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## Laser emission at 428 nm in $N_2^+$ : competition between two and three levels amplification

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### ABSTRACT

The lasing of  $N_2^+$  at 428 nm is studied as a function of the delay  $dt$  between two 800 nm ultrashort pumping pulses from a Ti:Sapphire laser. This lasing corresponds to a population-inverted transition between levels  $B^2\Sigma_u^+(0)$  and  $X^2\Sigma_g^+(1)$  of the singly ionized nitrogen molecule. An important erasing of the laser emission is observed under pumping with two consecutive pulses separated by large delays  $-20 \text{ ps} < dt < +20 \text{ ps}$ . From an analysis of the quenching results, we conclude that the mechanism of lasing corresponds to a coherent two-photon amplification scheme. Our results show that a two-photon process can dominate over a population-inverted one-photon amplification.

When an intense ultrashort laser pulse from a Ti:Sapphire laser passes through air or a gas of nitrogen, it creates an underdense plasma column by high field ionization<sup>1</sup>. A strong optical amplification is observed inside this plasma column upon injection of a short seed pulse at a wavelength of 391 nm or 428 nm<sup>2-4</sup>. These lines correspond to transitions  $B^2\Sigma_u^+(0) \rightarrow X^2\Sigma_g^+(0)$  and  $B^2\Sigma_u^+(0) \rightarrow X^2\Sigma_g^+(1)$  (hereafter denoted B0  $\rightarrow$  X0 and B0  $\rightarrow$  X1) in singly ionized nitrogen molecules, see Fig. 1(a). Despite a considerable amount of publications on this subject, there is still no consensus about the lasing mechanism. Three different processes have been proposed for the lasing at 391 nm: conventional amplification due to B0/X0 population inversion<sup>5-9</sup>, amplification without B0/X0 population inversion due to a coherent two-photon coupling between three levels B, X and  $A^2\Pi_u(2)$  in a V-scheme arrangement<sup>10,11</sup> and amplification due to an inversion of rotational populations between B and X states<sup>12,13</sup>, see Fig. 1(b) and (c) for a schematic representation of the first two processes. Recent calculations show that population inversion between B0 and X0 is very sensitive to the particular pump pulse conditions<sup>14,15</sup>. B0/X0 inversion occurs if the pump central wavelength from a Ti:Sapphire laser exceeds 780 nm and if the pump intensity exceeds  $1.5 \times 10^{14} \text{ W/cm}^2$ , but it disappears if one of these two values is decreased by less than 5%. Such a nearly equal population between B0 and X0 is supported by a recent measurement by x-ray absorption spectroscopy<sup>16</sup>. On the other hand, the same calculations show a robust population inversion between B0 and X1 levels at the same wavelengths<sup>15</sup>. Therefore, even if the case of 391 nm emission is still unsettled, it is natural to expect a conventional two levels amplification scheme to be operative for the

428 nm emission.

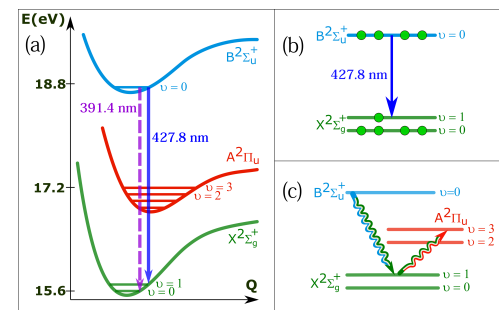


FIG. 1. (a) Energy diagram of  $N_2^+$  showing the levels relevant for lasing at 391 nm and 428 nm. (b) Schematic representation of the two levels amplification at 428 nm. (c) Schematic representation of coherent three levels amplification at 428 nm.

