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Theoretical treatment on externally-seeded superradiance from N_2^+ in femtosecond laser filamentation in low-pressure nitrogen

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We perform a theoretical investigation of a collective emission in tunnel-ionized nitrogen molecules triggered by a coherent seed pulse. A semiclassical theory of superradiance that includes the superradiant temporal profile, characteristic duration, time delay, intensity is achieved. The theoretical predictions of 391-nm forward emission corresponding to the transition between $N_2^+(B^2\Sigma_u^+, \nu' = 0)$ and $N_2^+(X^2\Sigma_g^+, \nu = 0)$ as a function of nitrogen gas pressure are compared with the recent experimental data [Ding P. *Lasing effect in femtosecond filaments in air*, Ph.D. thesis, Université Paris-Saclay (2016)]. The good agreement demonstrates that the time-delayed optical amplification inside the molecular nitrogen ions is superradiance.

KEYWORDS

optical bloch equations, superradiance (SR), strong-laser-field ionization, 391-nm lasing of N_2^+ , retarded optical amplification

1 Introduction

Tunnel ionization is a fundamental process for atoms/molecules in intense laser fields when the Keldysh parameter is less than unity [1, 2]. In the tunneling regime, not only sequential double ionization but also non-sequential double ionization exist [3, 4]. Tunnel ionization is taken as a foundation to understand other strong-field phenomena including high harmonic generation [5, 6], non-linear filamentation and laser-induced electron acceleration [7–9]. Because of the multielectron effects and the close ionization potentials between the outermost and a few lower-lying orbitals, the electrons from several molecular orbitals can be ionized in strong laser fields [10–12]. A direct consequence of the multiorbital ionization is that the molecular ions are populated not only in the ground but also in the excited electronic states. A good example is the molecular nitrogen ions inside the plasma generated by 800-nm femtosecond laser pulses, which is the subject of interest in this study. The fluorescence measurement of the plasma confirm that the molecular nitrogen ions exist in both ground $N_2^+(X^2\Sigma_g^+)$ and excited $N_2^+(B^2\Sigma_u^+)$ electronic states [13, 14]. For convenience, $B^2\Sigma_u^+$ and $X^2\Sigma_g^+$ will be termed as *B* and *X* in the following context, respectively.

Since the ionization rate decays exponentially with the electron binding energy, the populations in the excited states should be much less than those in the ground state, e.g., no population inversion should be expected between B and X state. However, it has been experimentally demonstrated that N_2^+ lasing at 391 and 428 nm wavelength can be generated and an externally-injected weak seeding pulse can be significantly amplified in laser filamentation in nitrogen gas, indicating the presence of high optical gain in the system [15–39]. To explain the seed amplification inside the molecular nitrogen ions, Xu et al., Yao et al. and Zhang et al. proposed multi-state coupling model [23, 25, 40]. The nitrogen molecules are populated in the lowest three electronic states of molecular nitrogen ions, i.e., X, A ($N_2^+(A^2\Pi_u)$) and B, by tunnel ionization at the peak of the 800-nm pump pulse. The falling edge of the 800-nm pulse, which can not cause further ionization though, still has high intensity. The two-level system of X and A with a transition wavelength of 787 nm experiences Rabi oscillation in the 800-nm pump laser field. As Rabi oscillation only occurs at the falling edge rather than the full laser envelope, the population transfers efficiently from X to A as a result of the couplings induced by the 800-nm pump pulse, which helps to establish the population inversion between B and X.

An important issue of radiation research is the time evolution of the radiated intensity of the system. Time-resolved measurements of N_2^+ lasing show that the external seeding pulse is almost unaffected after passing through the plasma, instead it triggers a retarded emission at the lasing wavelength [21, 22, 24, 29]. The intensity of the retarded emission increases gradually and reaches its peak value at a time delay τ_D of several picoseconds after the seeding pulse [22, 24]. The characteristic duration τ_W of the emission shares the same magnitude as τ_D [22, 24]. Within particular pressure range from few mbar to optimal pressure, it was found that the τ_D and τ_W are approximately proportional to the inverse of the gas pressures while the peak intensity of the retarded lasing scales as the square of the gas pressure, which coincides with the features of superradiance and therefore suggests the presence of macroscopic dipole (or molecular coherence) in N_2^+ lasing. Following experimental investigations have unquestionably confirmed the presence of molecular coherence between N_2^+ cations [28, 30]. Recently, by considering a three-level V-scheme system, a model of lasing without population inversion has been proposed based on the long-lasting coherences in $A \rightarrow X$ and $B \rightarrow X$ transitions to explain the time-delayed optical gain [29]. The model ascribes the long-lasting coherence in $A \rightarrow X$ transition to the post-pulse of the 800-nm pump pulse based on high contrast cross-correlator measurements.

