EPJ Web of Conferences **41**, 10004 (2013)
DOI: 10.1051/epjconf/20134110004

Intense, directional UV emission from molecular nitrogen ions in an adaptively controlled femtosecond filament

G. Andriukaitis[1], J. Möhring[2], D. Kartashov[1], A. Pugžlys[1], A. Zheltikov[3,4], M. Motzkus[2], A. Baltuška[1]

[1]Photonics Institute, Vienna University of Technology, A-1040 Vienna, Austria
[2]Physikalisch-Chemisches Institut, University Heidelberg, D-69120 Heidelberg, Germany
[3]Department of Physics and Astronomy, Texas A&M University, College Station TX, USA
[4]Physics Department, International Laser Center, M.V. Lomonosov Moscow State University, 119992 Moscow, Russia

Abstract. Using phase-shaped millijoule 1030-nm femtosecond pulses generating a filament in a cell filled with N_2, we obtain intense forward UV emission between vibrational manifolds of $B^2\Sigma$ and $X^2\Sigma$ states of N_2^+ for optimal pulse sequences. The effect is tentatively ascribed to wave-mixing between intense NIR pulses and weak supercontinuum components, resonant to the UV transitions, whereby a non-instantaneous nonlinear susceptibility is linked to rotational coherence in ions.

1 Introduction

UV emission from nitrogen in the plasma of femtosecond gas filaments has attracted attention as a prospective light source for stand-off spectroscopy [1, 2]. Distinct nitrogen fluorescence in the UV mainly results from plasma-chemical reactions in a filament which lead to the appearance of a large variety of neutral and ionic species in rotationally, vibrationally and electronically excited states. In this contribution we demonstrate, for the first time to the best of our knowledge, adaptive control over the efficiency of UV emission from nitrogen in a near-IR femtosecond filament using a spatial light-modulator (SLM). Applying an evolutionary algorithm, we determined an optimal pulse sequence to obtain intense forward-directed UV radiation. Although the studied generation mechanism shares many similarities with a seeded laser-type amplifier, the collected experimental evidence points toward a different, inversion-free mechanism—nonlinear wave-mixing between the high-intensity laser pulse, responsible for filamentation, rotational alignment/excitation and ionization, and a weak resonant component from the UV wing of the supercontinuum which covers electronic transitions between the vibrational levels of the ground ($X^2\Sigma_g$) and excited ($B^2\Sigma_u$) states.

2 Experimental results

Experiments were performed using a 0.5-kHz repetition rateYb:CaF_2 laser emitting 220 fs, 7 mJ, 1.03-μm pulses. The pulse shaper, comprising an SLM in a 4*f* fold of cylindrical mirrors, was installed between the pulse stretcher and the regenerative amplifier. The shaped laser pulses were focused with an f=1m focusing lens into a 1.5-m-long gas cell with transparent sidewalls and

Brewster input and output windows made of CaF_2. The enhancement effects reported here peak in the pressure range of 3-4 bars.

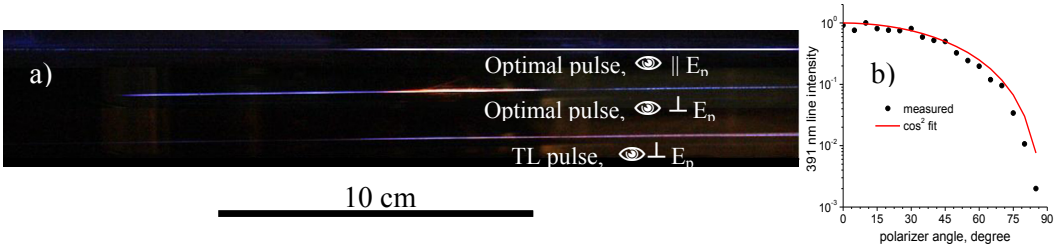

Fig. 1. a) Photographs of the filaments in the view planes parallel and perpendicular to the excitation laser polarization for an optimally shaped pulse and a transform-limited pulse. Note that the emission from N_2^+ (prominent central zone, middle trace) is strongly polarized along the laser field. b) Polarization degree measurement for the N_2^+emission at 391 nm indicating that this emission inherits the linear polarization of the weak supercontinuum.

