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Population trapping and rotational revival of N_2 molecules during filamentation of a femtosecond laser pulse in air

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Abstract
We study the fluorescence emitted from filaments in air using a pump–probe scheme with a femtosecond Ti–sapphire laser. The fluorescence intensities from the first negative band ($B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$) and the second positive band ($C^3\Pi_u \rightarrow B^3\Pi_g$) show enhancement and change periodically as a function of the pump–probe time delay. We attribute this phenomenon to the universal yet probably forgotten phenomenon of population trapping of nitrogen molecules in highly excited states together with field-induced alignment of nitrogen molecules followed by revivals of the rotational wavepackets. Theoretical calculation of the alignment dynamics of nitrogen molecules is consistent with the experimental data.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Filamentation of strong laser pulses in relatively high pressure gases has been extensively studied [1–6]. As a result it has been established that this phenomenon results from a dynamic interplay between self-focusing and defocusing by the plasma produced by multiphoton/tunnel ionization of air molecules. While qualitatively correct, this mechanism is an oversimplification since it ignores many processes related to laser–molecule interaction that take place inside the filament. Interestingly, one such process, i.e. laser-induced photoemission of neutral and ionic nitrogen molecules [7, 8], was extensively used for the characterization of filaments. This implies the possibility of using the filament behaviour as a tool for probing phenomena in laser–molecule interaction that are difficult to study with other means. The aim of this work is to establish such an approach by studying laser-induced molecular alignment, population trapping and, consequently, photoemission through pump–probe measurements on a filament generated in air.

Molecular alignment has been the subject of intense research for over two decades, fuelled by the interesting underlying physics and by a variety of applications in fields ranging from optics and spectroscopy to solution dynamics and molecular assembly. The vast majority of these investigations focused on the case of nonadiabatic alignment, with the important virtue of leaving the molecule aligned field free after turn off of the laser pulse. The physics, theoretical

and experimental methods and applications of nonadiabatic (or dynamic) alignment are surveyed in [9, 10]. Briefly, the interaction of a moderately intense laser pulse of short duration with respect to the rotational periods with the molecular (permanent or induced) dipole moment gives rise to the excitation of a rotationally broad wavepacket whose coherence properties can be shown to guarantee alignment upon turn off of the pulse. In linear or symmetric top molecules, where the corresponding classical motion is stable, the wavepacket undergoes periodic revivals after the turn off in each of which the initial alignment is precisely reconstructed. Fractional revivals are observed at fractions of the rotational period, depending on the molecular symmetry and spin statistics.

Another interesting although largely forgotten universal phenomenon involved in our study is population trapping. Population trapping, which is alternatively known as interference stabilization [11–15], is a phenomenon that accompanies ionization during the interaction of strong laser fields with atoms and molecules. Population trapping will reduce the ionization rate over an intensity range, an effect that could be observed in the ion versus peak intensity plots in the multiphoton regime of ionization [11, 12] or the high order harmonics versus intensity plot [12]. Observation of population trapping was reported, for instance, in rare gas atoms [11] and CO molecule [12] where it was attributed to the Λ-type interference stabilization. More detailed discussion of the phenomenon can be found in [13–15].

In this paper, population trapping, molecular alignment and rotational revival in air excited by a short Ti–sapphire laser pulse are observed and studied. We measured the fluorescence strength of two photo-emission bands of nitrogen (the first negative band systems ($B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$) ($v = 0 \rightarrow v = 0$) at 391 nm and the second positive band systems ($C^3\Pi_u \rightarrow B^3\Pi_g$) ($v = 0 \rightarrow v = 0$) at 337 nm) versus the delay between the strong fundamental pump and the weak second harmonic probe pulses of the Ti:sapphire laser. The observed enhancement and periodic variation of the signal with the delay time is attributed to field-induced alignment of molecules, rotational revival and population trapping. The experimental results are supported by theoretical simulation of the alignment dynamics of molecular nitrogen.

