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# 氧气对飞秒激光泵浦的氮气分子无腔激光效应的淬灭作用(特邀)

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**摘 要**:圆偏振飞秒激光泵浦氮气产生的无谐振腔激光效应在远程光学遥感方面具有非常重要的应用前景.然而,空气中氧气分子的存在显著地淬灭了该受激辐射效应.对比研究了氧气和氪、氩、氦这三种气体对于氮气分子前向和背向激射的影响,测量了纯氮气和空气中氮分子荧光的强度.结果表明,氧气和氪气这两种电离能非常接近的气体呈现出十分相似的淬灭作用;而氦气因为电离能极大,并不呈现出显著的淬灭作用.因此,可得氧气对于氮气分子激射淬灭作用的主要原因在于其引起的光丝内激光强度的下降,从而使得自由电子能量降低,导致碰撞激发效率下降.

**关键词**:非线性光学;空气激光;超快光谱技术;淬灭效应;飞秒脉冲

**中图分类号**:TN241;TN248.2 **文献标识码**:A **doi**:10.3788/gzxb20204911.1149013

# Quenching Effect of $O_2$ on Cavity-free Lasing of $N_2$ Pumped by Femtosecond Laser Pulses (Invited)

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**Abstract**: Nitrogen gas pumped by circularly polarized femtosecond laser pulses gives rise to bidirectional lasing emission, which holds unique potential for remote optical sensing application. However, the presence of oxygen molecules strongly suppresses this lasing effect. The influence of $O_2$, Kr, Ar and He on the lasing effect of nitrogen molecules was compared, and the fluorescence of nitrogen molecules in pure nitrogen and ambient air was examined. It is observed that the lasing presents a similar quenching effect

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with the partial pressure of $O_2$ and Kr, since Kr has a close ionization potential compared to $O_2$. In contrast, for He which has a much higher ionization potential, there is no significant quenching effect. Therefore it is suggested that the quenching effect of $O_2$ on nitrogen molecules mainly stems from the fact that $O_2$ leads to a reduction of the clamped laser intensity inside the plasma filaments, which results in a decrease of the kinetic energy of the free electrons and an inefficient collision excitation.

**Key words**: Nonlinear optics; Air lasing; Ultrafast spectroscopic technology; Quenching effect; Femtosecond pulses

**OCIS Codes**: 320.7120; 140.4130; 190.7110

## 0 Introduction

Ambient air can be turned into an optical gain medium under excitation with intense ultrafast lasers[1-5]. This cavity-free lasing action, coined as "air lasing", has attracted much attentions in recent years due to its unique potential to generate a coherent optical beam propagating from the sky to the ground observer[1,5-7]. With such a backward optical beam propagating to the earth, coherent optical spectroscopy such as stimulate Raman scattering can be used for remote sensing of trace elements in the atmosphere[8]. This could lead to revolutionary improvement of the optical remote sensing sensitivity, because information of the target molecules is now carried by the directional backward air laser beam, not by the fluorescence or spontaneous scattered photons emitted in the $4\pi$ solid angle as in most traditional optical remote sensing technique.

Up to now, all the three major constituents of air ($N_2$, $O_2$, Ar) have been demonstrated to be able to serve as gain medium for cavity-free lasing under excitation with properly chosen pump laser wavelength[9]. The activation of both $O_2$ and Ar lasing action depends on resonant excitation of the photon-dissociated oxygen atoms or Ar atoms, and requires a pump laser in the extreme UV range[1,7,10]. The strong attenuation of EUV beam during propagation in atmosphere prevents long distance creation of such air lasing. The cavity-free lasing of neutral nitrogen molecules in a $N_2$-Ar gas mixture by pumping with mid-IR pulses (3.9 μm or 1.03 μm) was first reported by KARTASHOV D and co-workers[5]. Later, similar results in $N_2$-Ar gas mixture were obtained with pump laser at 800 nm[11]. The emission line at 337 nm corresponds to the $C^3\Pi_u^+$-$B^3\Pi_g^+$ transition of the excited neutral nitrogen molecules. Unfortunately, high pressure Ar gas was necessary since the population inversion between the $C^3\Pi_u^+$ and $B^3\Pi_g^+$ state is achieved via collision with excited Ar atoms. Therefore, this scheme of backward lasing can not be utilized for application in ambient air.

