Supplemental Document

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Time-resolved study of laser emission in nitrogen gas pumped by two near IR femtosecond laser pulses: supplement

Rostyslav Danylo,[1,2] **Guillaume Lambert,**[1] **Yi Liu,**[2,3] **Vladimir Tikhonchuk,**[4,5] **Aurélien Houard,**[1] **and André Mysyrowicz**[1,*]

[1] *Laboratoire d'Optique Appliquée, ENSTA Paris, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, 828 Boulevard des Maréchaux, 91762 Palaiseau cedex, France*
[2] *Shanghai Key Lab of Modern Optical System, University of Shanghai for Science and Technology, 516, Jungong Road, 200093 Shanghai, China*
[3] *CAS Center for Excellence in Ultra-intense Laser Science, 201800, Shanghai, China*
[4] *University of Bordeaux-CNRS-CEA, CELIA, UMR 5107, 33405 Talence, France*
[5] *ELI-Beamlines, Institute of Physics, Czech Academy of Sciences, 25241 Dolní Břežany, Czech Republic*
[*] *Corresponding author: andre.mysyrowicz@ensta-paris.fr*

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Time-resolved study of laser emission in nitrogen gas pumped by two near IR femtosecond laser pulses: Supplementary Materials

1. DETAILED EXPERIMENTAL SETUP

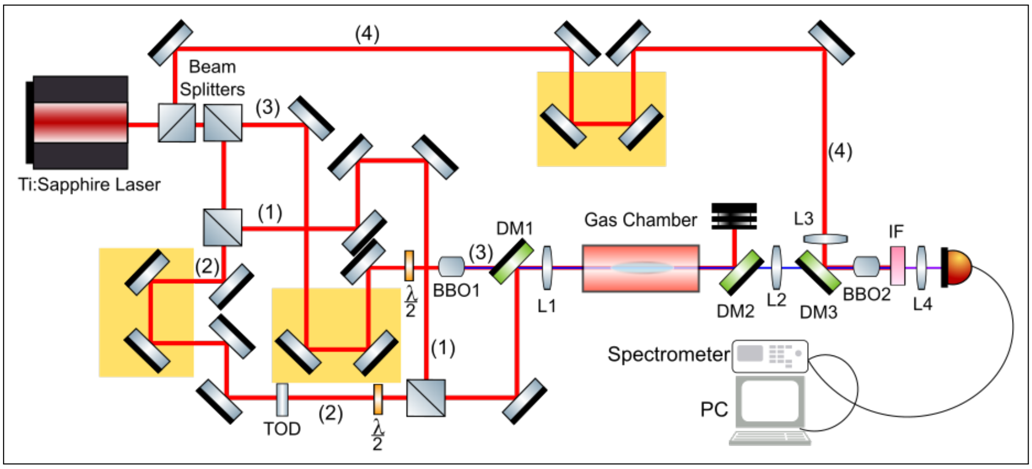

Fig. S1. Experimental setup for time-resolved measurements of lasing effect.

The experimental setup is shown on Figure S1. Ti:Sapphire femtosecond laser system ALPHA100 generates 800 nm laser pulses with 45 fs duration at 100 Hz repetition rate; maximal output pulse energy from the laser reaches $\simeq$15 mJ. Using a kit of beam splitters, the initial laser pulses is separated in four arms of the setup. The first arm (1) serves as 800 nm pump where the dominant part of the initial pulse energy is stored; this part of the setup maximally avoids any transmission elements. The second arm (2) plays a role of control where relatively weak 800 nm pulse locates. Control arm contains Tunable Optical Density (TOD) to vary control pulse energy and Half-Waveplate ($\lambda/2$) to rotate the polarization of control pulse as well as motorized translation stage to tune temporal delay between pump and control pulses. Pump and control pulses are recombined with another Beam Splitter. The third arm (3) serves as externally injected seed. This seed around 391 nm (bandwidth 1.2 nm) is generated in a strongly mismatched BBO crystal (BBO1), another Half-Waveplate ($\lambda/2$) is installed before the crystal to orient properly the polarization of generated external seed. The arm contains mechanical delay line, the temporal position is fixed in time and seed pulse arrives $\approx$300 fs after pump pulse to achieve the highest amplification ratio:

$$ratio = \frac{signal\ at\ 391\ nm}{external\ seed\ at\ 391\ nm} - 1 \tag{S1}$$

We use a Dichroic Mirror (DM1) (high reflector at 800 nm and high transmitter at 400 nm with a cutoff wavelength at 490 nm) to recombine pump and control with external seed pulse. Then all three pulses are focused with f = 40 cm lens (L1) into the gas chamber filled with pure Nitrogen at low pressure (p = 10-100 mbar), corresponding to a numerical aperture of 0.019 for pulses (1) and (2). 800 nm pump pulse forms a column of low-density plasma about 1 cm length. This plasma channel serves as a gain medium where singly ionized Nitrogen molecules emit laser-like emission at 391.4 nm. After the gas chamber 800 nm pump and control pulses are suppressed by another Dichroic Mirror (DM2), only external seed and amplified 391.4 nm emission pass through. Then this emission is recombined by the last Dichroic Mirror (DM3) with 800 nm scan pulse from the fourth arm. The fourth arm (4) contains an extremely weak 800 nm scan pulse and motorized delay stage to tune temporal position of scan pulse. Both scan pulse and amplified 391.4 nm emission are focused by the lenses L2 and L3 (both with a focal f = 30 cm) in the second mismatched BBO crystal (BBO2). BBO2 is optimized for Sum Frequency Generation (SFG) at ∼261 nm when mixing 391.4 nm and 800 nm wavelengths. SFG permits to detect the temporal profile of 391.4 nm emission. Interference Filter (IF) with 10 nm transmission bandwidth is used to cut 800 nm scan pulse and 391.4 nm amplified emission and transmits only SFG signal around 261 nm. SFG is collected by Fused Silica lens (L4) and detected by a fiber spectrometer Avantes, or by a high resolution spectrograph Princeton Instrument.

2. SATURATION OF LASING EFFECT

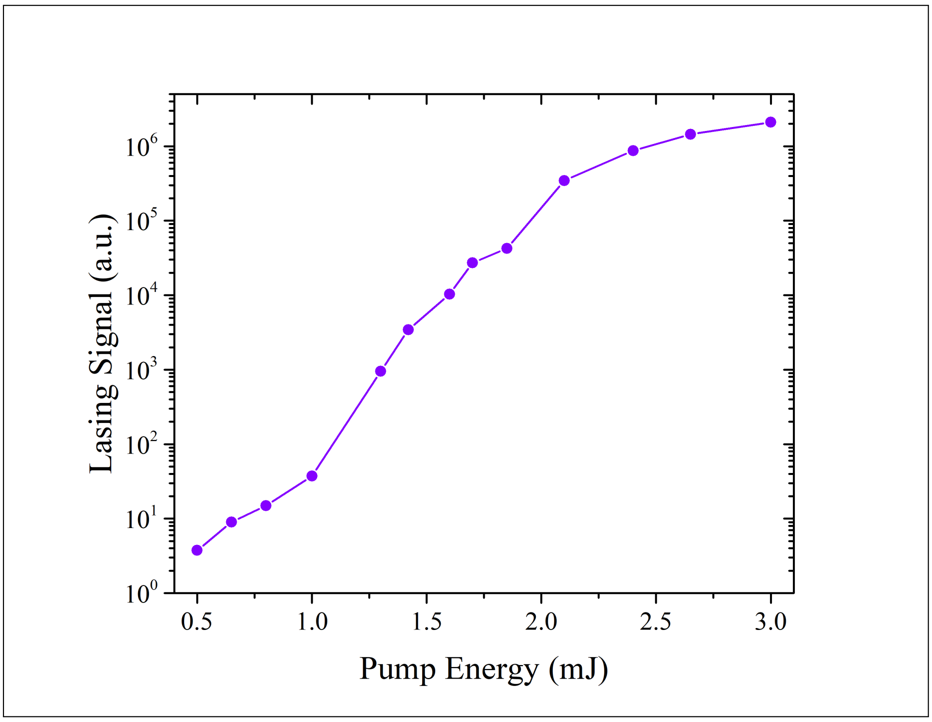

Fig. S2. Self-seeded lasing at 391.4 nm as a function of 800 nm pump pulse.

Previously, measurements of lasing from N_2^+ as a function of 800 nm pump pulse energy served to prove laser-like properties of the emission at 391.4 nm. Indeed, this dependence demonstrates a net threshold character and the emission disappears below a critical value pump pulse [1]. Another common property of any laser amplification is the saturation of the gain. Our aim in this experiment was to distinguish pump energy region where 391.4 nm gain remains exponential and the energy value when saturation takes place. We use the same focusing geometry and Nitrogen gas pressure as in Figure S1. Threshold of lasing estimates as 0.5 mJ and up to ≃2.0 mJ lasing responses as an exponential function. After 2.0 mJ saturation of lasing starts. Based on these results, we use energies around 1-2 mJ in all quenching experiments (Figures 2-5 in the main text).