2. Experimental details

The experimental setup is schematically illustrated in figure 1. A mode-locked femtosecond (fs) Ti–sapphire oscillator (Spectra Physics Tsunami^{TM}) sent seed pulses into a chirped-pulse amplification (CPA) module (Spectra Physics Spitfire^{TM}) for further amplification. The final output beam at around 800 nm has a 1 kHz repetition rate with a maximum energy of 2 mJ/pulse and a beam diameter of about 5 mm ($1/e^2$ level of intensity). The pulse duration was measured using a second-order single-shot autocorrelator (SSA, positive light) to be about 42 fs at full width at half maximum (FWHM). The laser beam was separated into two arms by a 50/50 beam splitter. One arm was used as the pump beam ($\sim$900 μJ/pulse). The other arm was used as the probe. In the probe arm, a second harmonic generation (SHG) crystal

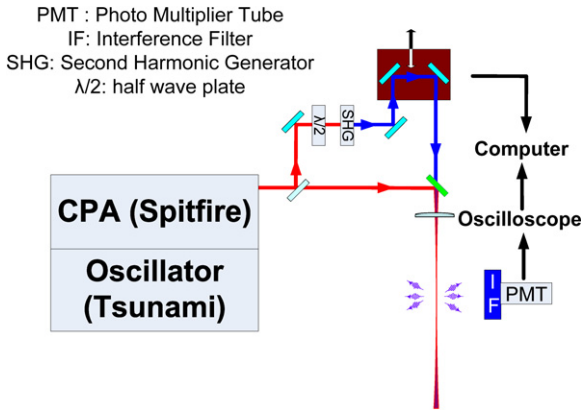

Figure 1. Schematic diagram of the experimental setup, carried out in air.

generated the blue probe pulses at 400 nm with an estimated pulse duration of about 50 fs (FWHM) and pulse energy around 28 μJ. Since in the type I BBO (beta barium borate) crystal, the polarizations of the generated maximum second harmonic pulses are perpendicular to the pump pulse's polarization, we used a half wave plate to rotate the pump polarization by 90° before the SHG crystal. Therefore, the polarization of the pump pulse is parallel to that of the probe pulse. The probe pulse was sent through a high resolution delay line with minimum steps of 0.26 fs (or steps of 80 nm). The observed fluorescence signal was collected versus delay time between the pump and probe pulses. Both the pump and the probe pulses were focused by a plano-convex lens in air.

The experiment was carried out at two different intensities. For this purpose two lenses with different focal lengths were used (f = 100 or 30 cm). The spatial superposition of the two pulses was verified with a far-field measurement while the temporal superposition of these two pulses was checked by the diffraction pattern of the blue pulses from the plasma of the filament of the near infrared pulses. This was used as a sign of the temporal superposition of these two pulses [16].

The fluorescence signal was collected from the side onto a gated micro channel plate photo multiplier tube (MCP-PMT, Hamamatsu R5916U – 52). A pulse generator (Hewlett Packard 8013B) was used to generate a delayed gate signal for triggering the gated MCP-PMT. Moreover, interference filters at 391 nm and 337 nm with 5 nm bandwidth were used for measuring the N_2^+ ($B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$) and N_2 ($C^3\Pi_u \rightarrow B^3\Pi_g$) bands at 391 nm and 337 nm, respectively. The output signal of the MCP-PMT was connected to an oscilloscope (Tektronix, TDS7254 2.5 GHz). The data were averaged over 100 shots.

3. Results and discussion

The delay time dependence of the fluorescence signal of the first negative system of N_2^+ ($B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ transition) (391 nm band) using $f = 100$ cm lens and the second positive system of N_2 ($C^3\Pi_u \rightarrow B^3\Pi_g$ transition) (337 nm band) using $f = 30$ cm lens are presented in figure 2. Positive delay time means that the probe pulse is behind the pump pulse. As can be seen from figure 2, there is a noticeable enhancement at zero time delay,

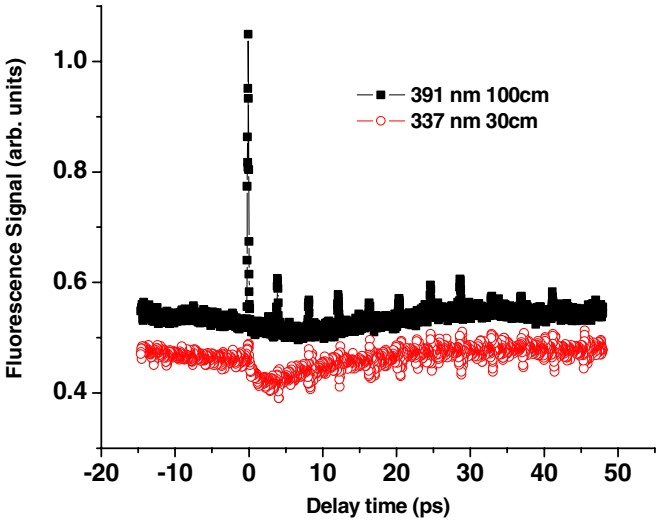

Figure 2. Fluorescence signal of the first negative system of N_2^+ ($B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ transition) at 391 nm and the second positive system of N_2 ($C^3\Pi_u \rightarrow B^3\Pi_g$) at 337 nm versus delay time with corresponding plano-convex lenses whose focal lengths are indicated.