However, there still exists an understanding gap between the presence of molecular coherence in N_2^+ and the temporal behaviors of the lasing pulse. Regardless of the underlying mechanism of optical gain in N_2^+ , it would be of important interest to understand the evolution of the lasing process under the circumstance of a macroscopic dipole of the excited N_2^+ . We noticed that Xie et al. have experimentally and numerically investigated the evolution of a weak seed pulse in the population-inverted N_2^+ system at the pressure of 200 mbar [41]. With the short dipole-dephasing time of 2.5 ps, they found multiple amplification routes including superradiance, seed amplification induced by the stimulated emission and free induction decay. In our article, the relative low pressures of nitrogen gas are used and the dipole-dephasing time is set to be 0. We deduce analytic expressions for temporal characteristics of the N_2^+

lasing pulse based on a semi-classical theory considering only the two-level system of X and B states. The obtained analytic results show clearly how the macroscopic dipole evolves and are compared with the experimental ones that have been reported recently, which shows good agreements.

2 Semiclassical theory of superradiance

After photonization and state couplings by 800-nm laser pulses, the molecular nitrogen ions are populated in B and X states that constitute a two-level system in the plasma. Since the intensity of the 391-nm seed pulse is weak, it can hardly induce ionization or non-resonant excitation. So only the interaction between the seed pulse and the two electronic states is significant. The electron-ion inelastic collisions and the electron-ion recombination can cause the population change (decay or increase) of the two-level system. Both of the them occur on nanosecond timescale [42–49], which is a much slower process compared to the 391-nm lasing emission that takes place in picosecond range. Hence, the population of the two-level system is considered to be conserved during the ultra-fast radiation.

Under the above conditions, the evolution of the two-level system of B ($\nu' = 0$) and X ($\nu = 0$) in the radiation field can be described by the optical Bloch equations by using the slowly varying amplitude approximation and the rotating-wave approximation [50–52]:

$$\frac{\partial u}{\partial t} = -\Delta\nu - \Gamma_2 u, \quad (1)$$

$$\frac{\partial v}{\partial t} = \Delta u + \Omega w - \Gamma_2 v, \quad (2)$$

$$\frac{\partial w}{\partial t} = -\Omega v - \Gamma_1 (1 + w), \quad (3)$$

where u and v denote dispersion and absorption terms, respectively, and w is the relative population difference between the upper and lower states. $\Omega(t)$ is the Rabi frequency which takes the form

$$\Omega = \begin{cases} \frac{\mu \bar{E}_p}{\hbar}, & 0 < t < \tau_r, \\ \frac{\mu \bar{E}_s}{\hbar}, & t \geq \tau_r, \end{cases} \quad (4)$$

with μ , \hbar , \bar{E}_p , \bar{E}_s and τ_r being the electronic transition moment between the two states, the reduced Planck's constant, the envelope amplitude of the seed laser pulse, the radiation field generated by the two-level system and the interaction time between the seed pulse and the two-level system, respectively. The resonant condition is considered here, i.e., $\Delta = 0$. It is noticed that the optical Bloch Eq. 1 are written in the coordinate system moving with the seed laser field. The transformation between the moving coordinate system (t, z) and the ground coordinate system (t', z') is

$$t = t' - \frac{z'}{c}, z = z', \quad (5)$$

where z and z' represent the propagation direction of the seed laser pulse, and c is the speed of light. z and z' range from 0 to L which is the filament plasma length. A pencil-shaped geometry for the active gain volume is used, i.e.,

