Effect of rotational wave packets on the stimulated emission of nitrogen with light filament

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Abstract

We measure forward emission of the nitrogen ion excited by light filaments at 800 nm. The radiation strongly depends on light polarization and orbital angular momentum. In all cases the rotational distribution of nitrogen molecules plays an essential role in the 428 nm stimulated emission.

Keywords: 52.38.Hb Self-focusing, channeling, and filamentation in plasmas, 33.20.Fb Raman and Rayleigh spectra (including optical scattering), 52.25.Ya Neutrals in plasmas

Light filaments [1], known as non-diffracting beams beyond multiple Rayleigh ranges, offer promising applications in remote sensing. The propagation of intense beam in air is affected by ionization, molecular and electronic Kerr effect, self phase modulation and diffraction [2, 3]. Recently there had been a great interest in controlling the stimulated emission in nitrogen molecular ions [4, 5] and nitrogen neutral molecules [6, 7, 8, 9]. We show the effect of rotational wave packet in enhancing the stimulated emission. The control of the rotational distribution of the medium is done in microscopic scale with light polarization (ellipticity) and macroscopic scale using orbital angular momentum of light.

The phenomena contributing to the emission from filaments can be categorized as: (i) direct radiation from individual atoms or molecules (uni-molecular effects, including self-phase modulation, white light generation [10]), and (ii) amplification by a group of atoms/molecules, a collective phenomenon affected by light propagation. In the latter category one finds non-resonant effects (i.e.

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9 generation and amplification of new wavelengths by four wave mixing) and res-
10 onant effects (gain by population inversion). The amplification of a particular
11 wavelength of the ultrashort pulse will proceed along a direction for which there
12 is group velocity matching [11] or Cherenkov radiation [12]. As in any laser
13 without cavity, the process of amplification has to be seeded, either by vacuum
14 fluctuation, spontaneous or conical emission.
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18 The Kerr effect introduces spectral broadening through upchirp in gases
19 with positive nonlinear index n_2 , while the negative index plasma introduces
20 a blue spectral shift [13]. Only a small percentage (0.01 to 0.1%) [14, 15] of
21 the molecules undergo ionization (higher electron densities are reported by
22 Kandidov[16]. While air is an isotropic medium, its response is no longer
23 isotropic when interacting with a polarized light. Strong field tunnel ioniza-
24 tion results in a controlled electron trajectory that can be explored in the THz
25 radiation being generated. Nitrogen and oxygen exhibit different polarizabili-
26 ties along and perpendicular to their molecular axis. As the dipole interaction
27 depends on the orientation of a molecule with respect to the field, there is a
28 preferential ionization [17] along the light polarization. For neutral molecules in
29 air and also for ions, there is a torque proportional to the projection of the dipole
30 moment along the light polarization $pE \cos \theta$, in which p is the polarizability, E
31 the electric field and θ the angle between the two vectors. In a non-perturbative
32 approach the laser field is treated classically and material quantum mechanically.
33 The rotational distribution (in “ J ’s”) is modified from its Boltzman distribu-
34 tion due to the light interaction, and for light the medium is birefringent. For
35 pulses shorter than the rotational period of the molecule (8.4 ps for nitrogen),
36 a rotational wavepacket is generated resulting in field-free alignment at delays
37 corresponding to multiple integers of the rotational period [18, 19]. Transient
38 birefringence is observed over the length of a single ultrashort pulse, leading to
39 polarization modification of the pulse itself [20].
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53 A simple setup is chosen for the investigation of forward radiation caused
54 by light filaments, as shown in Fig. 1. The 428 nm radiation is collected by
55 a monochromator with a CCD camera placed at its exit. The image on the
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Figure 1: Setup to study the forward emission at 428 nm generated in a N_2 filled cell (1 atm pressure) by a filament created with 15 mJ, 60 fs pulses at 800 nm (10 Hz repetition rate). The light polarization is controlled by a quarter wave plate and its orbital angular momentum can be changed with a spiral phase plate. The light is focused (NA=0.19) into the cell. The transmitted light is focused by a $f = 10$ cm fused silica lens onto the entrance slit of a spectrometer, calibrated with a mercury lamp using the spectral lines at 404.66 and 435.84 nm.

CCD spans a 42 nm bandwidth, is centered at 426 nm with the pixel resolution of 0.0088nm. The spectrometer is a coma-aberration corrected Czerny-Turner configuration with focal length of 0.64 meter. The image is taken with a Thorlabs CCD of 4872 x 3248 pixels (16000m-00-GE). For each measurement the image is integrated for 10 seconds to have a comparable image under various conditions. The two dimensional image on the CCD is integrated along the vertical axis and presented as a signal versus wavelength.

