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Lasing dynamics of neutral nitrogen molecules in femtosecond filaments

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We investigate the dynamics of cavity-free lasing from neutral nitrogen molecules in femtosecond laser filaments. An important difference of intensity and temporal duration is observed between backward and forward lasing, in both amplified spontaneous emission and seed-amplification regimes. Numerical simulations based on a nonadiabatic Maxwell-Bloch model reproduce well the observations, and attribute these differences to the finite gain lifetime and the traveling excitation nature of this gas laser.

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I. INTRODUCTION

Coherent emission from the atmosphere is attracting intense attention because of its potential in remote sensing applications [1–8]. Traditional optical remote sensing systems for probing the atmosphere rely on collecting backward-scattered incoherent light from targets in the atmosphere to detectors on the ground, a technique hindered by low efficiency and poor directionality. Availability of a light source which emits coherent light in the backward direction from a certain point in the sky back towards a ground-based detector could have a great impact on the efficiency and precision of optical remote sensing techniques [9]. Several possible approaches have been proposed for implementing a free-space mirrorless laser source emitting in the backward direction [1–3,5–14]. One of the most promising approaches so far is through two-photon dissociation and subsequent resonant two-photon excitation of neutral oxygen or nitrogen atoms by deep-UV laser pulses. However, the strong absorption of deep-UV pump laser pulse in air ultimately limits the distance at which this type of laser can be pumped [1,3,6]. This restriction disappears with a pump laser in the visible or IR region.

Recently, Mityukovskiy and co-workers demonstrated that the filament plasma column created by intense femtosecond laser pulse at 800 nm in air can act as an optical amplifier and give rise to lasing at 337.1-nm wavelength in backward directions [11]. Stimulated emission at this wavelength corresponds to a transition between levels $C^3\Pi_u^+ \rightarrow B^3\Pi_g^+$ in the triplet manifold of the neutral nitrogen molecule. Amplification takes place only if the infrared (IR) laser pulse is circularly polarized [11]. In the presence of external seeding around 337.1 nm in both backward and forward directions, the emission was enhanced by orders of magnitude [12,13]. Based on a study on the sideways fluorescence, inversion of population between states $C^3\Pi_u^+$ and $B^3\Pi_g^+$ responsible for optical gain has been attributed to the following scenario [see Fig. 1(a)] [14]. The intense filamentary pulse of peak intensity, $I = 1.5 \times 10^{14}$ W/cm², liberates free electrons from nitrogen and oxygen molecules through a high-field tunneling process [11,15]. The energy of the free electrons left after

the passage of the pulse depends sensitively on the laser polarization. For a linearly polarized pump pulse at 800 nm, most free electrons are left with low kinetic energy. By contrast, with circularly polarized light, the electrons have a peak in their energy distribution at $E = 2U_p = 14.5$ eV where $U_p = e^2/c\epsilon_0 m_e (I/2\omega_0^2)$ [Fig. 1(b)]. Fortuitously, this energy is optimal to promote a nitrogen molecule from its ground state to state $C^3\Pi_u^+$ via impact collision and to achieve thereby inversion of population with respect to $B^3\Pi_g^+$. We have previously reported seed-amplified lasing in pure nitrogen in the forward as well as the backward direction, wherein the backward geometry pump and seed pulses are counterpropagating [12]. However, the presence of oxygen with a concentration in excess of 10% led to a quenching of lasing [11,13].

We now routinely observe seed-amplified emission in air in the forward direction using a pump laser pulse of a few millijoules of energy and 50 fs duration (see Fig. 2); however, this is not yet the case in the backward direction. The main reason is the pronounced asymmetry of the lasing intensity between backward and forward geometries in favor of the latter. This is regrettable since backward lasing is particularly useful for long distance atmospheric diagnostics. It is therefore important to understand in detail the origin of this asymmetry in order to envision strategies allowing backward lasing in air.