$$r \ll L, \quad (6)$$

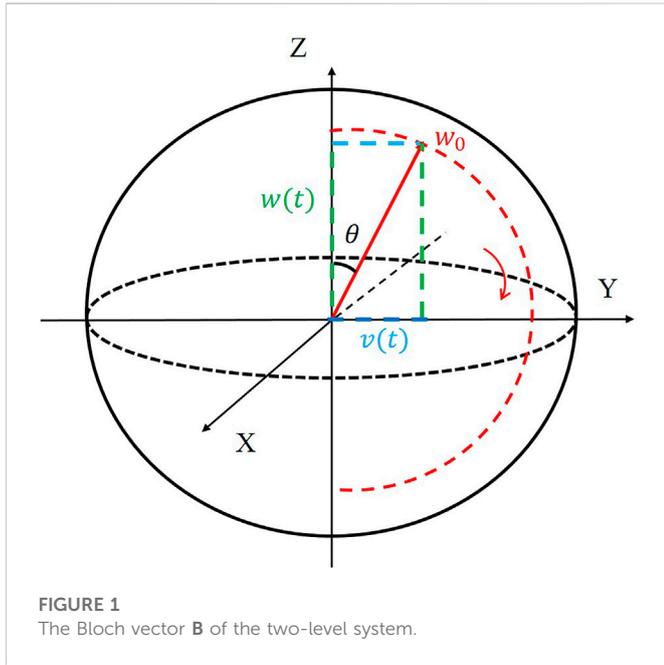


FIGURE 1 The Bloch vector **B** of the two-level system.

with r being the filament radius. It is reasonable because the plasma radius is usually around $50 \mu\text{m}$, and the filament plasma length is typically in centimeter range by using a lens of short focal length [53].

In Eqs 1–3, $\Gamma_1 = 11.76 \times 10^6 \text{ s}^{-1}$ is the transition rate [44], and the corresponding lifetime $1/\Gamma_1 = 85 \text{ ns}$ is about 4 orders of magnitude larger than the time delay and width ($\sim\text{ps}$) of the 391-nm retarded emission. Γ_2 is the dipole-dephasing rate, which can inhibit the formation of the macroscopic dipole moment. The radiation manifests characteristics of superfluorescence, damped superfluorescence and amplified spontaneous emission (ASE) if $1/\Gamma_2 \gg \tau_D$, $\sqrt{\tau_W \tau_D} \ll 1/\Gamma_2 < \tau_D$ and $\tau_W \ll 1/\Gamma_2 \ll \sqrt{\tau_W \tau_D}$, respectively [54–57]. The dephasing process is mainly caused by electron-ion elastic scattering in the nitrogen plasma generated by femtosecond laser pulses. Because of the lackness of elastic electron scattering cross-section for molecular nitrogen ions, we take the dephasing rate Γ_2 as a varying parameter and assume that $1/\Gamma_2$ is much greater than the time delay and width of the 391-nm retarded emission. As a reference, the ponderomotive potential of electron in the linearly polarized laser field with the laser intensity of 10^{14} W/cm^2 [13, 53]. If the elastic scattering cross-section for molecular nitrogen ions is similar to that for nitrogen molecules, the mean time between collisions is .94 ns for the density of the molecular nitrogen ions around 10^{16} cm^{-3} in the filament plasma [8, 29]. It still exceeds the time delay and width of the 391-nm retarded emission by 2 orders of magnitude. Therefore, within the time scale of 391 nm lasing pulse we considered, the spontaneous decay of upper state and the influence of collisional dephasing can be neglected, and we set both Γ_1 and Γ_2 as zero in the following derivation.

To solve Eqs 1–3, a Bloch angle is defined as

$$\theta(t) = \int_0^t \Omega(\tau) d\tau. \tag{7}$$

Then, simple solutions of the equations can be obtained as.

$$u(t) = 0, \tag{8}$$

$$v(t) = w_0 \sin \theta(t), \tag{9}$$

$$w(t) = w_0 \cos \theta(t), \tag{10}$$

with the initial conditions of $u(0) = 0, v(0) = 0$ and $w(0) = w_0$. By setting the eigen-energy of B and X to be $\hbar\omega/2$ and $-\hbar\omega/2$, the energy density of the two-level system can be expressed as

$$E_N(t) = \frac{1}{2} \hbar\omega N w_0 \cos \theta(t), \tag{11}$$

where N is sum of the population densities of the two states.