In order to verify this point, we have performed experiments using a sequence of two 800 nm laser pulses from a Ti:Sapphire laser with the goal to study the laser emission at 428 nm as a function of the delay between them, see Fig. 2 for experimental setup. Such a double pump experiment has been reported before for the 391 nm emission<sup>17-20</sup> and more recently for the 428 nm emission<sup>21</sup>. Surprisingly, analysis of our results indicate that the coherent three levels coupling plays the predominant role in the lasing process at 428 nm. We obtained the same conclusion from an analysis of the dou-

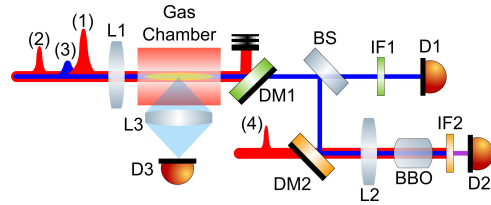


FIG. 2. Experimental setup. 800 nm pump pulse (1), 800 nm control pulse (2) and broadband 428 nm seed pulse (3) are focused by focusing lens L1 ( $f = 40$  cm or 50 cm) in gas chamber filled with pure nitrogen or air. Seed pulse (3) is injected  $\sim 300$  fs after pump pulse to achieve the strongest amplification at 428 nm. Pump and control pulses are reflected by dichroic mirror DM1. Transmitted lasing is splitted by beam splitter BS. One part is additionally filtered by interference filter IF1 and captured by detection system D1. The other part of lasing signal is recombined with 800 nm scan pulse (4) by dichroic mirror DM2. These pulses are then focused by 30 cm lens L2 in a BBO crystal ( $\theta=44.3^\circ$ ) and generate sum frequency signal at 281 nm permitting to measure the temporal profile at 428 nm lasing. Generated wavelength at 281 nm is selected by another interference filter IF2 and captured by detector D2 (for a more detailed description, see Refs.<sup>17,20</sup>). Sideway fluorescence signal is collected by lens L3 ( $f = 10$  cm) to the detection system D3.

ble pulse quenching of the lasing at 391 nm<sup>17,20</sup>.

An example of amplification of the emission at 428 nm upon injection of a weak seed pulse is shown in Fig. 3(a) for a pump pulse alone propagating in pure nitrogen at 100 mbar pressure. The lasing signal with no external seed is plotted in Fig. 3(b) as a function of incident pump pulse energy at atmospheric pressure. The lasing signal increases by 4 orders of magnitude when the pump pulse energy is increased by a factor  $\sim 1.5$  from 3.0 to 5.0 mJ, and then saturates. The saturation is related to the pump intensity clamping inside the filament<sup>22,23</sup>. Lasing saturation is excluded because the lasing signal is strongly amplified if an external seed pulse is injected, even with a pump pulse energy of 7.0 mJ. The pump intensity in the clamped regime is estimated to be around  $1.5 \times 10^{14}$  W/cm<sup>2</sup> in air at atmospheric pressure and is increased by a factor 2 in pure nitrogen<sup>24</sup>, which explains the difference in lasing signals between air and pure nitrogen shown in Fig. 3(b). The seeded lasing emission temporal profile presented in Fig. 3(c) for a pure nitrogen gas shows a lag in the onset of emission, with the lasing peak retarded from the pump and seed pulses by 3-4 ps depending on gas pressure. A similar delay of laser onset has been observed for the 391 nm emission<sup>25,26</sup>.

The effect of double pulse excitation on the emission at 428 nm in pure nitrogen gas with and without external seed pulse is shown in Figs. 4 and 5. Lasing amplification is shown in Fig. 4(a) as a function of negative delay  $dt < 0$  between seeded pump and control pulse, for a nitrogen gas pressure of 100 mbar and for two values of the control pulse energy of 100 and 500 J, where negative delay  $dt$  implies that the control pulse arrives after the pump. The level of lasing signal with pump alone is shown as the horizontal green line. The

