4 nm from singly ionized nitrogen molecules, in addition to the previously observed lasing in the forward direction [2, 3, 5-9, 21-23]. This backward lasing signal presents a sensitive dependence on the pump polarization state, with its intensity dropping down by a factor of 100 if the pump laser ellipticity exceeds 0.3. It is found that for linearly polarized pump pulses at 800 nm, the intensity of the backward 391.4 nm signal can be one order of magnitude higher than its neighboring spectral line at 428 nm. This is a clear demonstration of the optical amplification at 391.4 nm in the backward direction. The optical gain is further confirmed by 10 times amplification of an external seeding pulse injected in the backward direction. These observations suggest a new method for the generation of backward lasing from nitrogen molecules. We have investigated the pressure dependence of the backward and forward lasing emission, as well as the sideway fluorescence. An optimal nitrogen pressure about 10 mbar was identified for backward lasing emission, which was attributed to the finite gain life time and the pressure-dependent pump laser intensity inside the gas plasma. We finally compared the backward lasing signal obtained in pure nitrogen and air. It was found that the presence of oxygen molecules strongly depresses the backward lasing action and no lasing signal is obtained in ambient air.

2.   Experimental results and discussion

In the experiments, femtosecond laser pulses (800 nm, 12 mJ, 1 kHz) delivered by a commercial laser system (Elite DUO, Coherent Co. LTD) were focused by a convex lens (f = 300 mm) into a gas chamber filled with air or pure nitrogen at varying pressure, see Fig. 1. A bright plasma channel of 5-10 millimeters was formed, with its length dependent on the pulse energy and gas pressure. The backward emission from the plasma was collected by a fused silica lens (f = 100 mm) behind the dichroic mirror, which reflects the 800 nm femtosecond pump pulse and transmits the backward emission below 450 nm. The backward emission was send into a fiber and analyzed by a spectrometer. The forward emission from the plasma was also detected with the same fiber spectrometer. To filter out the intense pump pulse and the white light generated due to strong nonlinear interaction, two short pass dichroic mirrors (reflective for wavelength above 450 nm) were employed (not shown in the Fig.1) followed by glass filters (BG 39) to further reduce the residual emission around the fundamental wavelength. The fluorescence of the plasma was also measured in the sideway, after collection by an f = 50 mm fused silica lens.
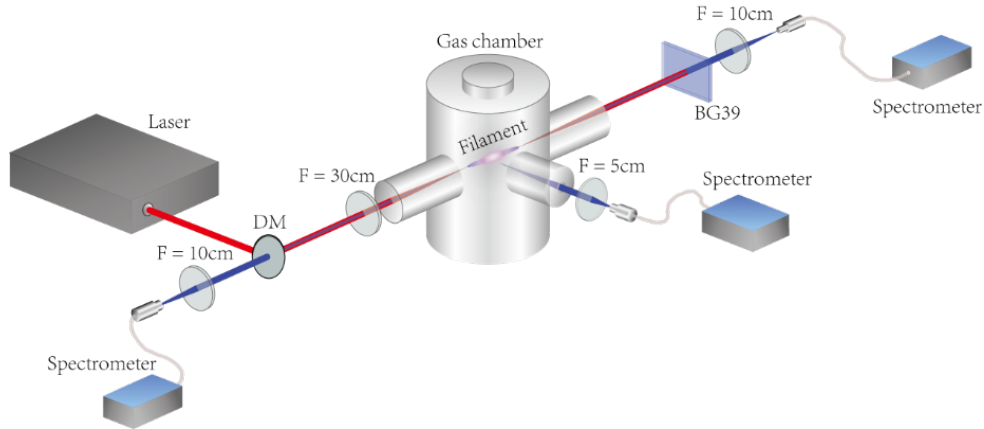

Fig. 1. Experimental setup. The femtosecond laser pulses were focused by the f = 300 mm lens into the gas chamber to form a column of plasma. The emission from the plasma in the backward, forward, and sideway were collected into a fiber connected to a spectrometer for analysis. The short-pass filters (BG 39) were used for detection of the backward and forward emissions. DM: dichroic mirror.