We define a Bloch vector **B** as

$$\mathbf{B} = (u, v, w) = u\mathbf{e}_X + v\mathbf{e}_Y + w\mathbf{e}_Z, \tag{12}$$

with $\mathbf{e}_X, \mathbf{e}_Y$ and \mathbf{e}_Z denoting the unit vectors of the X-, Y- and Z-axis, respectively. It is easy to obtain

$$|\mathbf{B}| = \sqrt{u^2 + v^2 + w^2} = w_0. \tag{13}$$

As $u = 0$, the Bloch vector **B** with a fixed length w_0 rotates around X-axis in Y-Z plane as shown in Figure 1. The rotation determined by the Bloch angle θ describes the evolution of the physical quantities including the energy density of the two-level system.

When $t < \tau_r$, the Rabi frequency Ω_p induced by the seed laser pulse is known, and the Bloch angle θ can be calculated by $\theta(t) = \int_0^t \Omega_p(\tau) d\tau$. Next, we investigate the development of θ when $t \geq \tau_r$. The relationship between the radiation field $E_s(t', z')$ and polarization $P(t', z')$ generated by the two-level system is governed by the Maxwell's equations

$$\frac{\partial^2 E_s}{\partial z'^2} - \mu_0 \epsilon_0 \frac{\partial^2 E_s}{\partial t'^2} = \mu_0 \frac{\partial^2 P}{\partial t'^2}, \tag{14}$$

where μ_0 and ϵ_0 denote vacuum permeability and vacuum permittivity, respectively. The radiation field $E_s(t', z')$ and polarization $P(t', z')$ can be written in the form

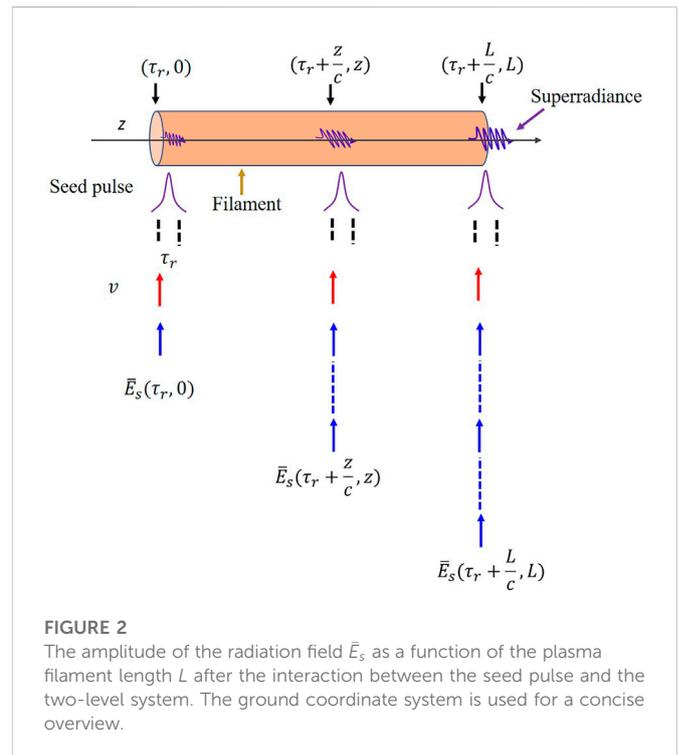


FIGURE 2 The amplitude of the radiation field \bar{E}_s as a function of the plasma filament length L after the interaction between the seed pulse and the two-level system. The ground coordinate system is used for a concise overview.

$$E_s(t', z') = \frac{1}{2} [\bar{E}_s(t', z') e^{-i\omega t' + ikz'} + c.c.], \quad (15)$$

$$P(t', z') = \frac{1}{2} [-iN\mu v(t') e^{-i\omega t' + ikz'} + c.c.], \quad (16)$$

where $\bar{E}_s(t', z')$ is the envelope amplitude of $E_s(t', z')$. By applying the rotating-wave approximation and slow amplitude approximation [59], Eq. 14 can be simplified to

$$\frac{\partial \bar{E}_s}{\partial z'} + \frac{1}{c} \frac{\partial \bar{E}_s}{\partial t'} = \frac{\mu_0 \omega c \mu N}{2} v(t'). \quad (17)$$