To this purpose, we have measured the temporal dynamics of the amplified spontaneous emission (ASE) and of externally seeded lasing at 337.1-nm wavelength in a filament in pure nitrogen, both in the forward and backward directions. These emissions were examined in the time domain with time-resolved techniques [11–13]. Significant differences in pulse duration and intensity are observed for ASE versus seeded lasing and for copropagating versus counterpropagating pump-seed geometries, features which are unexpected. We apply to our present case a theoretical model that successfully explained the collisional-induced amplification of XUV high-order harmonics injected into a plasma column. This model, which takes into account the pump-seed geometry and the initial free-electron energy distribution, reproduces the observed dynamics. It explains the strong lasing signal asymmetry in the forward versus backward gain, which originates from the short lifetime of optical gain and the traveling excitation nature of the filament amplifier. It also shows that an analysis relying solely

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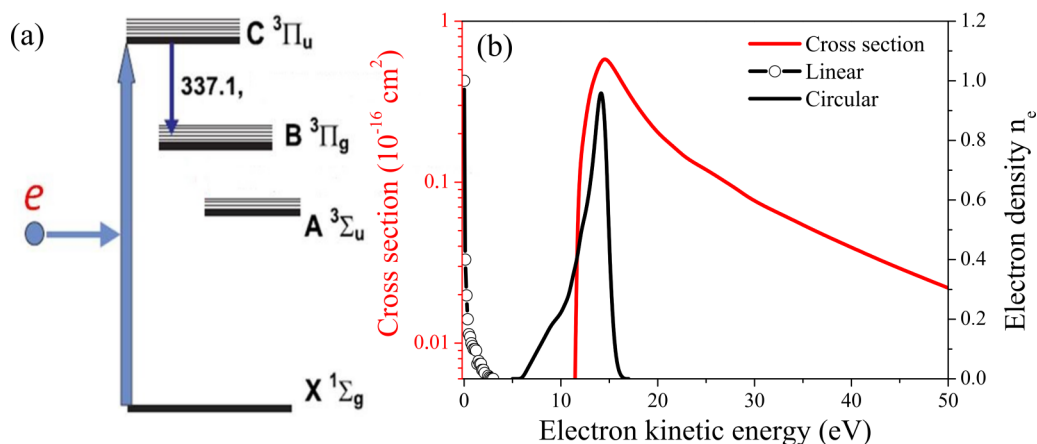


FIG. 1. (a) Schematic energy levels of the neutral nitrogen molecules. Collisional excitation of the ground-state ($X^1\Sigma_g$) N_2 molecules results in population inversion between the third excited state ($C^3\Pi_u^+$) and the second excited state ($B^3\Pi_g^+$). (b) Cross section of the collisional excitation to the $C^3\Pi_u^+$ state (red) and the electron energy distribution in the filaments pumped by circularly polarized femtosecond pulse (black line) and the linearly polarized one (black line-dot).

97 on rate equations would be insufficient to treat the results.
 98 Instead, full consideration of the medium polarization and of
 99 the nonadiabatic excitation process is necessary.

100 **II. EXPERIMENTAL RESULTS**

101 In the experiments, femtosecond laser pulses (wavelength
 102 centered at 800 nm, pulse duration of 45 fs, repetition rate
 103 of 100 Hz, and maximum pulse energy of 16 mJ) delivered
 104 by a commercial laser system (Thales, Alpha 100), were split
 105 into three replicas, *a*, *b*, and *c*. The time delays between these
 106 pulses were controlled by mechanical optical delay lines. For
 107 the creation of the nitrogen filament amplifier, a circularly
 108 polarized femtosecond pulse of ~ 9 mJ (replica *a*) was focused
 109 by a convex focal lens with $f = 1$ m into a nitrogen-gas-filled
 110 chamber. Both backward and forward ASEs at 337.1 nm were
 111 from the filamentary plasma with a length $l \sim 30$ mm were