For the detection of the UV emission in the *forward* direction, the central high-intensity filament part of the beam was blocked off and the annular transmitted beam was scanned by moving the entrance slit of spectrometer attached to an XY stage. The input pulse sequence created by the SLM was characterized by SHG-FROG in front of the focusing lens. Images of the visible fluorescence from the filament excited with optimized phase-shaped pulses compared to the case of fully compressed pulses are shown in Fig.1a. Key findings of the UV emission enhancement with phase-shaped 1030-nm pulses are summarized in Fig.2a-c.

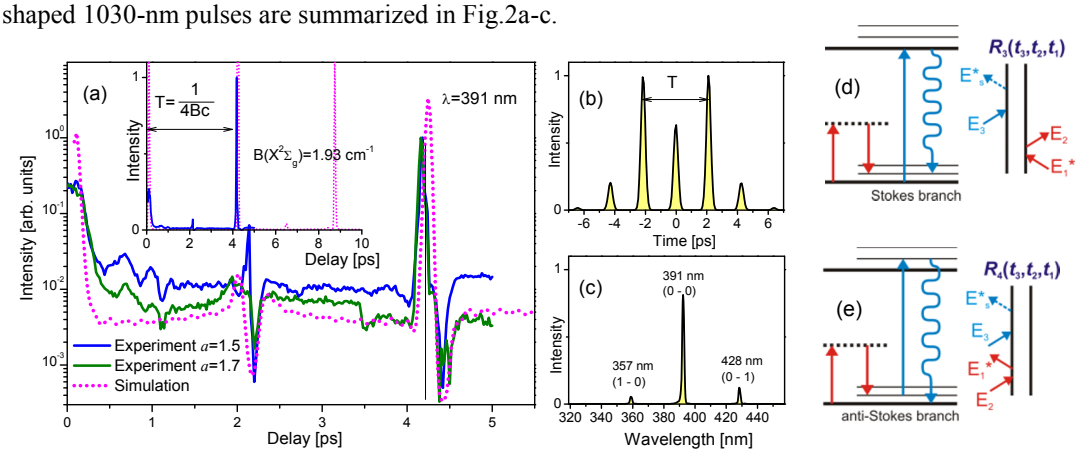

Fig. 2. a) Intensity of forward emission at 391 nm as a function of interpulse delay for a=1.5—1.7 and pressure 3 bar (solid curves). Dotted curve shows a simulation for a two-pulse stimulated photon echo assuming a non-instantaneous 3rd order nonlinearity caused by rotational revivals of N_2^+ in $X^2\Sigma_g$. b) measured optimal pulse train (a=1.6, b= 2.15 ps) corresponding to the highest 391-nm emission. Note that 4b=8.6 ps, which matches the full rotational revival of the ion in the ground ionic state. c) measured spectrum of forward UV emission with the corresponding vibrational transitions between $B^2\Sigma_u$ and $X^2\Sigma_g$ indicated in parenthesis. d) and e) give the relevant diagrams of four-wave mixing. Thick lines indicate vibrational levels of $X^2\Sigma_g$ and $B^2\Sigma_u$, thin lines represent rotational levels. The spectrum of nearly transform-limited near-IR pulses is sufficiently broad for Raman-type excitation of the rotational manifold.