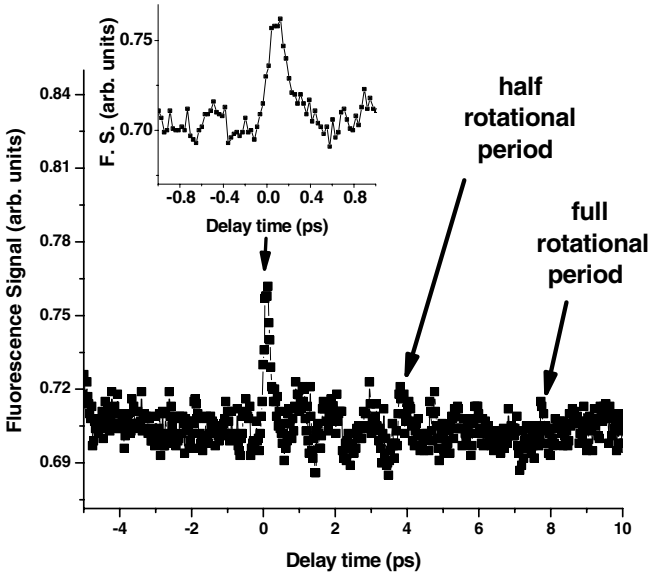

Figure 3. Fluorescence signal of the first negative system of N_2^+ ($B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ transition) at 391 nm versus delay time for 30 cm focal length lens.

which could only be partially attributed to the two-colour ionization enhancement of nitrogen. Two-colour ionization in strong laser fields has been studied in detail [17] and we will not elaborate on this case. When the probe pulses are behind the pump pulses, periodic enhancement is observed in the 391 nm band. The first peak is at a delay of about four picoseconds. This time is half the rotational period of nitrogen molecules. For the 337 nm band, apart from the decay behaviour which was explained elsewhere [18], an oscillatory structure of the fluorescence signal at the revival positions is observed. We shall explain the fluorescence signals observed in figure 2 through field-free alignment and population trapping.

The experiment was repeated with the two lenses reversed, i.e. the 30 cm focal length lens was used for 391 nm band and the 100 cm for 337 nm band (figures 3 and 4). In figure 3, again, a clear enhancement of a fluorescence signal can be seen around zero delay time similar to that in figure 2. However the relative enhancement percentage (defined as ([maximum fluorescence signal – average fluorescence signal in the range of positive delay time]/average fluorescence signal in the range of positive delay time) $*$ 100) is well below the relative enhancement in the 100 cm focal length lens case (compare figure 2), namely 13% in the 30 cm case versus 100% in the 100 cm lens case. For the 337 nm band probed with the 30 cm lens, the enhancement around zero delay time (inset of figure 4) and periodic behaviour are observed but are rather weak. In figures 3 and 4, the enhancement at zero delay time and the periodic revivals can be seen for both band systems as marked by arrows. We conclude that the variation in the fluorescence signal at specific delay times (for instance, at the full and half rotational period of nitrogen molecules) exists regardless of the focal length of the focusing lens. More discussion about the difference in signal strengths between figures 2, 3 and 4 will be given after the physics underlying the interactions is clarified.

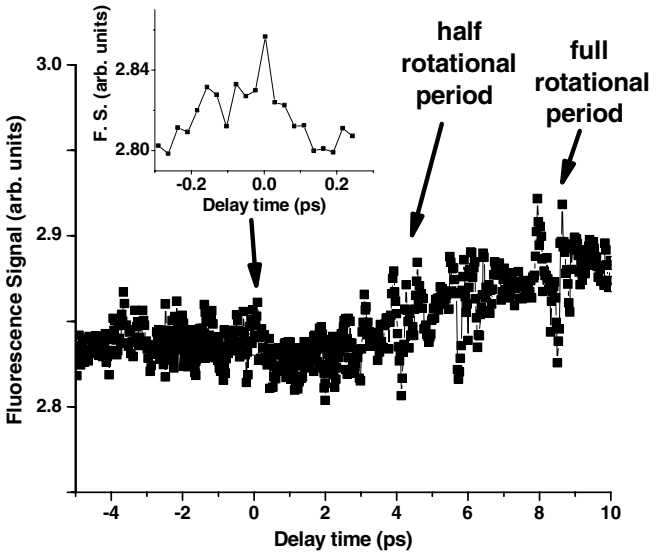

Figure 4. Fluorescence signal of the second positive system of N_2 ($C^3\Pi_u \rightarrow B^3\Pi_g$) at 337 nm versus delay time with 100 cm focal length lens.