Substituting (t', z') with (t, z) into Eq. 17, we get

$$\frac{\partial \bar{E}_s(t, z)}{\partial z} = \frac{\mu_0 \omega c \mu N}{2} v(t). \quad (18)$$

With the initial condition $\bar{E}_s(\tau_r, 0) = 0$, one further obtains

$$\bar{E}_s(\tau_r, L) = \frac{\mu_0 \omega c \mu N L}{2} v(\tau_r), \quad (19)$$

This derived expression suggests the amplitude of the radiation field emitted at the end of the plasma at $t = \tau_r$.

To get a clear physical picture of Eq. 19, Figure 2 illustrates the growth of \bar{E}_s with the increase of the plasma length L . The discussion below is performed in the ground coordinate system for a straightforward understanding. Considering the plasma as a series of consecutive micro regions $0 \sim \delta L, \dots, z \sim z + \delta L, \dots, L - \delta L \sim L$, the seed pulse interacts with the two-level system at each region, which is described by the optical Bloch Eqs 1–3. In the region of $0 \sim \delta L$, after the interaction there is a $v(\tau_r, 0)$, which generates a radiation field with the amplitude of $\bar{E}_s(\tau_r, 0)$. In the region $z \sim z + \delta L$ which corresponds to a propagation time of z/c , the seed pulse induce the same polarization and radiation field as that in the region of $0 \sim \delta L$. At the same time, all the radiation field generated before the infinitesimal region $z \sim z + \delta L$ have arrived. As a result, the amplitude $\bar{E}_s(\tau_r + z/c, z)$ is a superposition of the amplitudes generated by the infinitesimals $0 \sim \delta L, \dots, z \sim z + \delta L$. Therefore, the final amplitude of the radiation field can be expressed as Eq. 19.

At $t = \tau_r$, the energy flux density S of the radiation field from the cross section of the plasma is

$$S = \frac{1}{2} \sqrt{\frac{\epsilon_0}{\mu_0}} \bar{E}_s^2(\tau_r, L) = \frac{\mu_0 c \omega^2 \mu^2 N^2 L^2}{8} w_0^2 \sin^2 \theta(\tau_r). \quad (20)$$

The decrease of energy per unit time of the two-level system at $t = \tau_r$ according to Eq. 11 is

$$-\frac{dE_N}{dt} = \frac{1}{2} \hbar \omega N w_0 \sin \theta \left. \frac{d\theta}{dt} \right|_{t=\tau_r}. \quad (21)$$

From the law of conservation of energy, the decrease of energy per unit time from the active volume equals the energy flux from the cross section, which reads

$$-\frac{dE_N}{dt} \pi r^2 L = S \pi r^2. \quad (22)$$

By substituting both sides respectively and assuming no external factors affecting the system, one gets the derivative of the Bloch angle with respect to time

$$\frac{d\theta}{dt} = \frac{\mu_0 c \omega \mu^2 N L w_0}{4 \hbar} \sin \theta(\tau_r) \quad (23)$$

with the initial condition $(\tau_r, \theta(\tau_r))$. The solution of Eq. 23 is

$$\theta(t) = 2 \arctan \left(e^{\frac{t - \tau_D}{\tau_W}} \right) \quad (24)$$

with

$$\tau_W = \frac{4 \hbar}{\mu_0 c \omega \mu^2 w_0 N L}, \quad (25)$$

$$\tau_D = \tau_r - \tau_W \ln \left[\tan \frac{\theta(\tau_r)}{2} \right]. \quad (26)$$

The energy density $E_N(t)$ of this two-level system according to Eq. 9 and the radiation intensity I_s (the same as the energy flux density S) at the end of the plasma can be respectively expressed as

$$E_N(t) = -\frac{1}{2} \hbar \omega w_0 N \tanh \left(\frac{t - \tau_D}{\tau_W} \right), \quad (27)$$