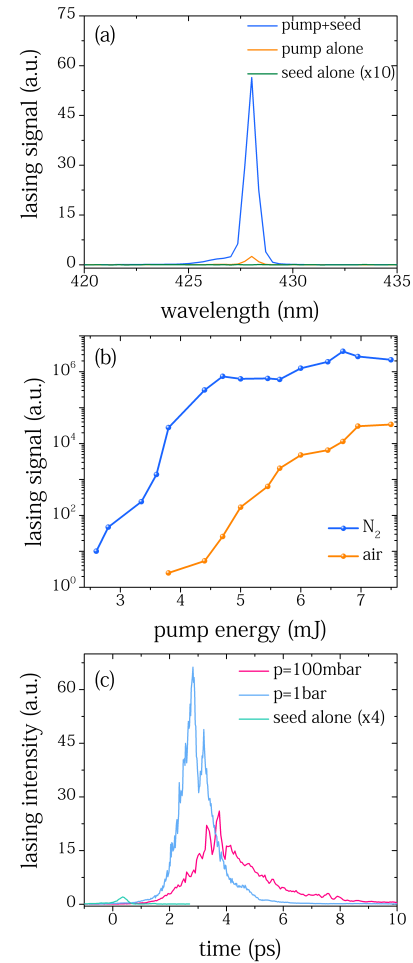


FIG. 3. (a) Spectrum of externally seeded and self-seeded lasing signals at 428 nm. Pump pulse at 800 nm of 7.8 mJ energy was focused by a  $f = 50$  cm lens in pure nitrogen of 100 mbar pressure. (b) Saturation curve of self-seeded lasing in pure nitrogen and air at atmospheric pressure. The other experimental conditions are the same as in (a). (c) Temporal profile of externally seeded lasing at 428 nm at two different pressures of pure nitrogen. External seed pulse arrives nearly 300 fs after pump pulse. The other parameters are the same as in (a).

lasing signal is reduced by two orders of magnitude when the control pulse with 500 J energy arrives with a delay of 1-3 ps after the pump pulse. Smaller laser signal reductions are observed at delays up to -20 ps. Effect of double pulse excitation for positive delays  $dt > 0$  is shown in Fig. 4(b) for a nitrogen

pressure of 100 mbar and a control pulse energy 100 and 500 J. Positive delay  $dt$  means that the control pulse arrives before the pump. The quenching effect is weaker than for short negative delays and becomes negligible with a control pulse energy of 100 J, below threshold of plasma formation. It also shows narrow lasing spikes when the pump pulse arrives at times corresponding to revivals of coherent rotational wavepackets of neutral nitrogen molecules induced by the probe pulse around  $dt = 4.19$  ps, 8.38 ps and 12.57 ps<sup>13</sup>. A zoomed view of panel (b) is shown in Fig. 4(c) together with a fit to the calculated partial alignment of neutral N<sub>2</sub> molecules occurring during revivals of coherent rotational wavepackets<sup>12,27-29</sup>. As shown in Fig. 5(a), lasing revivals are positive if both pump and control pulses have the same polarization, but change sign for orthogonal pump/control polarizations.

In contrast to the suppression of the lasing effect in the forward direction by the control pulse, no suppression has been observed in the fluorescence corresponding to the transitions B0→X0 and B0→X1, as shown in Fig. 5(b,c) for pump and control pulses of 3.0 and 2.5 mJ. As expected, an enhancement of the fluorescence is observed when pump and control pulses overlap in time (see discussion below). The difference by a factor of 3 in the intensities of fluorescence B0→X0 (391 nm) and B0→X1 (428 nm) is explained by the corresponding difference in the Einstein coefficients (see Ref.<sup>14</sup>, Tab. 1).

A remarkable feature of our observations is the appearance of lasing quenching. It indicates that, during a time interval  $dt$  of several ps, the combination of two successive pump and control pulses does not add to, but instead suppresses a large fraction of the pump induced lasing signal. We now examine possible explanations of this quenching in the frame of the two-levels, and three-levels amplification schemes. (An explanation of the quenching in the frame of the lasing due to inversion of rotational populations between B0 and X1 can be excluded because in this scenario lasing occurs only at specific time intervals). Several explanations have been considered with the two-levels scheme:

1. *Depletion of B0 population by the second oncoming pulse preventing lasing by the first pulse.* Fluorescence results in the presence of pump and control pulses shown in Fig. 5(b,c) show no decrease, but rather a small increase of B0 population. Therefore, this scenario can be excluded.
2. *Depletion of population at B0 level by the second pulse through a transfer of B0 ions to higher lying  $C^2\Sigma_u^+$  state.* Again, the fluorescence results shown in Fig. 5(b,c) for different delays and for a control pulse of increased intensity show no decrease of B0 population. Moreover, the intensity of the control pulse is too weak to produce a notable transfer from B0 to  $C^2\Sigma_u^+$  level lying 5 eV above B0, which would require a 4-photon process.
3. *Increased population at X1 level by the second oncoming pulse.* This level can only be optically populated by a transfer of ions from the higher excited levels A and B. This hypothesis might in principle explain the suppression of lasing at negative delays, but calculations of the population of ions in excited states A and B show a small effect of the control pulse. Moreover, it does not explain the suppression of emission at positive delays.