We present in Fig. 2 the spectrum of the side fluorescence, forward, and backward emission for the nitrogen pressure of 11 mbar, with 2.2 mJ pump laser pulse energy. The sideway fluorescence consists of many spectral lines, corresponding to the second positive band ($C^3\Pi_u^+$ to $B^3\Pi_g^+$) of the neutral nitrogen molecules and the first negative band ($B^2\Sigma_u^+$ to $X^2\Sigma_g^+$) of the singly ionized nitrogen ions with different vibrational quantum numbers. These optical transitions are identified in Fig. 2(a). Strong forward emissions centered at 391.4 and 388.5 nm in Fig. 2 (b) correspond to the P and R branches of

the $B^2\Sigma_u^+$ to $X^2\Sigma_g^+$ transition of nitrogen ions. This observation agrees with previous reports of nitrogen ions lasing [2, 4, 21-23]. The backward emission shown in Fig. 2(c) consists of a strong line at 391.4 nm, while other spectral lines at 337, 357, and 427.8 nm are barely observed. This is the first experimental demonstration of backward lasing of nitrogen ions. It is important to note that the ratio of R and P branches of the backward emission (Fig. 2 (c)) is different from that of the forward emission (Fig. 2 (b)), confirming that the backward signal is not due to the unwanted reflection of the forward lasing beam on the exit window of the gas chamber. Based on a comparison between the spectral intensity at 391.4 nm and 399 nm of fluorescence signal (Fig. 2(a)) and backward lasing (Fig. 2(c)), we estimated that the fluorescence contributes to about 12% of signal intensity in the backward lasing signal in Fig. 2(c).
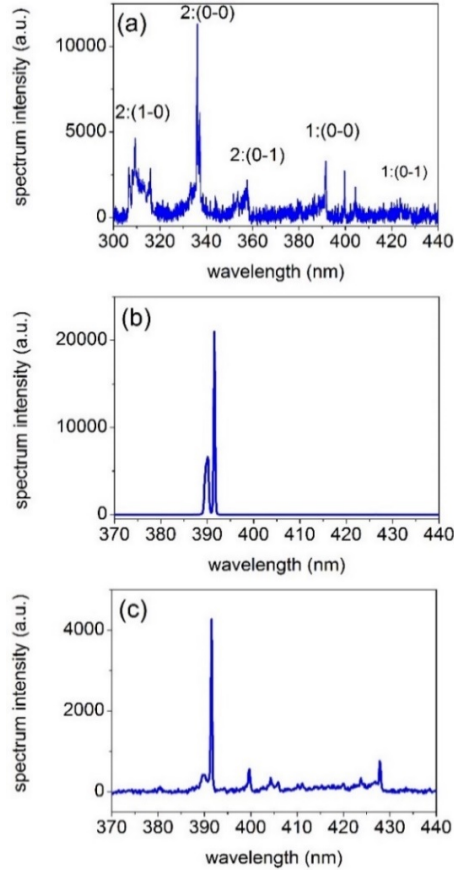

Fig. 2. Spectrum of the side fluorescence (a), forward (b) and backward (c) signal from the nitrogen plasma. The pump laser energy is 2.2 mJ and the gas pressure is 11 mbar. In (a), the number 1(2) before the parenthesis refers to the first negative (second positive) band of the nitrogen ions (neutral nitrogen molecules). The numbers inside the parenthesis presents the quantum number of vibrational levels for the upper state and lower state.