112 recorded. To obtain the seeding pulse at 337.1 nm, the second
 113 harmonic of pulse *b* was generated in a 1-mm-thick BBO
 114 crystal and then focused into a 20-mm-thick fused silica
 115 sample. A narrow spectral bandwidth (~ 10 nm bandwidth)
 116 of the emerging broadband continuum was selected with an
 117 interference filter around 337 nm. More technical details can
 118 be found in [12]. The seed pulse could be injected into the
 119 plasma in both directions. To characterize the pulse duration,
 120 the forward 337.1 nm ASE or amplified lasing radiation was
 121 focused together with the third 800-nm pulse replica *c* on
 122 a 2-mm-thick type-I BBO crystal cut specially at 50.7° , to
 123 efficiently produce a sum frequency generation (SFG) signal
 124 at 238 nm wavelength. By recording the sum frequency signal
 125 at 238 nm as a function of the time delay between the 337.1-nm
 126 lasing signal and the weak 800-nm probe pulse *c*, the temporal
 127 profile of the 337.1-nm lasing pulse is obtained.

128 We first present the optical gain dynamics of the 337.1-nm
 129 lasing emission measured in the pump-seed copropagation
 130 configuration. By measuring the seed amplification as a
 131 function of relative delay between pump and weak seed pulse,
 132 the temporal profile of gain can be probed. The result shown
 133 in Fig. 3 reveals a rapid gain buildup time of ~ 5 ps, and a
 134 slow decay time of ~ 35 ps. The duration of optical gain [full
 135 width at half maximum (FWHM)] is about $\tau_g = 13$ ps. Now
 136 we turn to the temporal profile of the stimulated radiation in
 137 the forward direction. The measured temporal structure of the
 138 forward ASE and seeded amplified emission are presented in
 139 Fig. 4(a). The 337.1-nm ASE pulse shows a buildup time of
 140 17 ps and a slow decay up to 53 ps, with a full width at half
 141 maximum duration of ~ 20 ps. In contrast, the seeded emission
 142 presents a much shorter rise time of ~ 3 ps and a duration of
 143 4 ps. The seeded lasing pulse precedes the ASE by about
 144 20 ps. The peak intensity is 50 times higher than for ASE, in
 145 good agreement with previous reports [12,13]. Note the slight
 146 temporal modulation structures with a period of about 11 ps
 147 apparent for both ASE and seeded emission, a feature related
 148 to the quantum beating effect that will be discussed later.

149 The temporal profile of the backward externally seeded
 150 337.1-nm lasing pulse measured with the cross-correlation
 151 method is shown in Fig. 4(b). It was normalized to its

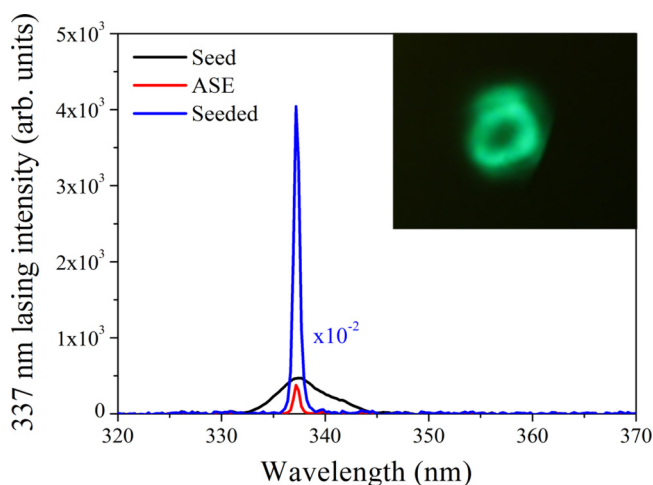


FIG. 2. Amplified spontaneous emission (red) and seeded emission (blue) obtained in ambient air in the forward direction. The ~ 9 -mJ circularly polarized femtosecond laser pulse was focused by an $f = 1$ m lens in atmospheric air. The inset presents the profile of the forward ASE obtained in pure nitrogen gas.