MITRYUKOVSKIY S, et al. employed intense circularly polarized 800 nm femtosecond laser pulses and observed a bidirectional lasing emission at 337 nm from neutral nitrogen molecules[6]. The underlying mechanism of population inversion between the $C^3\Pi_u^+$ and $B^3\Pi_g^+$ states was attributed to collisional excitation of the ground state ($X^1\Sigma_g^+$) neutral $N_2$ molecules by energetic electrons, $N_2(X^1\Sigma_g^+)+e=N_2(C^3\Pi_u^+)+e$. In this scheme, circular laser polarization is crucial since the kinetic energy of the electrons after laser-gas interaction depends on the polarization state of lasers, where electrons with energy up to $2U_p$ can be generated in case of circularly polarized pump pulses[12-13]. Here, $U_p=e^2/c\varepsilon_0 m_e\times I/2\omega_0^2$ is the ponderomotive energy of the electron in the laser field with the electron charge $e$, vacuum permittivity $\varepsilon_0$, the mass of the electron $m_e$, the intensity $I$ and frequency of the laser field $\omega_0$. For laser intensity of $I=1.5\times10^{14}$ W/cm$^2$, the ponderomotive energy is about 9 eV, sufficient to achieve population inversion between $C^3\Pi_u^+$ and $B^3\Pi_g^+$. Unfortunately, it was found that the presence of oxygen molecules strongly suppresses lasing effect. The forward 337 nm lasing emission in ambient air decreases to 1% of that obtained in pure nitrogen[13]. For the backward emission, MITRYUKOVSKIY S and co-workers reported that it is totally suppressed when the pressure of oxygen exceeds 13% [6], while in ambient air the concentration of oxygen gas is close to 21%. Later, a similar quenching effect was reported by YAO J, et al.[14]. As to the underlying mechanism of quenching effect, it was first suggested that the collisional dissociation of oxygen molecules by the excited neutral can play a role, $N_2(C^3\Pi_u^+)+O_2=N_2(X^1\Sigma_g^+)+O+O$[6]. Later, it was pointed out that the laser intensity decrease due to presence of oxygen can also be responsible[14]. Up to now, the mechanism of this quenching effect is not yet clear, which prevents the realization and optimization of backward lasing of $N_2$ in ambient air.

In this paper, we report on a comparative study of the influence of different gases on the backward lasing intensity of $N_2$. Three noble gases, Kr, Ar and He, are chosen since Kr has a similar ionization potential to $O_2$

while He is immune for ionization in this laser intensity range due to its higher ionization potential. For $O_2$ and Kr, similar quenching behaviors for increasing pressure are observed. In contrast, the lasing intensity keeps almost constant for He pressure up to 200 mbar. Analytic estimation reveals that the clamped laser intensity inside filaments in presence of $O_2$ or Kr is much less than that in pure nitrogen, due to their relatively smaller ionization potential energy. Therefore, we conclude that the quenching effect of $O_2$ mainly roots in the reduced laser intensity due to its presence inside filaments.

## 1 Experimental setup

In the experiment, the femtosecond laser pulses (800 nm, 35 fs, 1 kHz) delivered by a commercial Ti: sapphire chirped pulse amplification system (Coherent, Legend DUO) have a maximum pulse energy of 13 mJ. The experimental setup is schematically shown in Fig.1. A quarter wave-plate was installed on the beam path to change its polarization from linearly to circularly polarized states. The pump pulses pass through a Dichroic Mirror (DM: reflective for 800 nm, transparent for 337 nm beam) and then is focused with an $f$=750 mm lens into a gas chamber filled with pure nitrogen gas or its mixture with other gases. The pump pulses create a ~60 mm long bright plasma filament, which produces 337 nm lasing emission in both the forward and backward directions. The 337 nm emission was spectrally filtered out from the residual 800 nm pulse and the accompanying white light with BG 39 glass filter (high transmission in the range of 335~610 nm) and an interference filter of 337 nm (10 nm bandwidth). Finally, a convex lens with $f$=100 mm was used to collect the emission into the fiber spectrometer. The backward 337 nm emission from the plasma filament was similarly collected into the fiber spectrometer after a dichromatic mirror, a BG 39 filter and a 337 nm interference filter.