The photo-induced alignment dynamics of nitrogen molecules is calculated using a density matrix approach [19]. The expectation value of the alignment observable, namely $\langle\cos^2\theta\rangle$, is obtained as a trace operation $\langle\cos^2\theta\rangle(t) = \mathrm{Tr}\{\hat{\rho}(t)\cos^2\theta\}$, where $\hat{\rho}(t)$ is the rotational density operator that obeys the quantum Liouville equation $\frac{\mathrm{d}\hat{\rho}(t)}{\mathrm{d}t} = \frac{\mathrm{i}}{\hbar}[H_o + H_{\mathrm{int}}(t), \hat{\rho}(t)] + \left(\frac{\mathrm{d}\hat{\rho}(t)}{\mathrm{d}t}\right)_{\mathrm{diss}}$. Here H_o is the rigid rotor Hamiltonian of the nitrogen molecule and H_{int} represents the laser–molecule interaction [19]. The laser pulse is a Gaussian with 50 fs duration and a peak intensity of 5×10^{13} W cm^{-2} which corresponds to the clamped intensity in the filament in air under free propagation of the pump [16]. The commutator represents coherent dynamics and the bracketed term describes

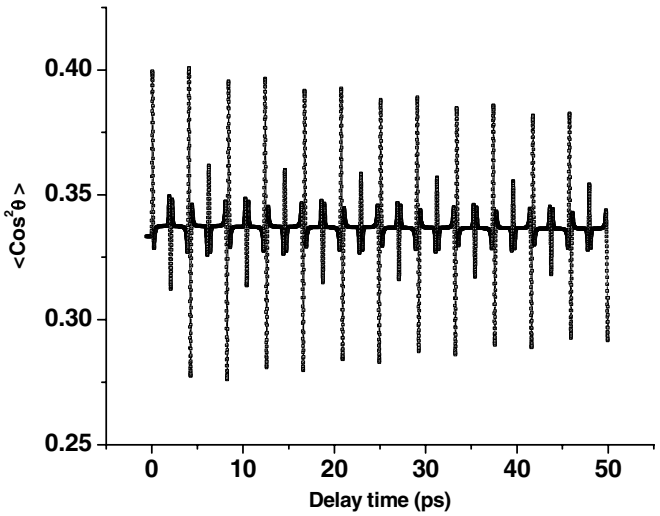

Figure 5. The calculated alignment parameter of nitrogen molecules interacting with 50 fs laser pulses at STP ($T = 295$ K and $P = 760$ torr) at an intensity of 5×10^{13} W cm^{-2}.

the relaxation and decoherence of the density operator due to collisions with nitrogen molecules. The dissipative dynamics is treated using a multilevel Bloch model [19], and the rate expression for population transfer between different rotational states under conditions of STP ($T = 295$ K and $P = 760$ torr) is obtained from the modified exponential gap law (MEG) [20, 21].

Figure 5 shows the plot of the alignment observable as a function of time following the application of a short laser pulse. The alignment revivals can be seen to undergo dissipative dynamics. Consequently, the alignment observable approaches thermal equilibrium as a single exponential decay with a time constant of about 150 ps.

The origin of the fluorescence signals from the 337 nm and the 391 nm bands has been explained in [18] and [22] respectively. Briefly, for the 391 nm band, the strong laser field, through inner valence electron excitation, populates the $B^2\Sigma_u^+$ ionic band, which decays to the $X^2\Sigma_g^+$ ionic ground state band. (In this paper, we measured only the strongest fluorescence signal from the 0–0 band around 391 nm.) The 337 nm band is the transition of the neutral nitrogen's $C^3\Pi_u$ band to $B^3\Pi_g$ band, and the strongest transition amongst all vibrational transitions from these bands is the 0–0 vibrational transition, around 337 nm. The upper band is dominantly populated by dissociative recombination through the process $N_2^+ + N_2 \rightarrow N_4^+$ followed by $N_4^+ + e \rightarrow N_2^* (C^3\Pi_u) + N_2$ at atmospheric pressure [18].