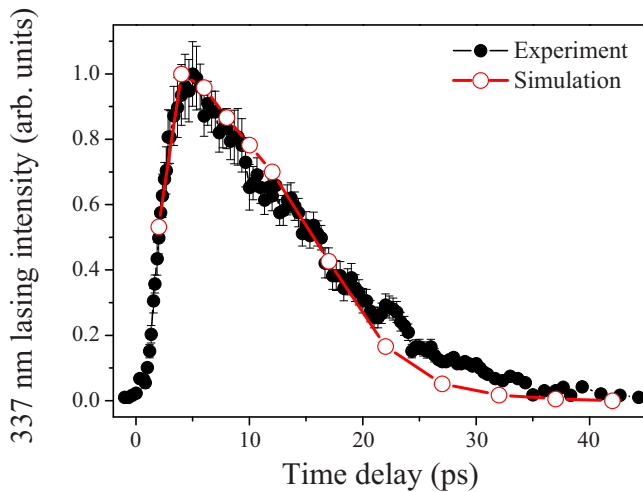


FIG. 3. Experimental (solid black dots) and simulated (red circles) results of the gain dynamics of 337.1-nm lasing emission with 7.5-mJ pump pulse energy under 1 bar of nitrogen gas.

counterpart in the forward direction. The radiation peak is delayed by 14 ps with respect to the seed pulse and a significant intensity modulation of period 11 ps is now obvious. The envelope of the backward seeded 337.1-nm lasing pulse gives a FWHM duration of ~ 15 ps. If we just concentrate on the strongest pulse following the seed pulse, it has a width (FWHM) of 4 ps. We have also found, by measuring the lasing pulse energy with a photodiode, that the backward seeded emission was about 150 times weaker than the forward one under the same excitation conditions. The backward 337.1-nm ASE lasing signal was too weak to be measured by the

cross-correlation method. We therefore turned to a Michelson interferometer to estimate its pulse duration by measuring the interference fringe visibility between two 337.1-nm ASE pulse replicas as a function of their relative delay. The interference pattern was captured by an intensified charge-coupled device (iCCD) (Princeton Instruments, PI-MAX). A periodic modulation of the fringe contrast with a period of ~ 11 ps is well apparent [see Fig. 4(c)], indicating that the ASE is also composed of multiple peaks in the temporal domain, similar to the seeded emission presented in Fig. 4(b). It yields a coherence time of ~ 120 ps, corresponding to a duration of 85 ps, as assuming a Fourier transform-limited ASE pulse.

III. THEORETICAL MODELING

In order to understand the different temporal dynamics of the forward and backward 337.1-nm emission observed in experiments, we adapted the one-dimensional time-dependent Maxwell-Bloch code DEEP-ONE [16,17] to our current case, following the lines of [18,19]. This code, originally developed to study the amplification of soft x rays ($\lambda = 10\text{--}40$ nm) in hot, dense plasma ($n_e > 10^{18}$ cm $^{-3}$, $T_e > 10\text{--}100$ eV), solves the paraxial wave equation for the electric field in the slowly varying envelope approximation,

$$\frac{\partial E_{\pm}}{\partial t} \pm c \frac{\partial E_{\pm}}{\partial z} = \frac{i\omega_0}{2} \left[\mu_0 c^2 P_{\pm} - \left(\frac{\omega_p}{\omega_0} \right)^2 E_{\pm} \right], \quad (1)$$

where E_+ , E_- are the electric fields propagating in the forward and backward directions, respectively; P_+ , P_- are the corresponding polarization densities of the medium; c is the light velocity in vacuum; ω_0 is the frequency of the electric field; ω_p is the free-electron plasma frequency; and μ_0