To verify that the nitrogen ions indeed give rise to optical amplification in the backward direction, we have performed a pump-probe experiment with another femtosecond laser system (100 Hz, 40 fs, 15 mJ, 800 nm). Similar to the setup in ref. 15, a weak probe beam with central wavelength at 391 nm generated in a BBO crystal was injected in the opposite direction of the pump laser. Here, the pump pulse duration was 40 fs and its energy was 7.5 mJ. The pump was focused by a convex lens f = 400 mm in pure nitrogen at 30 mbar. The experimental results are presented in Fig. 3. In this experiment, the pump pulse itself does not generate significant backward 391.4 nm signal due to loose focusing geometry and gas pressure deviated from the optimal value. In Fig. 3, we found that the backward-propagating seed pulse is amplified by a factor of 10 at 391.4 nm, which demonstrates undoubtedly optical amplification in the backward direction.
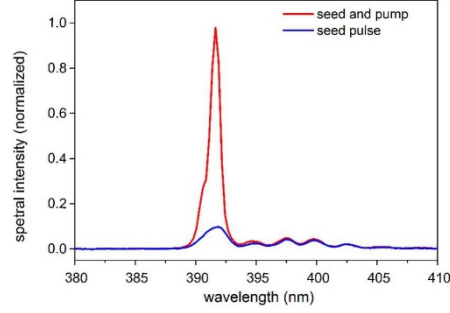
Fig. 3. Amplification of the backward-propagating seeding pulse in a plasma. Blue line: seeding pulse, red line: the amplified emission in presence of both the pump laser and seeding pulse. The energy of the pump laser is 7.5 mJ, it was focused in 30 mbar nitrogen gas with a convex lens f = 400 mm.

We further measured the gas pressure dependence of the backward 391.4 nm signal in pure nitrogen. The experimental conditions are the same as in Fig. 2. The results are presented in Fig. 4, together with those of the forward signal. With increasing gas pressure, both the backward and forward 391.4 nm signals become more intense up to an optimal pressure. The signals decrease at higher pressures. Similar pressure dependence of the forward 391.4 nm lasing signal has been reported in Ref. 4 and it was attributed to nonlinear propagation effects.
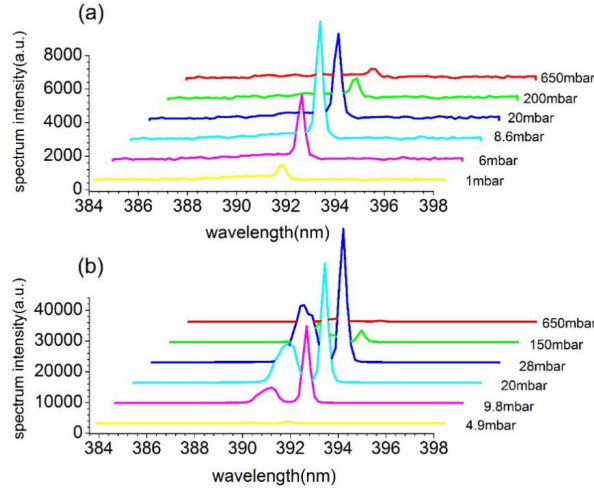

Fig. 4. Pressure dependence of the backward (a) and forward (b) lasing emission from nitrogen. The pump pulse energy was 2.2 mJ.