In figure 2, when the probe pulses precede the pump pulses (negative delay time), the fluorescence signals do not show any structure or variation. We also verified that the probe pulses alone do not generate any detectable fluorescence signal. At near zero time delay, enhancement of the fluorescence signals at both wavelengths occurs. At first glance, the enhancement could be explained by two channels of ionization, one by the pump pulse alone and the other by the pump and probe (two-colour scheme). To define the ionization regime, we use the Keldysh parameter. If the Keldysh parameter is larger

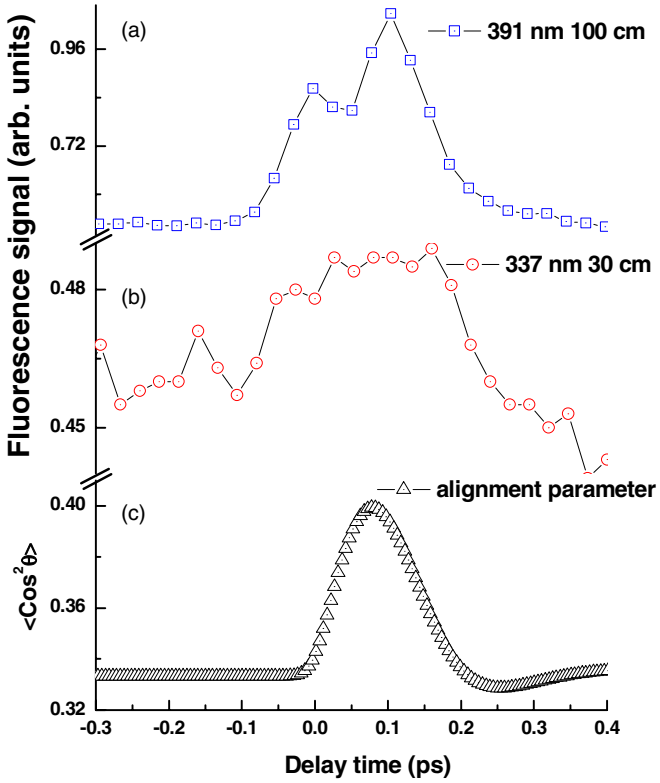

Figure 6. (a) and (b) Observed fluorescence signal of ionized and neutral nitrogen molecules and (c) calculated alignment parameter versus delay time in the vicinity of zero time delay, one of the parameters influencing the fluorescence signal is molecular alignment which is clear from the width of each graph, the other is two-colour ionization.

than 1, multiphoton ionization is the dominant process. Our experiment takes place in the multiphoton regime for the pump pulse with the Keldysh parameter exceeding unity. It is known that in multiphoton processes, a certain number of photons are needed for multiphoton ionization. In the case of near infrared pump pulses (800 nm) each photon has an energy of 1.55 eV. Therefore at least 11 photons are required for the ionization of nitrogen molecules. This is the major ionization channel. When the probe pulse nearly superposes the pump pulse, two-colour ionization also takes place. Two-colour ionization and its phase dependence for the atomic system were studied in [17]. When two pump photons could be replaced by one probe photon, ionization is enhanced. This would in turn result in the enhancement of the fluorescence signal at 391 nm because this fluorescence originates from the excited ion. The width of the enhancement peak near zero delay is twice that of the correlation of the two pulses (see figure 6(a)). However, it contains two peaks; this indicates that other processes also play a role in the enhancement near zero delay time.

The process of two-colour ionization is instantaneous and is thus responsible for the smaller peak around zero delay time (figure 6(a)). The second larger peak in figure 6(a) occurs about 100 fs after the first peak, i.e. after the pump pulse is gone or almost gone. It is the result of the probe pulse's interaction with molecules aligned by the pump pulse. There is a delay of the order of 100 fs between the excitation time at which the molecular rotational wavepacket starts to form

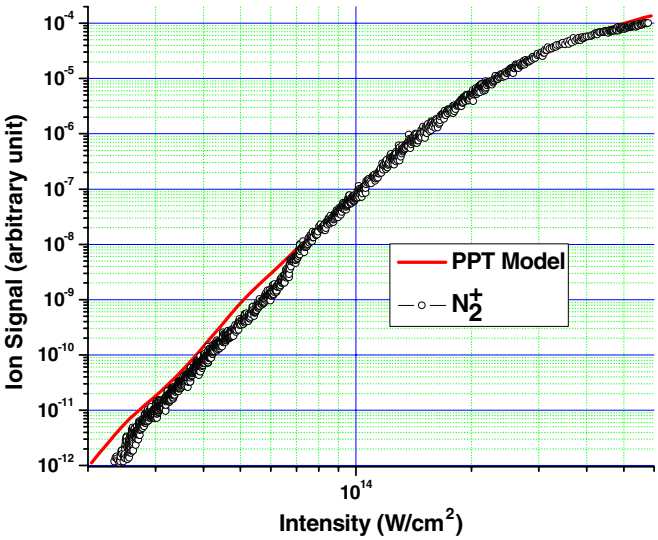

Figure 7. Ion yield versus intensity of the laser field for molecular nitrogen [28, 29].