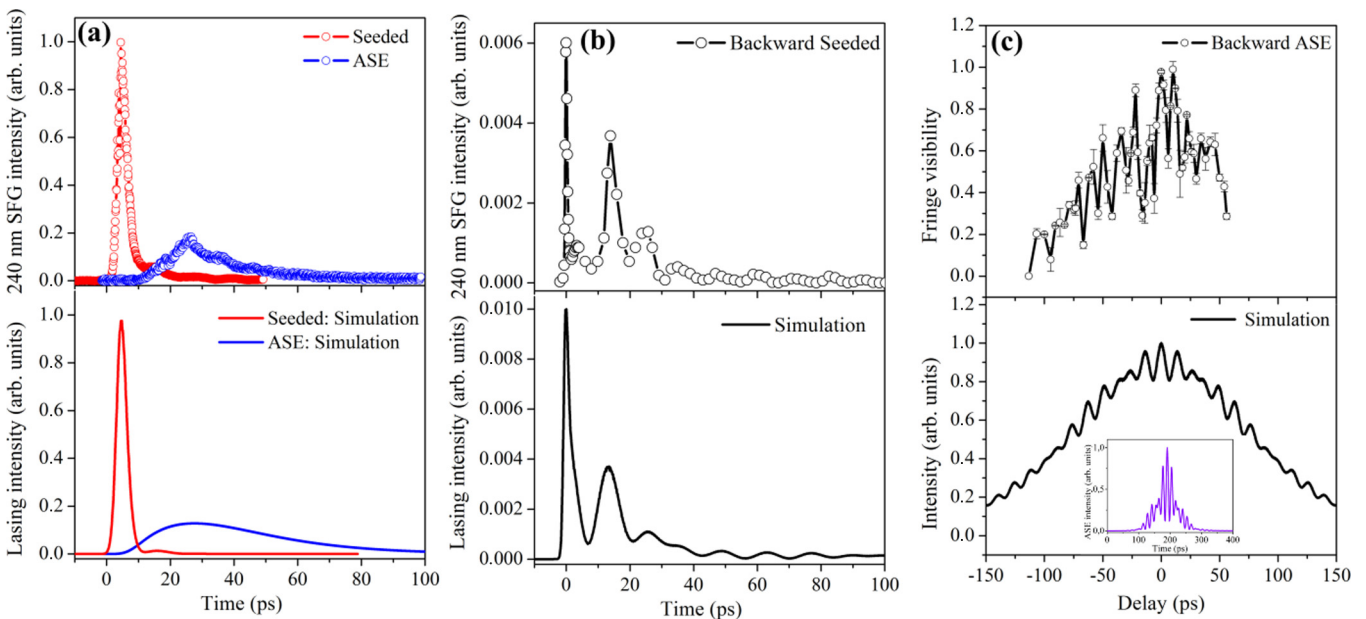


FIG. 4. (a) Experiment (upper panel) and simulation (lower panel) results of temporal profiles of forward 337.1-nm ASE lasing pulse (blue line) and externally seeded 337.1-nm lasing pulse (red line). The signal for the 337-nm ASE lasing pulse is enlarged by ten times for better comparison. (b) Experiment and simulation results of temporal profile of backward externally seeded 337.1-nm lasing pulse. (c) Experiment and simulation results of the autocorrelation trace of backward 337.1-nm ASE. The inset in the lower panel shows the simulated temporal profile of the backward 337.1-nm ASE.

190 is the vacuum permeability. Equation (1) is enhanced with a
 191 constitutive relation derived from Bloch equations,

$$\frac{\partial P_{\pm}}{\partial t} = \Gamma - \gamma P_{\pm} - \frac{iz_{ul}^2}{\hbar} E_{\pm}(N_u - N_l), \quad (2)$$

192 where Γ is a stochastic source term with vanishing correlation
 193 time modeling the spontaneous emission [20,21]; γ is the
 194 depolarization rate due to collisions; z_{ul} is the corresponding
 195 dipole matrix element (that can be deduced from Einstein's A
 196 coefficient); and N_u, N_l are, respectively, the population of the
 197 upper and lower levels of the lasing transition. Since the seed
 198 duration (~ 100 fs) is shorter than the typical collision time of
 199 the plasma (\sim ps), the adiabatic approximation is no more valid
 200 [22] and the full differential equation [Eq. (2)] must be used to
 201 describe the evolution of the polarization. These populations
 202 are computed by using rate equations,

$$\frac{\partial N_i}{\partial t} = \sum_k C_{ki} N_k \pm \mathfrak{S}(E^* P) \frac{1}{2\hbar}, \quad (3)$$

203 where $i = u, l$ and C_{ki} are the collisional (de)excitation and
 204 radiative deexcitation rates. These rates are computed from
 205 the cross sections reported in [23].