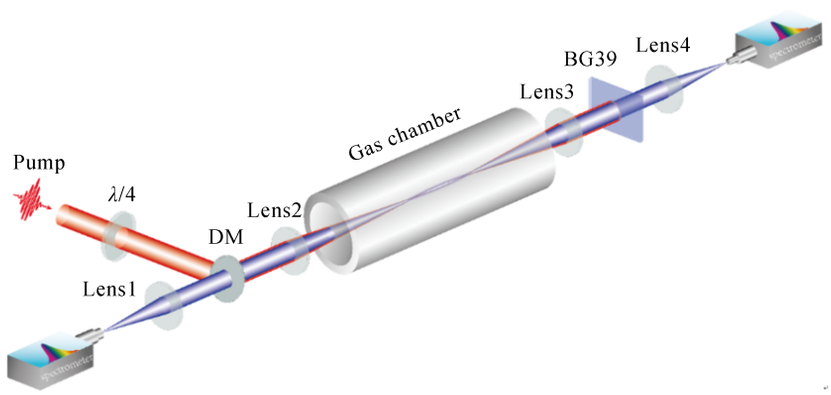


Fig. 1 Schematic experimental setup

## 2 Experimental results

### 2.1 Lasing emission at 337 nm from neutral nitrogen molecules

We first used circularly polarized 800 nm pulses to pump pure nitrogen gas at 1 bar pressure and optimized the 337 nm lasing emission in the forward and backward directions through careful adjustment of the rotation angle of the $\lambda/4$ wave-plate, as well as the azimuthal angle of the convex lens to compensate the slight astigmatism of the incident laser beam. The spectra of the forward and backward lasing emission are shown in Fig.2. It is seen that backward 337 nm emission intensity of the nitrogen molecule is much weaker compared to the forward signal. To assure this backward emission origins directly from the filamentary plasma, we need to exclude the reflection of forward 337 nm emission on the exiting fused silica window. In our experiments, we used a window with Brewster angle. Therefore, the reflected beam from the window cannot return into the detection fiber, which excludes the possibility that the backward signal is the reflected forward emission.

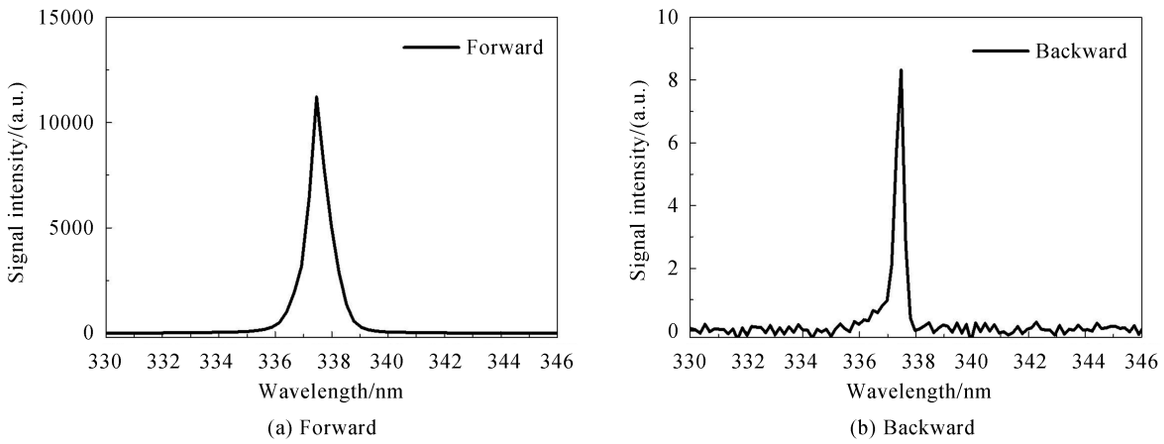


Fig. 2 Spectra of nitrogen molecule lasing emission for nitrogen gas plasma pumped by circularly polarized laser pulses of 12 mJ