In order to get further insight into the gas pressure dependence, we compared the intensity of the backward and forward 391.4 nm emissions to the corresponding sideway fluorescence signal in Fig. 5. There are three particular features that deserve our attention. First, the three signals present a rapid increase in the pressure range below 10 mbar. In this low gas pressure regime, the laser pulse propagates almost linearly and the laser intensity is independent of the gas pressure. Considering a Gaussian beam with width of 12 mm ($1/e^2$) focused by an f = 300 mm lens, the size of the focus is found to be $2w_0$ = 31.9 μm, with M^2 = 1.25 the beam quality factor. For a 35 fs pulse with incident energy of 2.2 mJ, the peak laser intensity reads as 2.32×10^{15}W/cm^2. In this linear propagation regime, the plasma density and the density of the ions in the excited states are almost linearly proportional to the gas pressure. This explains largely the rapid increase of the three curves up to gas pressure close to 10 mbar.
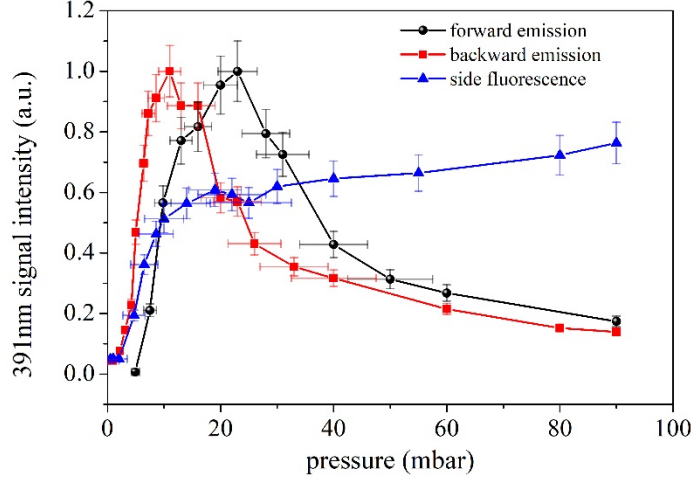
Fig. 5. Intensity of the sideway fluorescence at 391. 4 nm (black dot), the forward lasing signal (blue dot) and the backward 391.4 nm emission (red dot) as a function of the gas pressure. The pump laser energy was 2.2 mJ.

The second feature is that the sideway fluorescence is strongly saturated for gas pressures exceeding 20 mbar, while the forward 391.4 nm signal decreases. The sideway fluorescence signal origins from the excited nitrogen ions in the $B^2\Sigma_u^+$ state, and it therefore reflects the density of the plasma. Based on this observation, we argue that plasma density is strongly saturated for pressures above 20 mbar, which indicates that the ionization probability $\rho_{ion}/\rho_{neutral}$ of the neutral nitrogen molecules decreases in this pressure range. Since the ionization probability depends on the local laser intensity, we deduce that the laser intensity in plasma decreases progressively for the pressure range 20-100 mbar due to the plasma defocusing effect. This is expected since in the filamentation regime in atmospheric air the laser intensity is around 4.5×10^{13} to 1.5×10^{14}W/cm^2[24-26], much less than that in the linear propagation case. Therefore, the laser intensity is expected to decrease from 2.32×10^{15} W/cm^2 to $\sim10^{14}$ W/cm^2 with the gradual increase of gas pressure. This argumentation explains naturally the fact that the forward lasing signal decreases for pressures above 20 mbar, since the optical gain of the $B^2\Sigma_u^+$ to $X^2\Sigma_g^+$ transition depends on the laser intensity and there exists an intensity threshold for lasing action [22].

To further confirm the above argument, we have performed numerical simulations for pump pulse nonlinear propagation in pure nitrogen gas at different gas pressures. We have employed the laser pulse nonlinear propagation modeling and numerical code developed in Ref. [27, 28]. The parameters for simulations are close to those of the experiments. In Fig. 6, we present the pulse intensity and plasma density as a function of propagation distance for nitrogen pressure ranging from 5 mbar to 300 mbar. In the case of 10 mbar nitrogen, the peak laser intensity was found to be around 2×10^{14}W/cm^2. For increasing pressure, it decreases progressively to 1×10^{14}W/cm^2 for nitrogen pressure of 1 bar, which agrees well with the above qualitative analysis.
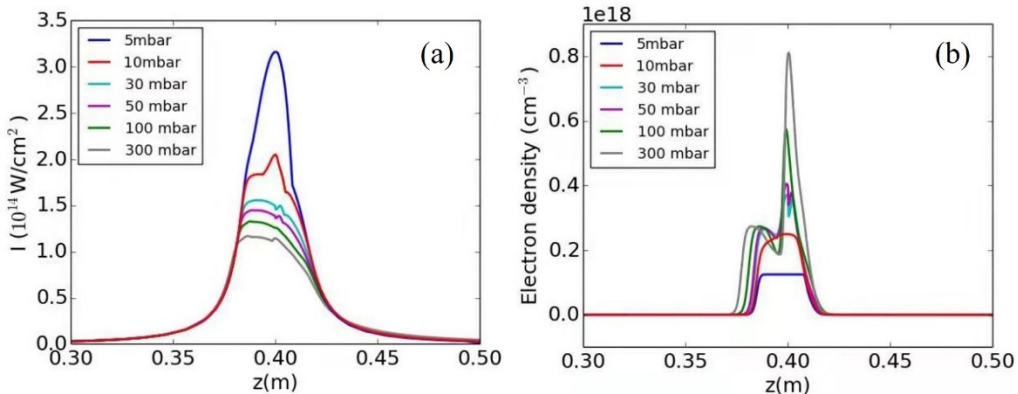