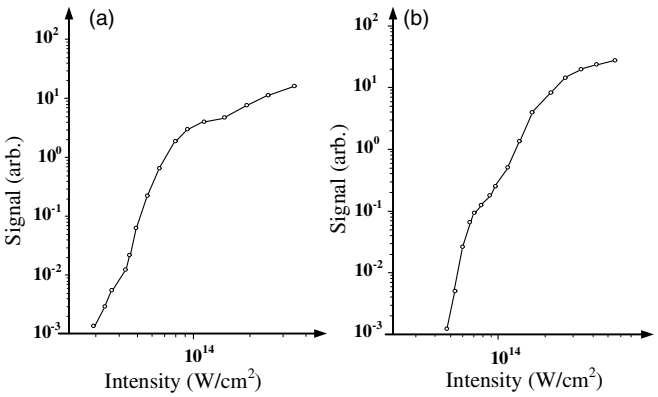

Figure 8. (a) 13th, (b) 15th harmonic signal versus laser intensity for molecular nitrogen.

(field-free alignment process) and the instance of maximum alignment [23]. The calculated alignment parameter in figure 6(c) shows the above-mentioned delay as well.

At the same time, there is population trapping induced by the pump pulse. A wide range of studies, including both experimental and theoretical investigations, was carried out on the phenomena of atomic stabilization. Lowest order perturbation theory (LOPT) predicts that the increase in ion yield versus the laser peak intensity (I) will follows an I^n power law (n being the number of photons participating in ionization). Stabilization is manifested by a decrease in ion yield with increasing intensity of the laser field. Amongst the large variety of publications about atomic stabilization, we note the studies of [11–15]. In atomic systems it was observed that such a decrease in ionization probability with increasing intensity is often due to population trapping in highly excited states, e.g. Rydberg states, which are dynamically brought into resonance with the ground state. Such an effect is referred to as interference stabilization [13, 14]. For rare gas atoms and CO molecules, the reduction in the ionization probability is due to a similar process [11, 12]. Trapping in nitrogen molecules was not reported as yet, but a decrease of the slope of the ion yield versus intensity at around or above 5×10^{13} W cm^{-2} was observed, for example in [24, 25]. For the sake of clarity, these results are reproduced in figure 7. The theoretical curve, based on the Perelomov–Popov–Terentev model [26], shows the expected ionization yield in the absence of population trapping. One should keep in mind that figure 7 is a log–log plot and the difference between the experimental and theoretical curves in the lower intensity zone spanning 2–7 $\times 10^{13}$ W cm^{-2} is significant. Specifically, the decrease in the yield of the 13th and 15th harmonics of nitrogen molecules [25] is clearer evidence of trapping. This is confirmed in figure 8. The dependence of the high harmonics on $N_0{}^2$ (N_0 is the gas density) enhances the signature of trapping [12]. In figure 8, the intensity range where the slopes change is around 3.5–7 $\times 10^{13}$ W cm^{-2}, in agreement with figure 7.

During the interaction, the pump pulse gives rise to population trapping in highly excited states of nitrogen molecules converging to both the ground state and excited ions. This is equivalent to dynamic resonant multiphoton ionization into the continuum coupled by a one photon down transition back to the resonant state (or Λ-type transition). The trapping in the molecular system is complicated and its details are beyond the scope of this paper.

At the same time, nitrogen molecules nonadiabatically align in the strong 800 nm pump laser field, see figure 5. Recently, it was found that nitrogen molecules aligned with respect to the laser polarization are roughly four times more likely to be ionized than nitrogen molecules aligned perpendicularly in the intensity range of 1.5×10^{14} W cm^{-2} [27]. The second largest peak in figure 6(a) could be interpreted as the molecular response to the strong ultrashort laser field when the molecules are first aligned parallel to the pump's linear polarization about 100 fs after the excitation 'kick'. The probe polarization being parallel to that of the pump, there is an enhancement of ionization, hence, fluorescence, due to the probe. In as much as we know, the energy difference between the two ionic states $B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ (391 nm transition) is 3.1 eV. The trapped Rydberg states of N_2 which converge to the ground state of N_2^+ absorb two probe photons (2×3.1 eV) thus populating the excited ionic state $B^2\Sigma_u^+$, which decays by fluorescence to the $X^2\Sigma_g^+$ state. This process is more probable for aligned molecules. If some of the trapped population is in highly excited Rydberg states, a single photon transition, which is more likely than a two photon one, can also affect this dynamics.