### 2.2 Effects of different gases on the nitrogen molecule lasing emission

In the experiment, we fixed the nitrogen pressure at 800 mbar and measured the 337 nm signal intensity as a function of oxygen gas pressure from 20 mbar to 200 mbar (measured every 20 mbar). The experimental results for the forward 337 nm emission are shown in Fig. 3 (a). We noticed that $O_2$ gas presents a strong quenching effect on the 337 nm lasing emission of nitrogen molecules. For $O_2$ pressure higher than 40 mbar, the intensity of the forward 337 nm emission reduces to 10% of that in pure nitrogen, which agrees with previous reports[6,14]. The experimental results for Kr, Ar and He are presented in Fig. 3 (b) ~ (d). Notice that the intensity of forward 337 nm signal decreases significantly with increase of the Kr pressure, in a similar tendency compared to that of $O_2$. In contrast, the introduction of He into $N_2$ just results in a slight decrease of the 337 nm

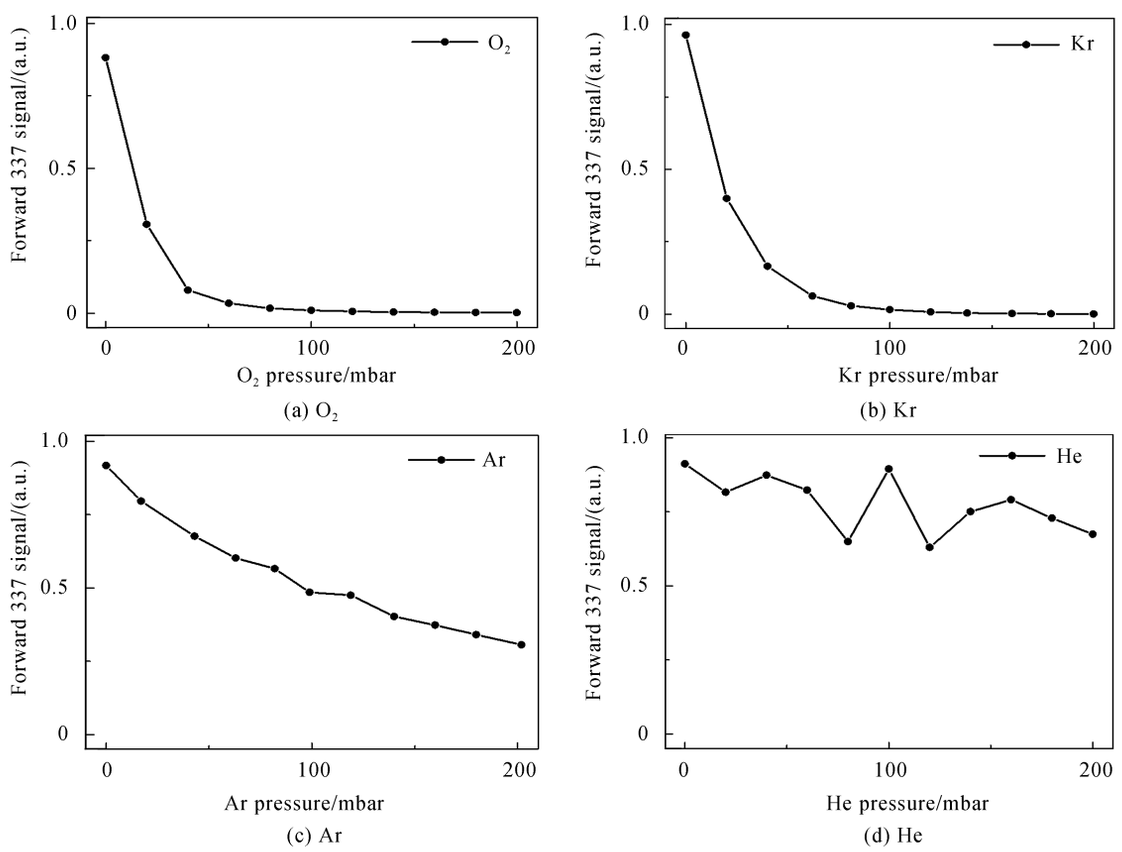


Fig. 3 Measured forward 337 nm lasing signal as a function of partial gas pressure of different gases. The pressure of the nitrogen is kept at 800 mbar

lasing signal up to a gas pressure of 200 mbar, as shown in Fig.3(d). For Ar gas, which has an intermediate ionization potential between Kr and He, the quenching effect is less severe compared to $O_2$, where the lasing intensity decreases to 10% at Ar gas pressure of ~200 mbar. Here we would like to point out that in Ref. [11] introduction of high pressure Ar into nitrogen gas leads to increase of the 337 nm signal[11]. We believe that the different observations origins from the entirely distinct experimental conditions of ours (800 mbar $N_2$, 0~200 mbar Ar) compared to that of Ref. [11] (300 mbar $N_2$, 900 mbar Ar).