Fig. 6. Numerical simulation of pump pulse peak intensity (a) and plasma density (b) as a function of propagation distance in nitrogen gas. The input pulse (35fs, 1.5mJ) is focused by a f = 40 cm lens.

The third feature in Fig. 5 is that the optimal gas pressure for backward lasing, around 10 mbar, is smaller than that for forward lasing (20 mbar). This confirms again that the backward signal is not due to the reflection of the forward 391.4 nm lasing beam, since otherwise they should present the same pressure

dependence. How should we understand this difference in optimal gas pressure? In a laser amplifier, the output amplified signal in the small signal regime is an exponential function of the length l, that is $I = I_0 \exp(gl)$, where g is the optical gain. For the forward emission, the length of the amplifier is equal to the geometrical length of the plasma, since a forward-propagating photon experiences the same optical gain along the plasma. Therefore, the optical gain g increases for the pressure range 4-20 mbar, while the gain length l is almost constant. In contrast, for the photons in the backward direction, the effective length of optical amplification is determined by the lifetime τ_g of the optical gain, $l_{back} = c\tau_g$ [15, 16], which is shown to decrease for higher pressures [9]. As a result, the product of the optical gain and the effective amplification length, gl_{back}, depends on the pressure in a more complex manner. For pressure range between 4-20 mbar, the optical gain is increasing, while the effective length of the gain decreases gradually. This explains why the maximum of the gain length product is achieved at a gas pressure below 20 mbar.

Finally, we examined the role of oxygen on the backward lasing action. The experimental results obtained in 11 mbar nitrogen and air are compared in Fig. 7. Unfortunately, it was observed that the backward 391.4 nm signal in air drops down by a factor of 9 compared to that in pure nitrogen. The intensity of the 391.4nm signal becomes comparable to that of the 427.8 nm signal, indicating that the 391.4 nm radiation is now of the nature of fluorescence instead of stimulated emission. Therefore, it is still difficult to obtain backward lasing emission in ambient air with this mechanism.
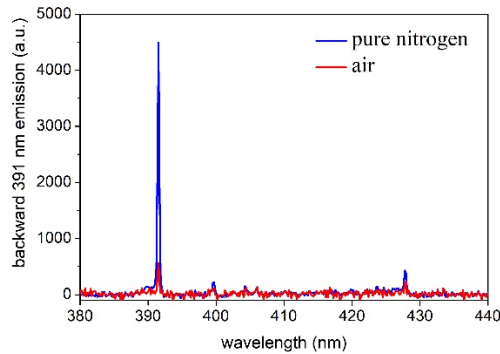

Fig. 7. Backward spectrum signal obtained in pure nitrogen and air at pressures of 11 mbar. The pump pulse energy was 2.2 mJ.

3. Conclusion

In conclusion, we have shown that the singly ionized nitrogen molecules pumped by intense 800 nm femtosecond pulses can give rise to coherent emission at 391.4 nm in the backward direction, in addition to the widely reported forward lasing radiation at the same wavelength. With the current focusing geometry, the optimal nitrogen gas pressure was found to be around 10 mbar, less than that of the forward radiation. This is explained by the fact that the effective length of optical amplification depends on the gas pressure for backward propagating photons. We compared the backward signal in low pressure pure nitrogen and air. It was found that the presence of oxygen molecules strongly depresses the backward emission and the stimulated emission retrogrades to fluorescence.

6

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