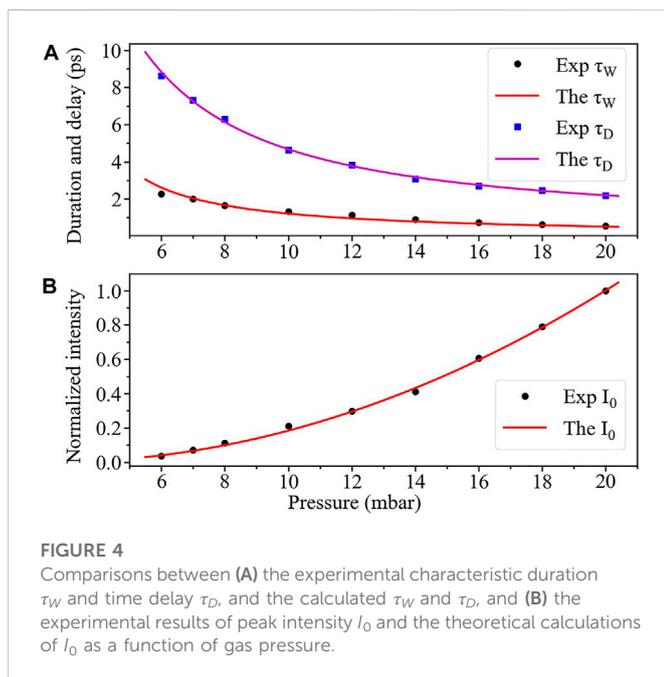
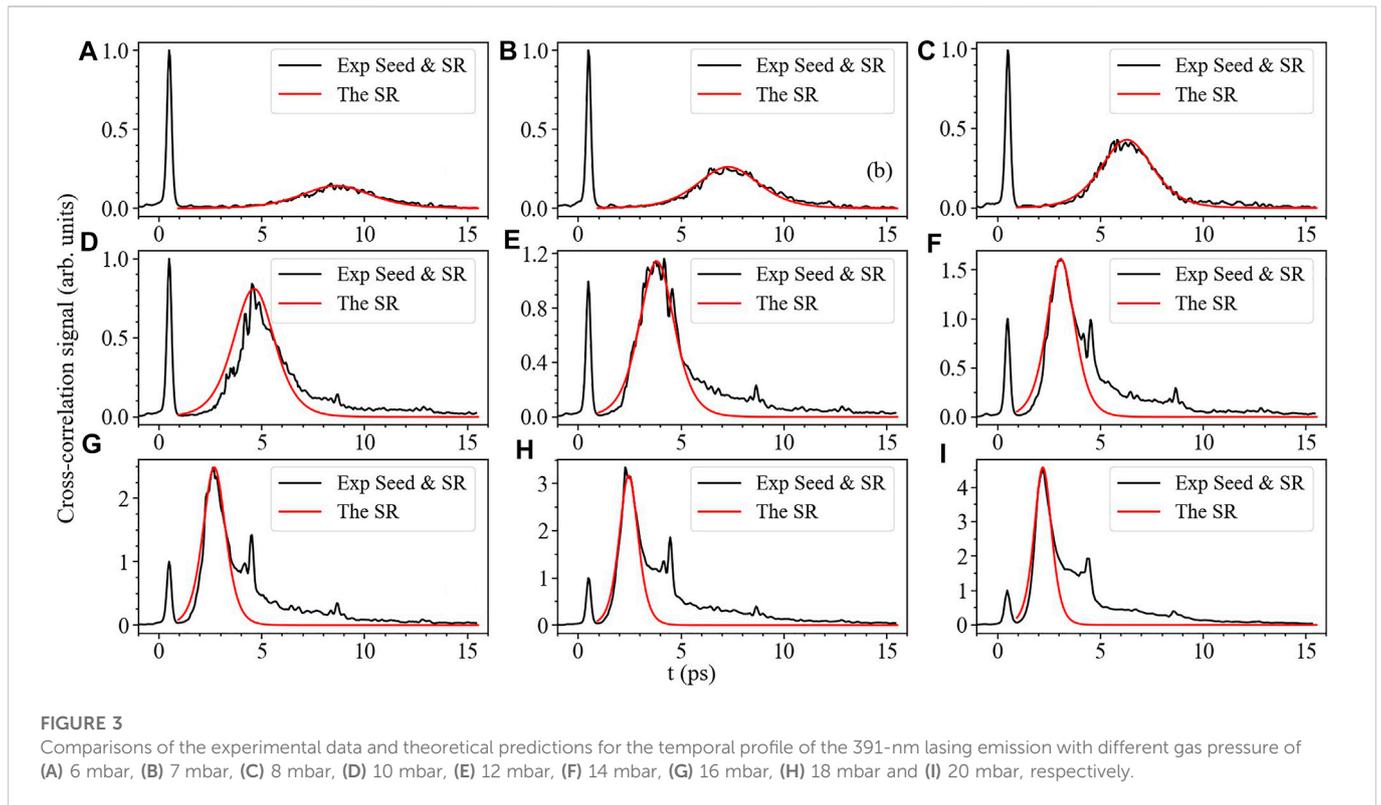
$$I_s = \frac{\mu_0 c \omega^2 \mu^2 w_0^2 N^2 L^2}{8} \operatorname{sech}^2 \left(\frac{t - \tau_D}{\tau_W} \right). \quad (28)$$