Similar experiments were performed for the backward 337 nm emission and the results are presented in Fig.4. Strong quenching effects were observed for $O_2$ and Kr. For He gases, the backward 337 nm emission remains almost unchanged with pressure up to 200 mbar.

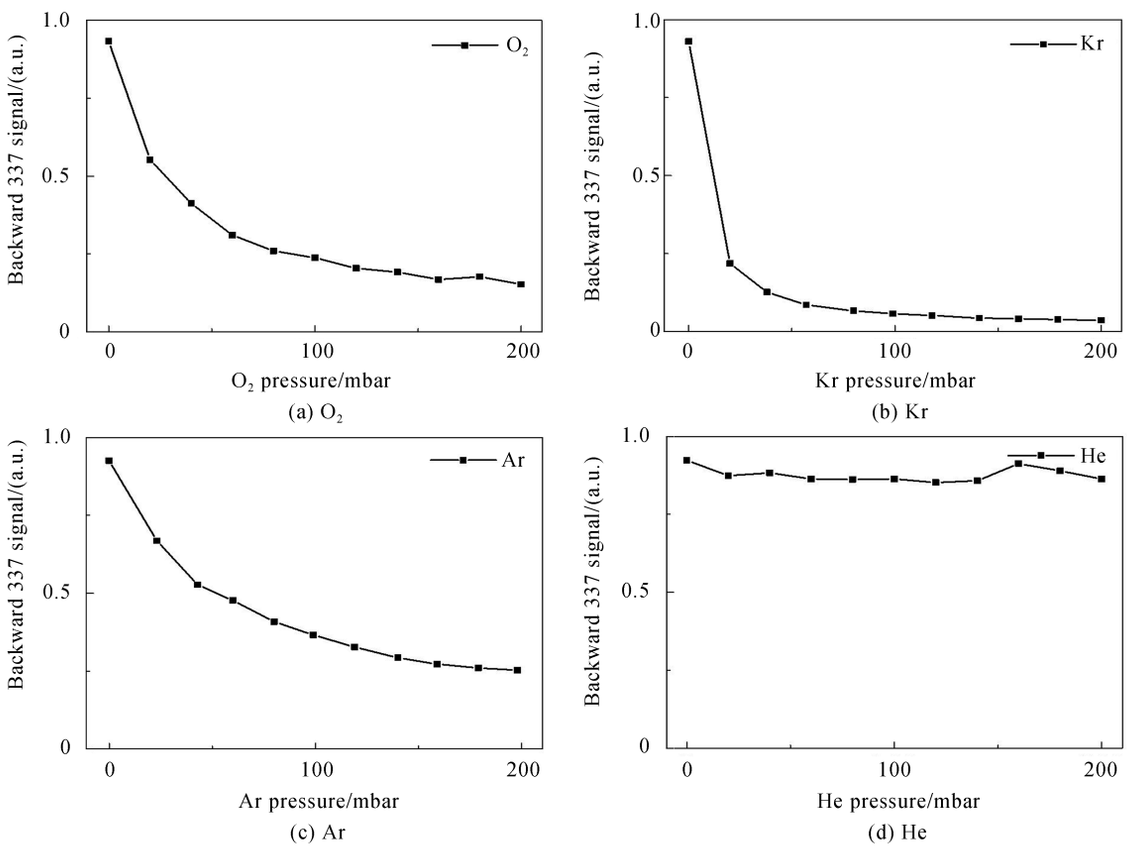


Fig. 4 Measured backward 337 nm lasing signal as a function of partial gas pressure of different gases. The pressure of the nitrogen is kept at 800 mbar