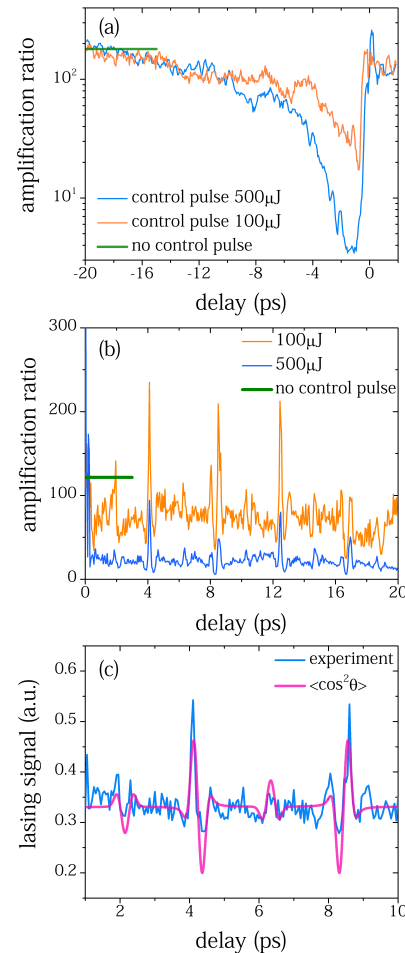


FIG. 4. (a) Erasing effect of externally seeded lasing at 428 nm for different control pulse energies. Negative delay implies that pump pulse comes first in the gas chamber. Pump pulse of 2.3 mJ energy is focused with  $f = 40$  cm focusing lens in pure nitrogen of 100 mbar. Amplification ratio is defined as the seeded lasing signal divided by the injected seed signal. (b) Alignment-dependent erasing effect of externally seeded lasing. Pump, seed and control pulse are of the same polarization. Positive delay means that control pulse arrives first in the gas chamber. The other experimental parameters are the same as in (a). (c) Simulation of the molecular alignment of neutral nitrogen molecule in the intense laser field. Experimental data are taken from (b).

4. *Negative interference between the lasing signals induced by pump and control pulses.* The lasing signal from pump pulse,

which is amplified by the seed pulse, is higher by 2-3 orders of magnitude than the lasing signal induced by control pulse. Therefore, any destructive interference would be minimal due to the large difference in lasing amplitudes.

5. *Plasma diffraction.* This scenario implies that the plasma generated by the first pulse diffracts the second pulse. This hypothesis may possibly explain a suppression of lasing at positive delays, assuming that it reduces slightly the intensity of the pump pulse, but it cannot explain the observed strong decrease of lasing signal at negative delays.

We now discuss the quenching in the frame of the coherent two-photon amplification via the V-scheme shown in Fig. 1(c). Considering first negative delays, we note that the quenching results of the 428 nm emission are very similar to those reported for the 391 nm emission and have the same explanation<sup>17,20</sup>. In order to be effective, the V amplification scheme requires the presence of B-X and A-X coherent couplings and a population inversion between B and A levels. To close the V loop, the third level is A3, which has the largest dipole moment for the transition to X1, see Fig. 1(c). The process of amplification within the V-scheme proceeds on a time scale of a few ps<sup>11</sup>. Therefore, a control pulse arriving within a few picoseconds after the pump pulse perturbs the coherence between the B0-X1 and A3-X1 transitions and thus effectively suppresses the two photons coupling<sup>20</sup>. The loss of coherence together with the increase of population at A3 level terminates the lasing process. This hypothesis is in agreement with the fact the quenching of emission is observed independently on the polarization of the control pulse, see Fig. 5(a). Moreover, the measured suppression time in Figs. 4(a) and 5(a) of 5-10 ps is in agreement with the measured duration of the 428 nm emission of 4-6 ps shown in Fig. 3(c).