The forward emission at 428nm is shown in Fig. 2 (a) and (b). Similar results had been recently published [21]. The inset in Fig. 2 (a) indicates the spectroscopic ro-vibrational branches of transitions between the $B^2\Sigma_u^+$ $\nu' = 0$ to $X^2\Sigma_g$ $\nu'' = 1$, where ν is the vibrational quantum number. The integrated signal is presented as function of wavenumber (cm^{-1}), in which zero corresponds to 23374 cm^{-1} (P transition to $J = 5$). Throughout the paper the rotational lines are defined with the final state of J , so the P transition $J' = 4 \rightarrow J'' = 5$ is labeled $J = 5$. At first glance the spectrum of Fig. 2 (a) may be mistaken

as a rotational Raman spectrum with $\Delta J = 2$. Our measurements show the direct transition of the *R* branch to be an order of magnitude stronger than the Raman transition of the *S* branch. Direct emission transitions sketched in

Figure 2: Time integrated spectral intensity around 428 nm, compared to the calculated lines based on Eq. (1). The *P* branch starts from $J = 0$; the *R* branch from $J = 1$. (a) the resolved lines are in agreement with *P* and *R* lines shown in red (solid lines) and blue (dashed) respectively. Multiple rotational lines can fit under the *P* branch. The inset shows the spectroscopic vibrational transitions (from upper state ν' to lower state ν'') of photons between various rotational states. (b) The same image is zoomed in and compared with Raman lines of *X* state with $\Delta J = 2$ [*O* branch (right)] and $\Delta J = -2$ [*S* branch (left)].

Fig. 2 (a), comprise a P branch ($\Delta J = -1$), and a R branch ($\Delta J = +1$). The generated 428 nm light undergoes Raman transition during propagation, with the emission of an O-branch ($\Delta J = 2$) and an S-branch ($\Delta J = -2$). The energy levels (in cm^{-1}) are given by:

$$\begin{aligned}
 E = & \omega_e(\nu + \frac{1}{2}) - \chi_e\omega_e(\nu + \frac{1}{2})^2 + Y_e\omega_e(\nu + \frac{1}{2})^3 \\
 & + B_\nu J(J+1) - D_e J^2(J+1)^2 + T_e.
 \end{aligned} \tag{1}$$

The first three terms are vibrational, the fourth and fifth terms are rotational, and the last term is the energy level of the electronic state for $J = 0$ and $\nu = 0$. The subscripts ν and e refer to vibration and electronic states.

The values for selected levels are (in cm^{-1}) $\nu'' = 0$, $T_e = 0$, $\omega_e = 2207.00$ cm^{-1} , $\omega_e\chi_e = 16.10$, $\omega_e Y_e = -0.040$, $B_e = 1.93176$, $D_e = 6.1 \cdot 10^{-6}$ for the X'' state (ion ground state) and $T_e = 25461.4$, $\omega_e = 2419.84$, $\nu' = 1$, $\omega_e\chi_e = 23.18$, $\omega_e Y_e = -0.537$, $B_e = 2.07456$, $D_e = 6.17 \cdot 10^{-6}$ for the B'' state (ion excited state). B_ν is calculated as $B_\nu = B_e - \alpha_e(\nu + \frac{1}{2})$, in which $\alpha_e = 0.01881$ for the X state and $\alpha_e = 0.024$ for the B state [22]. The P branch has a parabolic shape with a band head in the longer wavelengths, as a result there is an overlap between high J of the P branch and low J of the R branch. e.g. $J_R = 7$ overlaps with $J_P = 33$. The Boltzman distribution peaks at J_{max} given by $J_{max} = \sqrt{(k_B T)/2Bhc} - \frac{1}{2}$, in which “ k_B ” is the Boltzman constant, “ T ” the temperature of the medium in Kelvin, “ B ” the rotational constant, “ h ” the Plank constant and “ c ” the speed of light. $J_{max} = 7$ corresponds to $T = 300$ °K for a nitrogen gas in thermal equilibrium. Considering the low temperature of the plasma generated by a 10 Hz filament [23], we believe that the strongest lines observed in Fig. 2 are due to direct transition from B to X states. The Stokes and anti-stokes Raman transition with $\Delta J = \pm 2$ are in the normal ratio. The Raman branch S shows the maximum signal for $J_{max} = 6$ corresponding to room temperature. The parity is symmetric for odd J 's with weight $2/3$, and antisymmetric for even J 's with weight $1/3$. As a result the intensity alternates between odd and even lines, and lines originating from even J 's are expected to be more intense than odd ones, by a factor of two, as is observed.