### 2.3 Comparison of the plasma fluorescence in pure nitrogen gas and in air

In the above, we observed that the introduction of oxygen in nitrogen gas leads to gradual decrease of both backward and forward lasing signal. This indicates that the density of population inversion is reduced when we compare air to pure nitrogen. We therefore examined the molecule density change in the upper level $C^3\Pi_u^+$ state by observing the fluorescence signal of the excited neutral nitrogen molecules, since the fluorescence signal at 337 nm is directly proportional to the molecule density in the $C^3\Pi_u^+$ state. The experimental result is presented in Fig.5. We found that the 337.4 nm fluorescence signal in air decreases to 20% of that obtained in pure nitrogen. So, it is clear that the presence of oxygen molecules results in a strong decrease of the molecule density in the $C^3\Pi_u^+$ state.

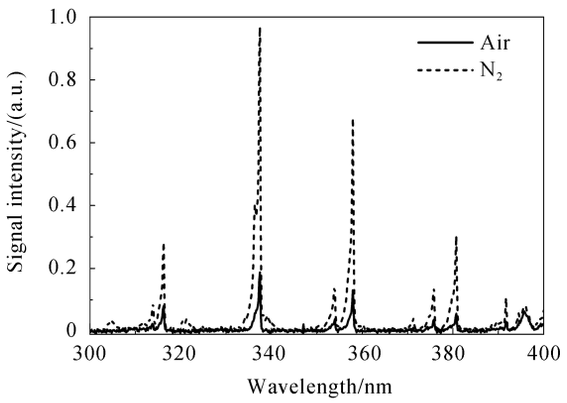


Fig. 5 Comparison of the fluorescence of nitrogen in air and pure nitrogen gas pumped by femtosecond laser pulses

## 3 Discussion

The quenching effect of $O_2$ has been discussed qualitatively in the previous works and several possible mechanisms have been suggested[6,11,14]. There exist three possible mechanisms for this quenching effect.

1) In the first report of $N_2$ lasing pumped by circularly polarized 800 nm pulses, the authors mentioned that the collisions between $N_2(C^3\Pi_u^+)$ and the $O_2$ molecules in ground state can cause the dissociation of oxygen molecules. As a result, the excited nitrogen molecules return to the ground state without radiation. This can lead to decrease of the concentration of excited nitrogen molecules in the $C^3\Pi_u^+$ state, which in turn reduces the lasing intensity[6].

2) Inside the filaments formed in ambient air, energetic electrons frequently collide with oxygen molecules, which can lead to vibrational and rotational excitation or dissociation of oxygen molecules[15]. These inelastic collisions result in rapid drop of the average energy of the electrons. Consequently, the collisional pumping of the ground state nitrogen to the $C^3\Pi_u^+$ state becomes less efficient, resulting in the quenching of nitrogen lasing emission.

3) It is known that the clamped laser intensity in the filament will decrease when $O_2$ is mixed into $N_2$ gas because of the relative lower ionization potential of $O_2$ ($U_i$=12.1 eV) compared to that of $N_2$ ($U_i$=15.6 eV)[16]. Therefore, one expects that the kinetic energy of the electrons will be reduced since it is directly proportional to the laser intensity. This can result in less efficient collisional excitation and thus quenching effect.

With the above experimental results in hand, we can now examine the role of these three mechanisms. First, we consider the collisional dissociation of the $O_2$ by collisions with excited $N_2$ ($C^3\Pi_u^+$). It is now well known that the lasing dynamics of $N_2$ occurs on the time scale of 10～100 ps[13,17]. In contrast, the mean collision frequency between molecules in 1 bar air is known to be $\nu=7\times10^9\,s^{-1}$, corresponding to a mean collision time of 150 ps. The characteristic time scale of the $N_2$ lasing is even less than the mean collision time between molecules. Therefore, we believe that the influence of the first mechanism should be negligible.

For the second and third possible mechanisms, the comparison between Kr, Ar, He and $O_2$ provides important insight. The ionization energy of Kr ($U_i$=13.9 eV) is close to that of $O_2$, which ensures that the laser intensity in the filament remains roughly identical when same concentrations of Kr or $O_2$ are mixed into $N_2$ gas. Consequently, the electron density and the electron energy distribution are similar in these two situations. On the other hand, Kr is a monoatomic molecule which has no rotation and vibration degrees of freedom. So, the collision of electrons with argon molecules is basically elastic, and the energy of electrons will not change after collisions. Therefore, similar tendencies in Fig.3(a) and Fig.3(b) indicate the relaxation of electron energy due to collision with $O_2$ should not play an essential role for the quenching effect, otherwise no significant quenching should be observed in Kr.