We consider now positive delays, where the control pulse arrives first in the sample. A longer duration of the quenching is observed, which translates the fact that it is no more limited by the duration of the lasing signal but depends on the plasma lifetime. The lasing revivals observed around 4.19, 8.38 and 12.57 ps fit very well the calculated curve of alignment of neutral molecules<sup>12,27-29</sup> and can be explained as follows: The control pulse, arriving first in the sample, aligns after  $\sim 100$  fs a fraction of neutral molecules along its polarization direction. The coherent rotational wavepacket thus formed experiences spontaneous revivals at each half period of rotation of neutral molecules, leading to revivals of molecular alignment. The pump pulse of same polarization arriving in coincidence with a revival produces a stronger ionization and produces preferentially these ions in the B state, since the parallel orientation of molecule axis and pump electric field favors dipole transitions with magnetic quantum number 0. Since amplification takes place 1-3 ps after alignment, at that time orientation of molecules is random and therefore the V scheme amplification can proceed as usual. Therefore, one expects the appearance of a stronger lasing signal if the pump pulse of same polarization arrives at these particular moments. Conversely, one expects an increased quenching when the pump pulse is perpendicular to the molecular axis. This is in agreement with the observation shown in Fig. 5(a). The relatively small decrease of the 428 nm emission at other positive delays is probably

related to the weak plasma ionization produced by the weaker control pulse with 500 J energy. When the control pulse energy is decreased down to 100 J, nearly no lasing reduction takes place, as shown in Fig. 4(b). While the level of excitation is not sufficient to produce an observable emission, an intensity of control pulse of  $1 \times 10^{14}$  W/cm<sup>2</sup> leads to an ionization level of 0.3%<sup>30</sup>, which can slightly refract the pump pulse arriving a few ps later. For a control pulse of lower energy, the ionization is very low and quenching nearly disappears, see Fig. 4(b).

The significant suppression of the 428 nm emission implies a dominant role of the three-levels process, which is not what is expected from the theory<sup>11</sup>. Calculations of the B-X emission when both two levels (B-X) and three levels (B-X-A) transitions are allowed predict that the direct B-X transition dominates as soon as there is a B0/X0,1 population inversion<sup>11</sup>. However, the N<sub>2</sub><sup>+</sup> molecule is a complex object and a combination of only three levels with associated long-lived coherent couplings is probably not sufficient for an interpretation of this experiment. Simultaneous transitions from B0 to X0,1 and from A0,1,2,3 to X0,1 have to be considered. Interferences between all these transitions could make a strong effect on the probability of emission. This requires a more detailed theoretical analysis.

We now discuss the interference of pump and control pulses at small delays around  $dt = 0$ . A recent manuscript<sup>21</sup> reported an extremely enhanced lasing in air at 428 nm by pumping ambient air with two pulses of comparable intensity intercepting at a small angle. We find a similar amplification for a delay close to 0, but we differ in the interpretation. This gain of lasing signal is explained, in our opinion, by a positive interference between the two overlapping laser fields. The interference of the pump and control pulses of comparable intensities explains the enhanced lasing signal at 428 nm. By combining the unseeded pump and control pulses of energy 2.5 mJ and 1.5 mJ, we increased lasing signal by five orders of magnitude in agreement with Fig. 3(b), but the lasing signal was never higher than the saturated value obtained with a single pulse.

In conclusion, we have observed an important quenching of the laser emission between levels B0 and X1 under excitation of nitrogen gas with two consecutive femtosecond pulses from a Ti:Sapphire laser. Several features indicate that this quenching effect can be explained only if a coherent three levels coupling in a V-scheme arrangement plays a primordial role in the large amplification of 428 nm emission in the plasma column. The three level amplification process is promising for various applications including atmospheric remote sensing and x-ray or gamma ray coherent amplification.

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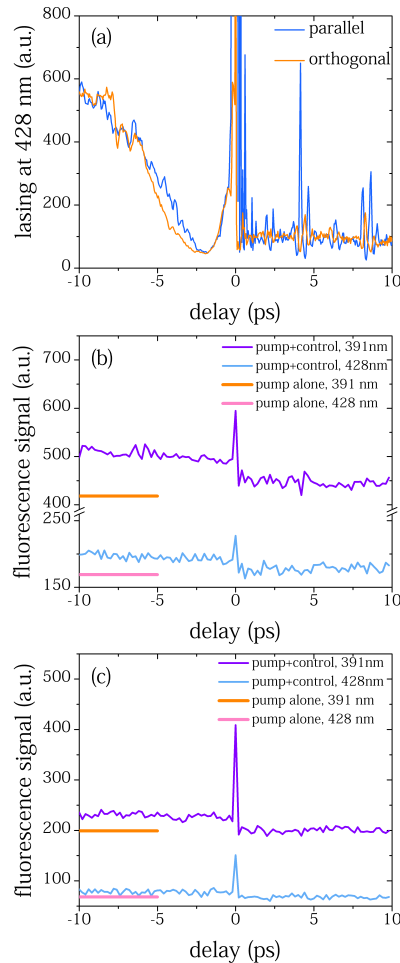