The forward radiation around 428 nm is compared for linear and circular polarized light of the same energy in Fig. 3, showing a reduction by two orders of magnitude for circular polarization. One might at first interpret this difference as being due to the higher peak field and ionization efficiency in linear polarization. However, the conclusion of Fig. 3 still holds when comparing circular polarization with linear of the same peak field: the R branch is quite resolvable in linear polarization, barely visible in circular. The P branch is significantly reduced in circularly polarized light, even with the same initial peak field. It is also known that the critical power for circularly polarized filaments is higher than the linear due to a weaker Kerr nonlinearity by $2/3$ [24].

Figure 3: Comparison of 428 nm radiation for excitation by linear versus circular polarization. The radiation with circularly polarized excitation is two orders of magnitude smaller than the one for linearly polarized light. In order to quantitatively compare the forward emission for circularly polarized light and linear one, the measurement of Fig 3 is done at the same exact condition and the 428 emission of linearly polarized light is saturating the detector.

The emission from a filament is far beyond a simple uni-molecular radiation and is strongly affected by the spatial profile and focusing geometry. We explore the effect of orbital angular momentum on the forward radiation of filaments. The orbital angular momentum is a spatial property that can affect collectively

a group of atoms or molecules, while individual molecules sense only the local light polarization. An 800 nm beam with angular momentum is prepared by transmission through a spiral phase plate made as to achieve an azimuthal phase retardation of $e^{im\phi}$ across the beam profile ($m = 1$ in our experiment). As the field vanishes at the center of the beam because of the singularity, the beam breaks into two filaments at the same distance from the core [25]. In the results presented here, the 400 nm probe beam has zero orbital angular momentum, and only probes the excitation induced by a 800 nm filament with or without angular momentum. The polarization of the pump and probe are parallel for both cases of linearly and circularly polarized light.

The 428 nm radiation shown using the phase mask (Fig. 4) with linear or circular polarization is seen to ride on an intense background, not present with the Gaussian $m = 0$ beam. One notes also that the 428 nm radiation for the circularly polarized beam, hardly detectable in the case of the beam without orbital angular momentum (cf. Fig. 3), dominates even the radiation produced by the linear polarized beam in the case $m = 1$. The R branch for both circularly and linearly polarized light is visible and accompanied by a strong continuum background. If we take as reference the total integrated signal under the P branch produced by linearly polarized light ($m = 0$), one gets 50% of that amount with circularly polarized-phase masked ($m = 1$) excitation, 40% with linearly polarized-phase-masked ($m = 1$) excitation, and only 1% when excited with circular polarization without mask. The spectrum around 428 nm for linear polarization is maximum at the $J = 11$ line of the R branch; the Raman radiation of the S branch peaks at $J = 7$. While the R branch is clearly between the two ion states B and X , the Raman line can have various neutral and ionic origins. To explore this difference, we perform a study on the rotational population of nitrogen molecules in a filament.

A fraction of the 60 fs pump at 800 nm is frequency doubled to provide a weak (10 μ J) probe. A weak probe (10 μ J) at 400 nm follows the filament produced by the 60 fs pulses at 800 nm. Both beams are focused (colinearly) with lenses of $f = 50$ cm positioned to have their waist at the same location

Figure 4: Forward emission around 428 nm for circularly polarized light with orbital angular momentum $m = 1$. For this case the circular polarization generates stronger signal than linearly polarized light with the same pulse energy of 15mJ.

in the nitrogen cell. The 400 nm probe beam has zero orbital angular momentum and only probes the excitation induced by the 800 nm filament with and without angular momentum. The polarization of the pump and probe are parallel for both cases of linearly and circularly polarized light. The spectrum of the probe is recorded as a function of its delay (scanned with 7 fs steps) with respect to the pump. During the interaction with a strong laser field, nitrogen molecules undergo ionization as well as non-adiabatic alignment. The probe beam undergoes phase upshift and downshift at delays corresponding to molecular alignment and anti-alignment [18, 26]. This phase sweep results in shift of pulse wavelength as a function of delay, as shown in Fig. 5 (a) for linearly polarized light $m = 0$. The Fourier transform of such a measurement shows the J distributions for linear/circular polarization [Fig. 5 (b)] and light with and without angular momentum [Figs. 5 (c and d)]. Because the transition between the rotational lines is proportional to the rotational constant B of the level [Eq. (1)], and the slight difference between the rotational constant of the ion (1.922 cm^{-1}) and neutral (1.998 cm^{-1}) states [22], we can confirm that the Raman modulation of the 400 nm radiation is mainly determined by the neu-