Now, we discuss the third mechanism. The laser intensity inside the femtosecond filament can be estimated by considering a balance between the Kerr self-focusing and the plasma induced defocusing, $n_2 I = \rho(I)/2\rho_c$[18]. Here, $n_2$, $\rho(I)$ and $\rho_c$ are respectively the nonlinear refractive of air, the intensity-dependent plasma density, and the critical plasma density. In air, one can roughly estimate the plasma density as

$\rho(I)\sim\sigma_K I^K\rho_0\tau_p$, where $\sigma_K$, $K$, $\rho_0$, and $\tau_p$ denote the multiple photon ionization coefficient, the minimum number of photons necessary for Multiple Photon Ionization (MPI), the density of neutral molecules, and the incident pulse duration, respectively. The laser intensity can be estimated as $I\sim\left(\frac{2n_2\rho_c}{\sigma_K\tau_p\rho_0}\right)^{K-1}$. The nonlinear refractive index for $O_2$ and $N_2$ are similar and its role can be neglected. Considering the ionization potential of $O_2$ and $N_2$ and the photon energy of the 800 nm pump laser ($\hbar\omega=1.5$ eV), the minimum numbers of photons necessary for MPI of $O_2$ and $N_2$ are respectively $K_{O_2}=8$ and $K_{N_2}=11$. The MPI coefficient for $O_2$ and $N_2$ are known to be $\sigma_8=2.8\times10^{-96}\,s^{-1}cm^{16}/W^8$ and $\sigma_{11}=6\times10^{-140}\,s^{-1}cm^{22}/W^{11}$[18]. As a result, the ratio of the clamped laser intensity inside the filament in $O_2$ and $N_2$ gases read as $I_{O_2}/I_{N_2}=\sigma_{11}^{1/10}/\sigma_8^{1/7}=0.54$. Here, we see that the clamped laser intensity inside the filament experiences significant decrease when $O_2$ is mixed inside $N_2$ gas. We would like to point out that this decrease of laser intensity has been reported before through numerical simulations[19]. The decrease of the laser intensity directly leads to lower kinetic energy of the electrons. As a result, the collisional excitation of $N_2\left(X^1\Sigma_g^+\right)$ becomes ineffective when the electron energy becomes less than the threshold energy $E_{th}$ (14 eV). It is now clear that the quenching effect of $O_2$ mainly origins from the fact that it lowers the clamped laser intensity inside the filaments, leading to less energetic electron generation and thus inefficient collisional excitation of the $N_2$ responsible for population inversion. With mixture of He into $N_2$, the clamped laser intensity is almost unchanged due to the much higher ionization potential of He ($U_i=24.5$ eV). As a result, the presence of He has negligible influence on the 337 nm lasing intensity. In view of this mechanism for the quenching effect, one possible method to overcome this detrimental effect is to increase the clamped laser intensity by using tighter focusing geometry. At the same time, a reasonable plasma gain length should be also maintained for sufficient optical amplification. Therefore, we believe that optimization of the focusing geometry and employment of more powerful femtosecond laser pulses should be possible solutions to achieve backward lasing of $N_2$ in ambient air.

## 4 Conclusion

We studied the mechanism of the quenching effect of $O_2$ on nitrogen molecule lasing emission. In the experiment, we first optimized the 337 nm signal in the forward and backward directions from the nitrogen gas plasma. We then mixed different noble gases with different ionization potential at variable pressures into $N_2$ gas to systematically study the effects of different gases on the nitrogen molecular lasing emission. Through comparison of the influence of $O_2$, Kr, Ar, and He on the lasing emission of nitrogen molecules, we found that the decrease of the clamped laser intensity in the filament should be the most important reason for the quenching effect. This understanding suggests that to overcome the quenching effect of $O_2$ molecules and to achieve the nitrogen molecule lasing under atmospheric conditions, the possible solutions is to use tighter focusing conditions and higher energy pump pulses to compensate the decrease of laser intensity in the filaments.

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