From above analytical expressions, we can see that the radiation from the two-level system possesses the nature of superradiance [54–56, 60–64] featured with the characteristic duration of τ_W and delay of τ_D with respect to the seed pulse. τ_W is inversely proportional to the population density N and the radiation intensity is proportional to the square of N^2 , typical signatures of superradiance.

As described by Eq. 27, there is still energy stored in the system after interaction with the seed pulse. The release of remaining energy results in the retarded emission. The initial relative population difference w_0 determined by the pump pulse and the Bloch angle $\theta(\tau_r)$ caused by the seed pulse govern the temporal evolution of the retarded emission.

3 Comparison between the experimental results and theoretical predictions

In this section, we perform comparisons between the experimental results and theoretical predictions of the temporal profile of the 391-nm lasing emission based on analytical expressions derived. The experimental results are directly taken from previous work [65], where detailed descriptions of the experiments can be found. Figure 3 shows the temporal profiles of 391-nm lasing pulse measured for different nitrogen pressures from 6 to 20 mbar. The interaction time τ_r for the calculation is set to be $3.6\tau_p$, with $\tau_p = 0.26$ ps being the duration of the seed pulse. The characteristic duration is $\tau_W = \tau_{FW}/1.763$ for the hyperbolic secant pulse, where τ_{FW} is the experimental FWHM of the 391-nm lasing pulse. By substituting τ_W and τ_D at different pressures into Eq. 28, we obtain the corresponding temporal profile as shown by the red line in Figure 3. When the gas pressure exceeds 10 mbar, the theoretical predictions are in agreement with the experimental signals for the main part of superradiance as illustrated in Figures 3D–I. Following the strongest radiation, there are the other two gains at ~ 4.4 and 8.6 ps that correspond to the revival of the optical gain due to the molecular alignment effect [21, 66], which was not considered in the theoretical derivation. Besides, there exist a gradual decay in the measured temporal profiles at high pressures. By increasing the dephasing rate Γ_2 of the transition dipole in KCl: O_2^- , Malcuit et al. found that the temporal profiles of the 629.4-nm emission from KCl: O_2^- changed from a standard superfluorescence



to having an obviously long decay [54]. In our case, with the nitrogen gas pressure increasing, the dephasing rate Γ_2 increases. Even though the dephasing rate is not larger enough to inhibit the formation of the macroscopic dipole moment (superradiance), it indeed leads to the gradual decay in the measured temporal profiles at higher pressures, which was also found in Xie et al.'s experiment [41]. In general, the theoretical results agree well with the experimental data, which is a

strong evidence that the 391-nm emission inside ionized nitrogen molecules is superradiance.