3. QUANTUM ERASING OF LASING EFFECT FROM N_2^+: SIMULATIONS

Numerical simulations describing the quenching of lasing effect from N_2^+ consist in four subsequent steps: (i) modeling of the Nitrogen gas ionization and initial excitation by the 800 nm pump pulse; (ii) evolution of the excited Nitrogen ions in the plasma with the A-X coherence maintained by the post-pulse and subsequent lasing triggered by the external seed; (iii) perturbation of plasma filament by 800 nm control pulse and re-excitation of N_2^+; (iv) evolution of the re-excited Nitrogen ions and lasing from the redistributed system.

The first step of simulation refers to the solution of density matrix equations in a prescribed high laser field by taking into account ionization and excitation of Nitrogen molecular ions in the ionic ground level $X^2\Sigma_g^+(0)$ and four excited states: $X^2\Sigma_g^+(1)$, $A^2\Pi_u(2)$, $A^2\Pi_u(3)$ and $B^2\Sigma_u^+(0)$. This numerical module is described in Section 2 of Reference [2]. Amplitude of the pump pulse, its wavelength and duration come from the experiment. At the end of pump pulse, the ionization level, populations in all four excited states and all polarizations are evaluated and averaged over a random molecular orientation. Populations of the states $X^2\Sigma_g^+(0)$, $A^2\Pi_u(2)$ and $B^2\Sigma_u^+(0)$ and the corresponding polarizations are considered as initial conditions for the second step: simulation of the amplification process at 391.4 nm. In the example of Figure 4 of the main text, the ionization reaches 10.4%, populations in $X^2\Sigma_g^+(0)$, $A^2\Pi_u(2)$, $A^2\Pi_u(3)$ and $B^2\Sigma_u^+(0)$ states are 37.3%, 10.0%, 17.1%, 7.1% and 28.5% respectively. The polarization corresponding to the $A^2\Pi_u(2)$-$X^2\Sigma_g^+(0)$ transition is about 0.12, the other two polarizations of interest, $B^2\Sigma_u^+(0)$-$X^2\Sigma_g^+(0)$ and $B^2\Sigma_u^+(0)$-$A^2\Pi_u(2)$, are very small, less than 10^{-4}. Inversion of populations between levels B and A permits seed amplification in the V-scheme.

In the second step, a system of Bloch equations with three levels coupled to two electromagnetic fields corresponding to the transitions $B^2\Sigma_u^+(0)$-$X^2\Sigma_g^+(0)$ and $A^2\Pi_u(2)$-$X^2\Sigma_g^+(0)$ has been solved. This numerical module is described in Section 3 of Reference [2]. Coherence was maintained by an exponentially decaying in time post-pulse. The amplitude of the post-pulse, the decay time and the frequency detuning from the $A^2\Pi_u(2)$-$X^2\Sigma_g^+(0)$ transition are not measured in experiment and considered as free parameters in the simulation. Lasing is initiated by a weak external seed of 0.2 ps duration, injected 300 fs after pump pulse. A double peak in Figure 4 of the main text just after the seed pulse corresponds to the spontaneous emission due to nonzero polarizations. It is followed by a much stronger emission due to the parametric amplification starting at time of $\sim$1 ps. Spatial distribution of populations and polarizations along the filament at the moment of control pulse arrival are stored and taken as the input for the third step of simulation.

The interaction of the 800 control pulse with the filament is considered with density matrix equations as in the first step but high-field ionization was not taken into account and the N_2^+ system is reduced to three levels only: $X^2\Sigma_g^+(0)$, $A^2\Pi_u(2)$ and $B^2\Sigma_u^+(0)$. The control pulse is 2-3 times weaker than the pump pulse and therefore only the coupling to three resonant levels is important. However, the control pulse amplitude is still sufficiently high to produce a major perturbation in the populations, in particular, by promoting ions from $X^2\Sigma_g^+(0)$ to $A^2\Pi_u(2)$ level and to strongly modify the polarizations. In the considered case, before the control pulse arrival there is still a significant inversion $n_B > n_A$ with about 19% molecules in the level $A^2\Pi_u(2)$ and 26.5% in the level $B^2\Sigma_u^+(0)$. The control pulse increases population at the A level up to 22%, further it destroys the coherence between X-A and X-B polarizations, so than amplification in the V-scheme cannot proceed anymore.

Population and polarization distributions along the filament after the end of control pulse are stored and used as the input for the system of Bloch equations, same as in the second step but with updated input. Gain was still observed at this stage of simulation but the signal amplitude was significantly reduced by a factor 30-100 depending on the control pulse amplitude.

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