206 The evolution of electron density and temperature are
 207 computed using the model presented in [18,19], with the initial
 208 electron temperature for circular laser polarization obtained
 209 in [14,24]. The electron temperature T_e and the vibrational
 210 temperature T_v are given, respectively, by

$$\frac{3}{2} \frac{\partial N_e T_e}{\partial t} = -Q_c N_a N_e \left(1 - \frac{T_v}{T_e}\right), \quad (4)$$

$$\frac{3}{2} N_a \frac{\partial T_v}{\partial t} = Q_c N_a N_e \left(1 - \frac{T_v}{T_e}\right), \quad (5)$$

211 where N_e is the electron density; N_a the neutral density; and
 212 Q_c is the cooling rate, that for $T_e < 2$ eV is given analytically
 213 by the following formula:

$$Q_c \approx 3.5 \times 10^{-8} \exp\left(-\frac{5}{3T_e}\right) + 6.2 \times 10^{-11} \exp\left(-\frac{1}{3T_e}\right). \quad (6)$$

214 Since the initial electron temperature of 16 eV cools in
 215 less than 6 ps to a value lower than 2 eV, the value of Q_c is
 216 extrapolated for $T_e > 2$ eV.

217 The electron density N_e and positive N_p and negative N_n
 218 ion densities are given by

$$\frac{\partial N_e}{\partial t} = \nu_{\text{ion}} N_e - \beta N_p N_e - \eta N_e, \quad (7)$$

$$\frac{\partial N_n}{\partial t} = \eta N_e - \beta_{np} N_n N_p, \quad (8)$$

$$N_p = N_e + N_n, \quad (9)$$

219 where ν_{ion} is the collisional ionization rate, β is the electron-
 220 ion recombination rate, β_{np} is the ion recombination rate,
 221 and η the attachment coefficient. The following analytical
 222 approximations, obtained from [18], are used to calculate the

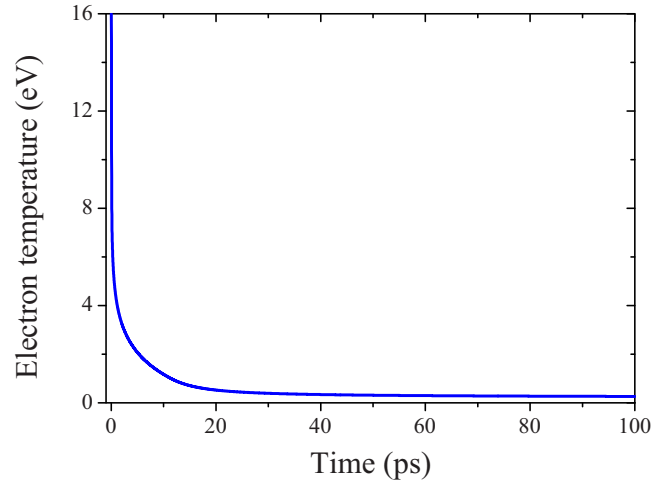


FIG. 5. Temporal evolution of the electron temperature along 100 ps.

aforementioned parameters; i.e.,

$$\nu_{\text{ion}} = \nu_{\text{ion}}(N_2) + \nu_{\text{ion}}(O_2); \quad (10)$$

$$\nu_{\text{ion}}(X) = \nu_X \left(\frac{T_e}{U_X}\right)^{3/2} \left(\frac{U_X}{T_e} + 2\right) \exp\left(-\frac{U_X}{T_e}\right),$$

$$X = N_2, O_2; \quad U_{N_2} = 15.6 \text{ eV}; \quad U_{O_2} = 12.1 \text{ eV};$$

$$\nu_{N_2} = 7.6 \times 10^{11} \text{ s}^{-1}; \quad \nu_{O_2} = 10^{11} \text{ s}^{-1}; \quad (11)$$

$$\beta \left(\frac{\text{cm}^3}{\text{s}}\right) \approx 1.5 \times 10^{-8} T_e^{-0.7}, \quad \text{for } T_e < 0.1 \text{ eV}; \quad (12)$$