tral molecules. Circularly polarized light sent through the phase mask ($m = 1$), results in a rotational population shifted to higher J states, while for linearly polarized light the effect is less pronounced and in the opposite direction. The linearly polarized light with $m = 0$ is slightly more effective in pumping to higher J states. It can be seen that the population in $J = 14$ is doubled using the phase mask and is comparable to the case of linearly polarized light. The measurement of 428 nm radiation in the forward direction (Figs. 3, 4) shows a stronger signal at $J = 13$, which, considering the Boltzmann distribution of a medium at thermal equilibrium, corresponds to a temperature of 900 Kelvin. It should be noted however that in dealing with molecular alignment with the strong fields of filaments, a thermal equilibrium with a Boltzman distribution may not be present and the alignment itself is affected by the laser intensity.

The data of Fig. 5 clearly indicate that the rotational population of the medium is modified by light polarization and by the phase profile of the orbital angular momentum. This is indeed surprising in view of the non-local nature of the orbital angular momentum of the laser beam. The explanation has therefore to be sought in the formation and propagation properties in the filament itself. The dependence of the critical power on angular momentum m is given by [27]: $P_{cr}^m = [(2^{(2m+1)}\Gamma(m+1)\Gamma(m+2))/(2\Gamma(2m+1))]P_{cr}$, which indicates that a beam with angular momentum $m = 1$ has four times the critical power of a Gaussian beam. Each of the pair of filaments created by the $m = 1$ beam has the same clamping intensity than a filament created by a beam without angular momentum. The intense background in Fig. 4 is a clear evidence of seeding in the case of the beam with angular momentum. The larger seeding in the case of a beam with angular momentum is consistent with the observation that the white light is generated in the formation stage of the filament [28, 29] where a larger distribution of wave vector can satisfy the group velocity or phase matching conditions of either x-waves [30, 11] or four wave mixing. The polarization profile across the filaments with angular momentum [25] and interplay between the filaments can also play a role in four wave mixing and effective seeding at 428 nm. The dynamics of frequency modulation for filaments prepared with

Figure 5: (a) Mean frequency of a 400 nm probe pulse propagating in the path of a 800 nm pump pulse, as function of its delay, for a linearly polarized pump pulse. Fourier transform of such a wavelength shift versus delay, which reveals the rotational population of the neutral nitrogen molecules. b) Linear versus circular c) Linear with and without phase mask and d) circular polarization with and without phase mask.

angular momentum beam is under investigation.

1. Conclusion

Our study shows clearly that the rotational distribution influences the 428 nm radiation. The enhancement in forward emission of 428 nm is always accompanied with population in rotational states higher than the thermal equilibrium. Ionization and non-adiabatic alignment of molecules affect both the rotational

distributions.

Molecules aligned with respect to the laser polarization see a stronger polarizability and more chance for ionization [17]. The rotational distribution is transferred to the ionic state, and the alignment continues in the ion. Since the polarizability in the two ion states of X and B have the opposite signs with respect to the light polarization due to their electronic structure, their alignment at the presence of the laser field is substantially different [31]. The change of polarizability for the ion states can enhance the gain of “air lasing” transitions.

Our measurement suggests that the rotational distribution at “B” state has the maximum of $J = 11$, which is considerably higher than the room temperature $J = 7$ of the S branch of the Raman radiation. This shift can be associated with the ionization process, since the ionization is dominated by radiation in the higher intensity filament core, hence better alignment. Circularly polarized light is less effective in aligning a diatomic molecule (in the plane of polarization [31]), which can be the reason for reduced emission. The stimulated emission in filaments could be enhanced by engineering of the rotational distribution in the medium, either by using pre-aligned samples, or by using a sequence of ultrashort infrared pulses [32].

Although the ionization and radiation are taking place at a molecular level, the stimulated emission known as “air lasing” is strongly dependent on the beam profile and propagation. In the case of a beam prepared with orbital angular momentum, each single molecule sees the same light polarization and is not affected by the spatial phase across the beam cross section. The enhanced radiation at 428 nm for circularly polarized light with orbital angular momentum can be attributed to more effective seeding and overlap with the continuum generated in the beam. It has been shown [33] that the phase retardation of 0.85π would result in a strong increase of spectral bandwidth and blue shift in filamentation.

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