We proposed the same interpretation for the second positive band (337 nm band) around zero delay time; i.e. the above-mentioned enhanced ionization would lead to the enhancement of the population in the neutral band $C^3\Pi_u$ through collisions. Their decay by fluorescence into the $B^3\Pi_g$ band results in the fluorescence enhancement observed at 337 nm (figure 6(b)). The duration of the fluorescence enhancement is more than 250 fs, which is the compound of the two processes responsible for the twin peaks of the 391 nm band (figure 6(a), upper curve). The reason why we cannot resolve the two peaks could be that the pump and probe pulses are too long. In addition, calculation of the alignment

of nitrogen molecules with the pump pulse has confirmed that other processes play a role in the vicinity of zero time delay. A comparison between the width of the peaks in figures 6(a)–(c) confirms that effects other than molecular alignment participate in the process. The fact that the width in figure 6(c) is smaller than that in figures 6(a) and (b) is explained as a consequence of two-colour ionization.

At positive delay times, figure 2, periodic enhancement of the 391 nm band is observed. The pump pulse populates trapped highly excited electronic states, including Rydberg states, below the ionic ground states. These states have a long enough lifetime to participate in subsequent processes in the picoseconds time scale. Moreover, field-free periodic molecular alignment (rotational revivals) is created by the pump pulse. This is shown in figure 5 and, in more detail, for time delay around the full revival of nitrogen molecules, in figure 9(c). The first enhancement at positive delay time occurs at the half revival (figures 2, 3). The reason why we do not observe quarter revivals could be technical. The observation of the fluorescence signal is very difficult. In addition it could be in the range of laser energy fluctuation. For the trapped molecules that are aligned parallel to the laser polarization at 4.15 ps after the pump laser pulse, the probe pulse could create more excited ions through two photon absorption processes. This would in turn enhance the 391 nm fluorescence. Figure 9(a) shows the enhancement of the 391 nm line at the full revival, which could be interpreted in the same way as the half revival enhancement. It occurs in the general region of the calculated full revival time (figure 9(c)).

Summarizing the dynamics underlying the 391 nm band, neutral molecules are aligned by the linearly polarized pump pulse at 800 nm and revive periodically. The arrival of the probe pulse, whose linear polarization is parallel to that of the pump, will ionize molecules that are trapped in highly excited states through one or two photon absorption. Absorption by aligned nitrogen molecules is more probable. When the molecules are aligned parallel to the polarization of the laser pulses, at the periodic revival times, ionization is enhanced. This results in periodic fluorescence enhancement of the first negative band at 391 nm. The consequence of this process is that the number of N_2^+ ions is increased, which influences the signal at 337 nm.

The fluorescence signal from the 337 nm band in figure 9(b) shows a clear oscillatory structure. This fluorescence is the result of collision between neutral and ionic nitrogen molecules through the following process: $N_2^+ + N_2 \rightarrow N_4^+$ followed by $N_4^+ + e \rightarrow N_2^* + N_2$. Another process that influences the signal detection is the anisotropy of the fluorescence with respect to the alignment. Considering the nitrogen molecules as radiating dipoles, one expects radiation minimum parallel to the molecular axis and maximum perpendicular to the axis. For randomly oriented molecules, the detected fluorescence signal represents an average. The detection direction is perpendicular to the laser propagation axis and parallel to the pump and probe polarization axis. Hence, the detected fluorescence is minimized when the molecules are aligned and maximized when the alignment direction is perpendicular to the observation axis. The

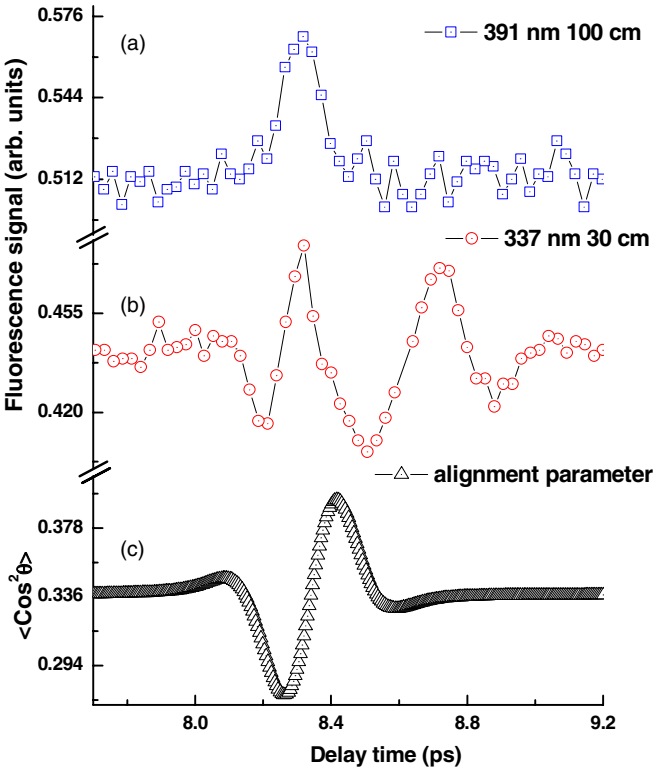

Figure 9. (a) and (b) Observed fluorescence signals of ionized and neutral nitrogen molecules and (c) calculated alignment parameter versus delay time in the vicinity of the full revival period of nitrogen. One of the parameters influencing the fluorescence signal is field-free alignment.