$$\beta \left(\frac{\text{cm}^3}{\text{s}}\right) \approx 2.0 \times 10^{-8} T_e^{-0.56}, \quad \text{for } T_e > 0.1 \text{ eV}; \quad (13)$$

$$\eta[\text{s}^{-1}] = \alpha_2 N_a + \alpha_3 N_a^2; \quad (14)$$

$$\alpha_2 \left[\frac{\text{cm}^3}{\text{s}}\right] = 2.75 \times 10^{-10} T_e^{-0.5} \exp\left(-\frac{5}{T_e}\right); \quad (15)$$

$$\alpha_3 \left[\frac{\text{cm}^3}{\text{s}}\right] = 1.5 \times 10^{-32} T_e^{-1} \exp\left(-\frac{0.052}{T_e}\right). \quad (16)$$

IV. NUMERICAL SIMULATION RESULTS AND DISCUSSION

224 With this model, we performed numerical simulation on the
 225 AND DISCUSSION
 226
 227 With this model, we performed numerical simulation on the
 228 optical gain dynamics. As shown in Fig. 3, the results agree
 229 well with the experiments. To understand the ultrafast gain
 230 buildup (~ 5 ps), we plot the temporal evolution of electron
 231 temperature in Fig. 5. The filament has an initial electron
 232 density of $N_e = 2 \times 10^{-16} \text{ cm}^{-3}$ and electron temperature of
 233 $T_e = 16$ eV. As shown in the figure, the free electrons cool
 234 in the first picoseconds of evolution, due to collisions with
 235 neutrals. When the temperature is low enough, the collisional
 236 excitation pumping is no longer effective, which therefore
 237 leads to a rapid buildup of the gain.

238 Now we turn to the temporal dynamics of the forward
 239 emission. The forward ASE starts from spontaneous emission
 240 spanning all the gain duration due to the long lifetime of the
 241 upper level of the transition $\tau_u \approx 40$ ns. Amplification and
 242

241 saturation effects shorten its duration. After passing through
 242 3 cm of plasma, the ASE pulse has a duration (FWHM) of
 243 44 ps, which agrees with our observations [see Fig. 4(a)].
 244 In the external-seeding case, the evolution of the temporal
 245 structure of the seed pulse is mainly driven by its initial
 246 spectral profile, related to its duration and the linewidth of
 247 the transition, dominated by collisions (i.e., a Lorentzian line).
 248 The mismatch between the linewidth of the seed pulse and
 249 that of the transition triggers the creation, during the first
 250 centimeter, and subsequent amplification of a retarded wake
 251 pulse [16,22]. When the wake is intense enough, saturation
 252 effects reduce its duration until it attains the value of 4.5 ps
 253 (FWHM) after propagating through a 3-cm-long filament
 254 plasma, again in agreement with the experiment results [see
 255 Fig. 4(a)].

256 Figure 4(b) (lower panel) shows the simulated temporal
 257 evolution of the seeded backward emission. Note that the seed
 258 and the IR pump pulses enter the plasma from opposite sides,
 259 at $t = 0$ ps. Thus, the seed propagates without amplifying until
 260 it meets the IR pump in the middle of the filament ($t = 50$ ps).
 261 From this point on, the seed is amplified. At the exit of
 262 the filament amplifier, the simulation predicts a moderately
 263 amplified seed pulse, followed by a wake radiation extending
 264 over more than 60 ps, in good agreement with our experiment
 265 observations. We would like to point out that this kind of wake
 266 emission has been widely observed in the simulation of x-ray
 267 amplification in gas plasma [22].