FIG. 5. (a) Erasing of lasing at 428 nm versus the time delay without injected seed. Pump pulse energy is 2.7 mJ, control pulse energy is 500 J of parallel (blue) or perpendicular (orange) polarization. (b) Fluorescence of  $B0 \rightarrow X0$  transition at 391 nm (purple curve) and 428 nm (blue curve) versus the time delay between the pump and control pulses in the presence of external seed. Pump and control pulse energies are 3.0 and 2.5 mJ. (c) Fluorescence of  $B0 \rightarrow X0$  transition at 391 nm (purple curve) and 428 nm (blue curve) versus the time delay between the pump and control pulses when no external seed is injected. Pump and control pulses are the same as in (b).

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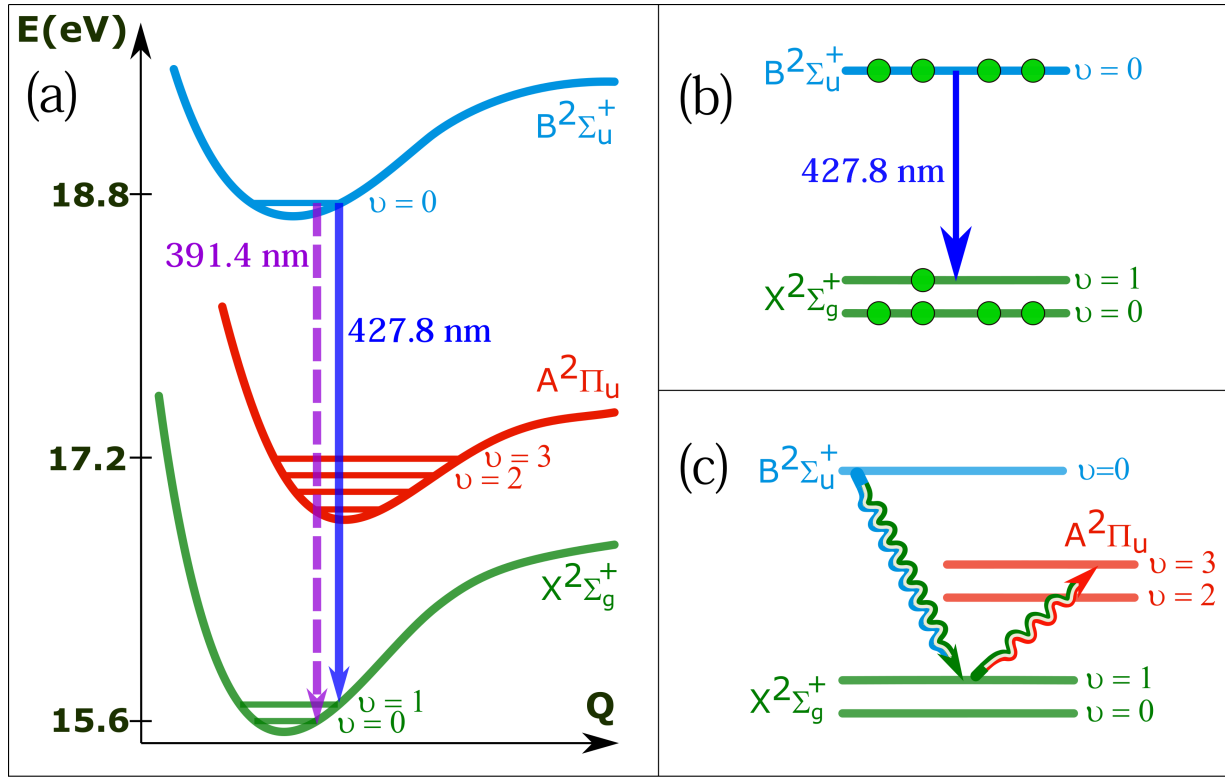