To control the UV emission from nitrogen, we applied a sinusoidal mask $\varphi(\omega)=a\sin(b\omega+c)$ to the spectral phase of the compressed laser pulse, where a determines the relative amplitudes and the number of pulse replicas and b the temporal delay between them [3]. The idea behind the use of such a pulse train is that relatively weak pre-pulses in the train produce alignment of nitrogen molecules at

pulse separations corresponding to fractional revivals of the rotational wavepacket [4]. At the same time, a higher plasma concentration is expected when the intensity of the pulses in the train reaches the ionization threshold because the ionization probability for nitrogen molecules aligned along the laser polarization is about 4 times higher than for perpendicular orientation [5].

Application of a genetic algorithm for a two-dimensional (*a*- and *b*-) pulse optimization (Fig. 2b) results in a strong forward emission at 358, 391 and 428 nm consisting of very narrow spectral lines corresponding to well-known N_2^+ transitions (Fig.2c). The emission is strongly polarized along the laser polarization with a polarization contrast higher than 10^3 (Fig.1b), proving that this emission is not spontaneous. Also, we observed full correlation between the appearance of this emission and the generation of white light in the filament, suggesting that the white light formation is required for the appearance of the emission at N_2^+ lines. Noteworthy, the effect of multicolor emission at N_2^+ fluorescence lines was observed recently in a spark produced by a tightly focused IR femtosecond laser beam in air at 1.9 μm wavelength [6], and in our recent experiments with a high-power 4-μm femtosecond laser source.

3 Discussions and conclusions

Our explanation of the observed phenomenon is based on the interpretation of the rotational revival signatures in the presented shaped-pulse filamentation experiment and supporting evidence of additional time- and frequency-resolved measurements that will be presented elsewhere. Although the behavior of the observed emission resembles that of a laser-like amplifier, time-resolved data prove that spatially and temporally coherent UV emission decays over many picoseconds, well outside the temporal overlap with the resonant field of the supercontinuum pulse. The emitted signal is strongly modulated in time with the periodicities corresponding to the rotational revivals of both the ground and the excited ionic states, whereas its carrier frequency is modulated by the rotational revivals of neutral nitrogen molecules and is consistent with description given in Ref. [4]. Remarkably, all attempts to "seed" the "amplifier" at resonant UV transition frequencies invariably end up in optical losses because of resonant absorption. The four-wave mixing (FWM) scenario, outlined in Fig.2d,e, on the other hand, adequately describes all experimental observations performed to date. The investigated FWM process involves two successive interactions with a high-intensity non-resonant near-IR field, one of the roles of which, beside the generation of ions, is the excitation of their rotational Raman manifold. Subsequent interaction with a weak resonant UV field leads to an automatically phase-matched emission of intense 3rd order polarization at Stokes and anti-Stokes frequencies around the resonance. Although the precise role of each sub-pulse in the train (Fig.2b) remains to be asserted, the main effect can be captured by approximating the interaction with two most intense pulses, where at least the second main pulse generates a supercontinuum stretching into the UV. A two-pulse stimulated echo simulation, shown in Fig.2a by dotted curve, supports the validity of this approach.

References

1. Q. Luo, W. Liu, S.L. Chin, Appl. Phys. B **76**, 337-340 (2003)
2. P. R. Hemmer, R. B. Miles, P. Polynkin, T. Siebert, A. V. Sokolov, P. Sprangle, and M. O. Scully, PNAS **108**, 3130-3134 (2011)
3. D. Zeidler, S. Frey, K. L. Kompa, and M. Motzkus, Phys. Rev. A **64**, 023420 (2001)
4. J.-F. Ripoche, G. Grillon, B. Prade, M. Franco, E. Nibbering, R. Lange, A. Mysyrowicz, Opt. Comun. **135**, 310-314 (1997).
5. D. Pavičić, K. F. Lee, D. M. Rayner, P. B. Corkum, and D. M. Villeneuve, Phys. Rev. Lett. **98**, 243001 (2007)
6. J. Yao, B. Zeng, H. Xu, G. Li, W. Chu, J. Ni, H. Zhang, S. L. Chin, Y. Cheng, and Z. Xu, Phys. Rev. A **84**, 051802 (2011)