result is the oscillation of the fluorescence signal observed at 337 nm (figure 9(b)), in qualitative agreement with the calculation (figure 9(c)). The shift between the peaks in figures 9(b) and (c) could be due to the experimental error in the absolute calibration of the delay time. Incidentally, this is a very convenient technique to observe the alternation of parallel and perpendicular alignment.

The above-mentioned oscillatory change of the signal is not observed in the signals at 391 nm because the fluorescence originates from molecular ions. At a revival instant, the aligned trapped Rydberg molecules are preferentially ionized by the probe, resulting in the fluorescence enhancement at 391 nm. However, Coulomb repulsion among these initially aligned fluorescing ions would quickly destroy the alignment making the medium isotropic. The detected fluorescence enhancement would thus be isotropic.

The reason why the periodic revivals and enhancement are weaker in the case of the 391 nm fluorescence after we reversed the focusing lenses from 100 cm to 30 cm focal lengths (figures 3 and 4) can now be understood. Tighter focusing leads to a higher clamped intensity and generates a higher plasma density [16]. External focusing increases the clamped intensity inside the filament. The clamped intensity for free propagation is 5×10^{13} W cm^{-2}. Based on the calculation of plasma density in the filament core and the experimental data in figure 7, the clamped intensity of 7×10^{13} W cm^{-2} and 1×10^{14} W cm^{-2} can be attributed to the 100 cm and 30 cm lenses, respectively. Theoretical calculation

was carried out at the clamped intensity of free propagation. At higher intensity the number of ionized molecules is larger than the population of trapped states [11, 12]. For the 391 nm band, the enhancement around the zero delay (figure 6(a)) using the 100 cm lens shows that the first peak (due to two-colour ionization) is weaker than the second peak (due to aligned molecules). This is in contrast to the inset of figure 3 where, using the 30 cm lens, we find that the first peak is stronger than the second (shoulder). The inset of figure 3 indicates that the enhancement due to ionization of the trapped states (second peak or shoulder) is below that of two-colour ionization (first peak). This is because the intensity in this case is higher using the shorter focal length lens and there is now more ionization and fewer trapped states. Because the number of trapped states is lower, enhancement of the 391 nm signal at the revival positions becomes insignificant (see figure 3, positive delay times). But when we use the 100 cm lens, the clamped intensity (hence the plasma density) is lower than that with 30 cm lens. At this lower intensity, more trapped states survive; hence, more enhancement of the fluorescence at the revival positions is observed (figure 9(a)).

In the case of the 337 nm band, the fluorescence strength depends on the plasma density [18]. The higher the plasma density, the higher the population of $C^3\Pi_u$ states will be, hence, the stronger the 337 nm signal is. Thus, the tighter focal length lens (30 cm) yields more ions; as a result, relatively clearer fluorescence enhancement signals could be observed using the 30 cm focal length lens as compared to those observed using the 100 cm lens (compare figure 2 (lower curve) and figure 6(b) with figure 4 (lower curves)).

4. Conclusion

Using a pump–probe technique, we observed the periodic enhancement of nitrogen molecular fluorescence in a filament in air induced by a Ti–sapphire laser pulse. The period corresponds to the periodic revival of the aligned nitrogen molecular wavepacket. The effect of molecular alignment on the fluorescence signal has been confirmed by theoretical calculations. Population trapping of nitrogen molecules in highly excited states seems to be at the origin of the enhancement. We would like to underline, before ending, the importance of the phenomenon of population trapping. Though proposed more than 20 years ago [13] and demonstrated [11, 12, 15] more than 15 years ago, it received little attention in recent years. Our results show that trapping is an important phenomenon accompanying ionization during ultrafast strong laser field interaction with atoms and molecules. This phenomenon also shows up in the field of femtosecond laser filamentation in air, where the enhancement of nitrogen fluorescence by terahertz field is shown to be a new technique to remotely detect terahertz radiation, which otherwise would be absorbed by the humidity in air [28, 29].

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