The time-dependent quantum-wave-packet calculations show that the initial relative population difference w_0 varies from 0 to 0.55 with the 800-nm laser intensity ranging from 2.2 to 4×10^{14} W/cm² [23]. The 800-nm pump laser intensity in our experiment is estimated to be 2.6×10^{14} W/cm² with the energy of 1.8 mJ, duration of 45 fs and filament diameter of ~ 70 μ m. Thus, it is reasonable to set w_0 as .1. Since the pump laser pulse energy is unchanged for different gas pressure ranging from 6 to 20 mbar [29, 53], the two-level-system population density is proportional to gas pressure. In actual case, the superradiance will vanish when the pressure is lower than a certain value p_0 . The minimal gas pressure that causes the superradiance is $p_0 = 2.5$ mbar in our case. The relationship between the population density and pressure can be expressed $N = k(p - p_0)$, where k is a scaling factor. Using the experimental point ($p = 8$ mbar, $\tau_W = 1.666$ ps), we obtain $N = 0.228 \times (p - 2.5)$ (mbar) $\times 10^{16}$ cm⁻³ with $L = 10$ mm and $\mu = 1.7 D$ [67]. Hence, the characteristic duration τ_W dependent on the gas pressure can be calculated. The calculated τ_W (red line) as a function of the nitrogen pressure show good agreement with the experimental data (black circle), as shown in Figure 4A.

The seed pulse in Figure 3 has a peak intensity of 10^7 W/cm², and can be well fitted by a Gaussian shape with the pulse duration $\tau_p = 0.26$ ps. The initial Bloch angle is accurately calculated

$$\theta(\tau_r) = 2 \int_0^{\tau_r} \frac{\mu}{\hbar} \bar{E}_p(t) dt = 0.057\pi, \quad (29)$$

which is far smaller than $\pi/2$. The two-level system does not experience several cycles of Rabi oscillation under the presence of the seed pulse. Only little energy emits with the seed pulse

simultaneously, which explains the almost unchanged intensity of the seed pulse after passing through the filament plasma. It can also be seen that the theoretical calculations for τ_D are in good agreement with the experimental results (blue square) in Figure 4A.

The peak intensities I_0 of the superradiance are investigated as a function of nitrogen pressure. The values at $t = \tau_D$ of the experimental 391-nm emission in Figure 3 are taken as the peak intensities. The theoretical peak intensity is expressed as $I_0 = (1/8)\mu_0 c \omega^2 \mu^2 w_0^2 N^2 L^2$, which is proportional to N^2 . Using the linear relationship between N and p , the normalized I_0 (read line) of the theory is calculated, as shown in Figure 4B, also showing good agreement with the experimental data. Additionally, it is noticed that the peak intensity of superradiance scales as N^2 but the total emitted energy is proportional to N according to Eq. 23. This fact inevitably leads to the characteristic duration τ_W to be proportional to N^{-1} . We emphasize that N is the sum of population density of B and X . The intensity is influenced by not only the population in the excited state but also that in the ground state.

From the comparisons in terms of the temporal shape, the characteristic duration, the time delay and the intensity between the theoretical and experimental results above, it can be seen that the assumption $1/\Gamma_2 \gg \tau_W$ and $1/\Gamma_2 \gg \tau_D$ is reasonable. If the dephasing rate Γ_2 is sufficiently large, no macroscopic dipole moment ever develops and each electric dipole simply responds to the instantaneous value of the laser field. In this case a very noise output pulse shows up, of which the pulse duration and the peak intensity no longer scale as $1/N$ and N^2 [54].

4 Conclusion

In conclusion, we theoretically investigate the evolution of energy in the population-inverted molecular nitrogen ions induced by the coherent seed pulse. By solving the Bloch angle of the two-level system of $N_2^+(B^2\Sigma_u^+, \nu' = 0)$ and $N_2^+(X^2\Sigma_g^+, \nu = 0)$, the semiclassical superradiance theory is proposed to describe the characteristic duration, time delay, intensity and total emitted energy. We explain the time-delayed optical amplification in molecular nitrogen ions irradiated with intense femtosecond laser pulses, and further confirm the superradiance nature of the 391-nm forward emission by the comparisons between the theoretical and experimental results.

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Data availability statement

Publicly available datasets were analyzed in this study. This data can be found here: https://www.researchgate.net/publication/316084384_Lasing_effect_in_femtosecond_filaments_in_air.

Author contributions

QW conceived the concept. PD provide the experimental data. QW, SW, and MA-K performed the theoretical derivation. QW, PD, YZ, ZL, and BH analyzed the data. The manuscript was prepared by QW and PD, and was discussed among all authors.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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