268 Backward 337.1-nm ASE is much weaker than in the
 269 forward case. This is due to the particular geometry of
 270 longitudinal pumping. When the spontaneous emission pho-
 271 tons propagate in the same direction as the IR pump beam,
 272 they interact continuously with a newly created population
 273 inversion. On the contrary, in the case of backward ASE,
 274 where the ASE counterpropagates with respect to the IR
 275 pump beam, the population inversion experienced by the
 276 spontaneous emission photon is depleted gradually by the
 277 forward ASE that follows the IR pump pulse. In fact, due
 278 to the short lifetime of the gain $\tau_g = 13$ ps, the effective
 279 amplification length for a spontaneous photon propagating in
 280 the backward direction is $l = c\tau_g = 4.2$ mm, which is much
 281 less than the 30 mm geometrical length of the plasma filament.
 282 After propagating through the 30 mm of N_2 plasma, the pulse
 283 duration (FWHM) of the backward ASE is found to be 77 ps,
 284 in good agreement with the autocorrelation measurement [see
 285 Fig. 4(c)].

286 What is the origin of the temporal modulation of the
 287 lasing observed mainly in the backward geometry? Actu-
 288 ally, the lasing line at 337.1 nm, corresponding to the P
 289 branch of the $C^3\Pi_u^+ \rightarrow B^3\Pi_g^+$ transition, is composed of
 290 three finely separated branches P_1 , P_2 , and P_3 , due to the
 291 interaction between orbital angular momenta and spin [25,26].
 292 The separation between these lines is about 0.33 Å [25],
 293 which is beyond the resolution of our spectrometer. The
 294 fast collisional excitation process brings the molecules in a
 295 coherent superposition of the three branches. An intensity
 296 modulation in the temporal domain is then expected due to
 297 quantum beating between them. The corresponding temporal
 298 modulation period T can be expressed as $T = \lambda_0^2/(c\Delta\lambda)$,
 299 where $\lambda_0 = 337.1$ nm is the central wavelength of lasing

emission, c is the light speed, and $\Delta\lambda = 0.033$ nm is the
 wavelength separation of the three branches. Therefore, one
 obtains $T = 11.5$ ps, in good agreement with the experiment
 result. Due to the employment of the slowly varying envelope
 approximation, we are treating the field envelope, not the laser
 field. As a result, the beating effect cannot directly appear in
 our Maxwell-Bloch simulations. Thus, we postprocessed our
 electric field data assuming an interference effect between
 sublevels to examine if the experimental oscillations were
 caused by the beating. When the arbitrary phases between the
 three fields are correct, the position of experimental maxima
 and minima are retrieved. The simulation results agreed well
 with the experimental ones, as shown in Fig. 4.

How far are we from obtaining a backward lasing from
 filaments in air? Our results help define a strategy to reach
 this goal. The small ratio between backward and forward ASE
 signal is due to the difference in optical gain path length. It is
 clear that the effective optical gain length must be increased.
 This can be achieved along the lines suggested in Ref. [9] by
 generating a series of aligned filaments. The weak backward
 ASE from the most remote filament would be amplified in
 successive steps by crossing the other filaments downstream.
 Another necessity is to reduce the quenching effect of oxygen
 molecules. We are presently investigating approaches towards
 that goal.

V. CONCLUSION

In summary, we have performed experimental measure-
 ments and numerical simulations of the temporal dynamics
 of the bidirectional 337.1-nm lasing pulse from filaments
 in nitrogen gas pumped by circularly polarized femtosecond
 laser pulses. The emission in both the ASE and externally
 seeded regimes was examined with time-resolved techniques.
 The 337.1-nm ASE pulse in the backward direction has a
 pulse duration around 85 ps, much longer than that of the
 forward one (20 ps), and a much weaker peak intensity. In both
 forward and backward directions, the seeded emission presents
 a shorter pulse duration and a much enhanced radiation
 intensity compared to the corresponding ASE. Numerical
 simulations based on a nonadiabatic Maxwell-Bloch equation
 agree well with the experimental results and reveal that this
 difference originates from the traveling excitation scheme of
 a pump laser and the limited lifetime of optical gain. These
 results provide valuable temporal information concerning the
 337.1-nm radiation from free-space nitrogen-gas plasma
 pumped by circularly polarized 800-nm femtosecond laser
 pulses, which can find important applications in lasing-based
 stand-off spectroscopy in the atmosphere.

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