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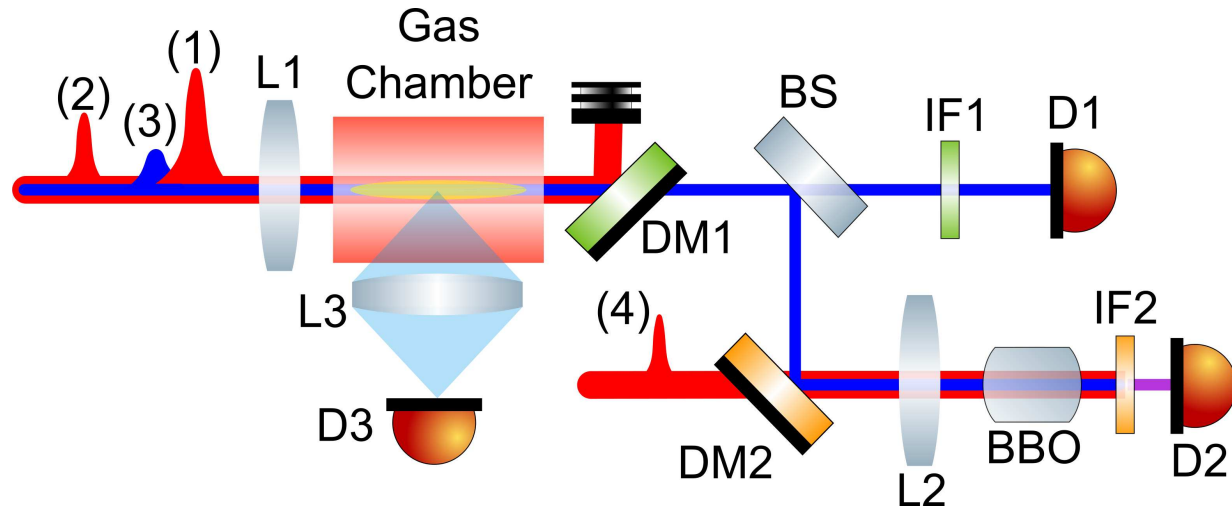
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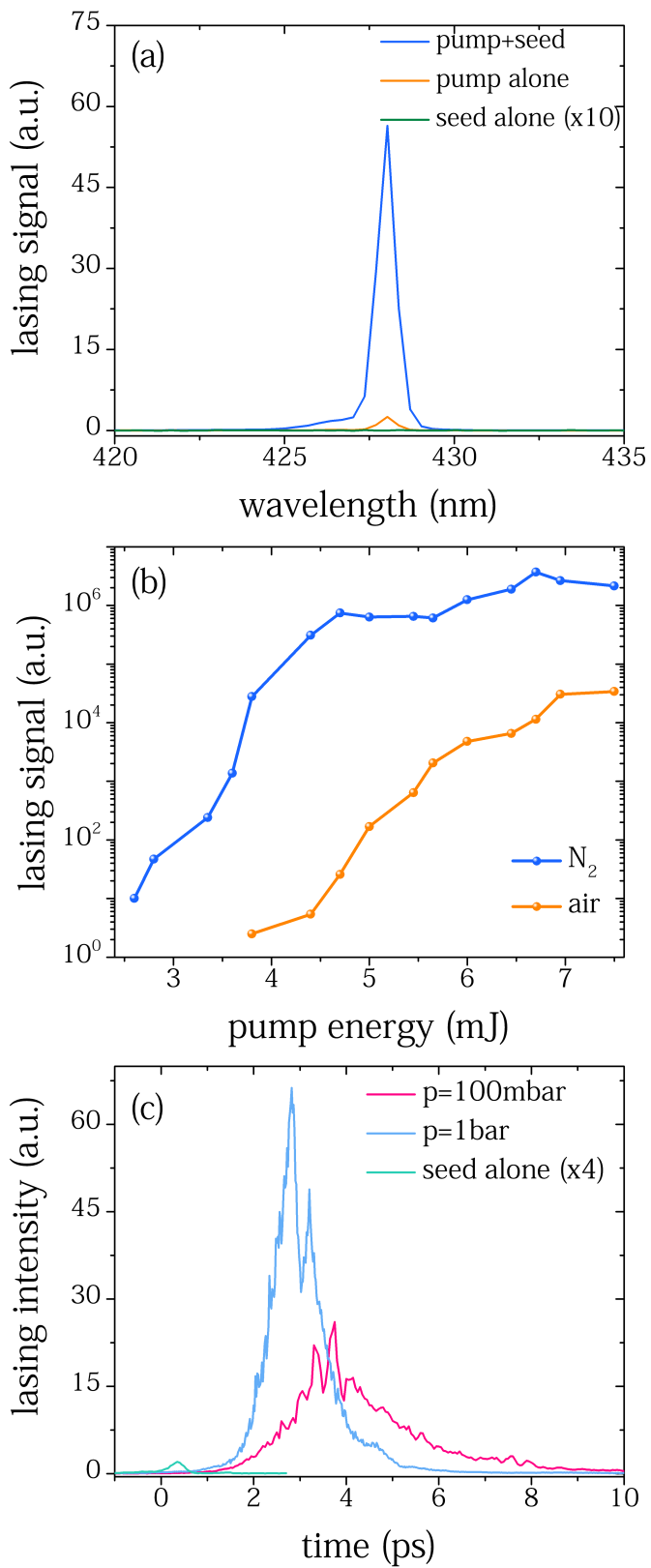
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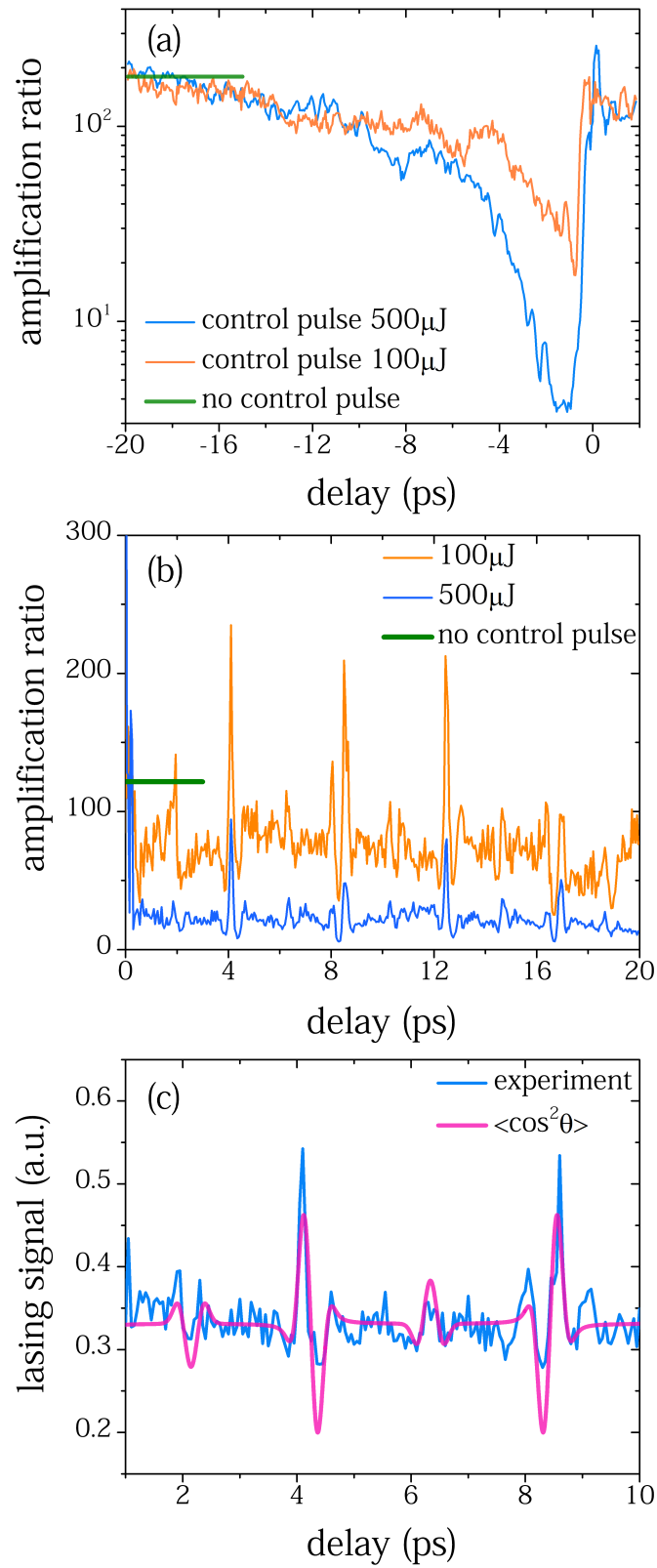
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