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Intensity clamping and re-focusing of intense femtosecond laser pulses in nitrogen molecular gas

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ABSTRACT We present results of measurements of fluorescence spectra due to the interaction of a Ti:sapphire laser pulse with N_2 molecules at different gas pressures and pulse energies. The analysis of the data together with the results of numerical simulations, using a propagation model, reveal signatures of the phenomena of intensity clamping and of re-focusing of the laser pulse at high gas pressure. The laser pulse energy for intensity clamping as a function of the gas pressure is determined.

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1 Introduction

There has been in the past few years considerable progress towards an understanding of the propagation of ultra-short intense laser pulses in atomic and molecular gases at high pressure. At high laser intensities, due to multiphoton ionization of the gas a plasma is generated and a balance between the two effects of Kerr self-focusing and of plasma defocusing is usually reached at low plasma densities (e.g. [1]). Due to this balance a self-guided light filament is created in which the pulses have been shown to propagate up to several tens of meters in the laboratories [2–4] and up to 12 km in the atmosphere [5, 6]. Possible applications are lightning control [7] or the detection and identification of atmospheric components using a Lidar [5, 6].

The combined effect of self-focusing and plasma defocusing is known (e.g. [1–4, 8–13]) to affect the spatial and temporal shape of the pulse, leading to phenomena such as intensity clamping [1–4, 8–10, 13] and re-focusing of the laser pulse [4, 11, 12]. Since measurement systems cannot be incorporated into the intense beam directly, precise measurements of the parameters of the pulses in the filaments, such as their temporal and spatial intensity distribution, are difficult. For example, the clamping of the intensity in gases was observed indirectly by measurements of the energy and the diameter of the pulse after the interaction chamber [8] or by reflection of the pulse [2].

An alternative way of determining information about the intensities inside the filament is offered by the observation of quantities outside the interaction volume [12]. Since excitation and ionization of atoms and molecules in intense laser pulses are highly non-linear processes, observables such as ion signals, harmonic generation yields, photoelectron spectra or fluorescence spectra depend on the intensity distribution in the focal volume and, even more crucially, on the peak intensity of the pulse.

The purpose of this study was to investigate, via the fluorescence spectrum of nitrogen molecular gas, the phenomena of intensity clamping and re-focusing. To this end we measured spectra at different gas pressures and input laser energies. The experimental data are analyzed together with results of numerical simulations, using a 3D pulse propagation model.

2 Experimental setup

The Ti:sapphire laser system and the setup of the fluorescence measurement have been described in detail elsewhere [12]. Briefly, pulses at 800 nm, 250 fs (FWHM) with energies of up to 100 mJ are delivered from the laser system. The laser beam (diameter about 2 cm) was focused through a 100 cm lens in an interaction chamber filled with N_2 (background pressure below 5×10^{-2} Torr). A filament around the geometrical focus was visually observed at high gas pressures and was imaged onto the center of the entrance of the spectrometer. In all measurements the fluorescence signal of the whole filament and, hence, of a large focal volume was observed. The spectra were averaged over 100 laser shots and the background was subtracted.

3 Results and discussion

In Fig. 1 the peak intensities of the strongest band heads of two band systems [12, 14], corresponding to the transitions in N_2, $C^3\Pi_u$ ($\nu = 0$) – $B^3\Pi_g$ ($\nu' = 0$) at 337 nm (open squares), and in N_2^+, $B^2\Sigma_u^+$ ($\nu = 0$) – $X^2\Sigma_g^+$ ($\nu' = 0$) at 391 nm (solid circles), are plotted as a function of the input pulse energy. The data are obtained at 0.63 Torr, 400 Torr and atmospheric pressure. While the mechanism by which the $C^3\Pi_u$ electronic state of N_2 is populated is not yet unambiguously determined [12], the excitation of the molecular ion to the vibrational levels of the $B^2\Sigma_u^+$ electronic state of N_2^+ has been identified [12, 14,

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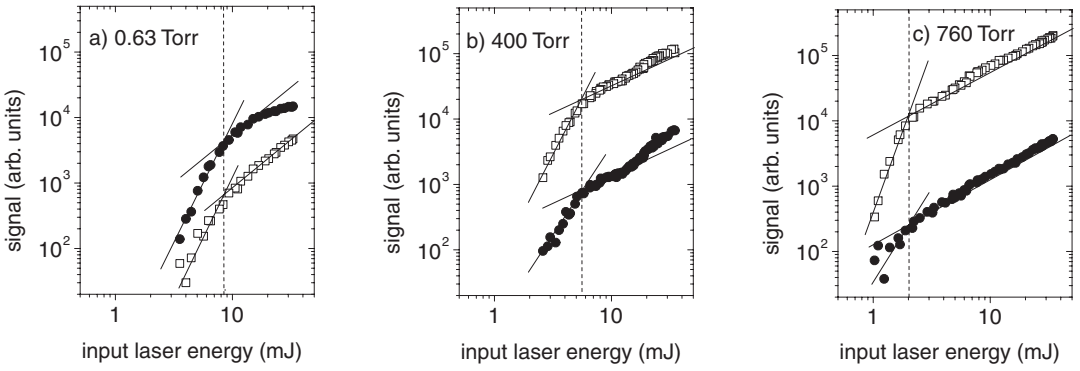

FIGURE 1 Strengths of the strongest band heads of the second positive band system in N_2 (*open squares*) and of the first negative band system in N_2^+ (*solid circles*) as a function of the input laser pulse energy at **a** 0.63 Torr, **b** 400 Torr, and **c** 760 Torr

15] as being due to direct multiphoton ionization of an inner-valence electron of the neutral molecule. As has been observed before [12, 14], the band head of the second positive system in N_2 was found to depend strongly on the gas pressure, while that of the first negative system in N_2^+ is nearly independent of it.

At all pressures, the fluorescence signals show first a quick rise with the increase in the input laser pulse energy before the slope changes towards a slower increase. A characteristic pulse energy for this clearly marked change of the slope is given by the crossing point of two linear fits through the data on the log–log scale, as shown in Fig. 1. At each pressure this change of slope is found at the same input laser energy for both the band heads. This characteristic energy depends on the gas pressure. It is significantly smaller at atmospheric pressure than at low pressure. At 400 Torr we observe a subsequent second rise of the signal at pulse energies above 15 mJ, while at atmospheric pressure we note a small step in both signal curves at about 7 mJ.

In the low-pressure case (Fig. 1a) the line intensity of the strongest band head of the first negative system in N_2^+ (solid circles) as a function of the laser energy has been analyzed before [14, 15]. At the present wavelength the $B^2\Sigma_u^+$ state of the ion is populated by a laser-induced inner-valence electron ionization of the neutral nitrogen molecule. Since the ionization by the field is a highly non-linear process, the ionization probability and, hence, the band head strength crucially depend on the peak intensity of the laser pulse. Thus, the rise of the fluorescence signal below its characteristic change of slope reflects the increase in the peak intensity in the laser focal volume with the increase in the laser pulse energy.

It has been further shown [14, 15] that the change of the slope in the band head strengths of the first negative band system of N_2^+ at low pressure is due to the depletion of neutral molecules in the central part of the focal volume by multiphoton ionization. This effect is known [16] to set in when the peak intensity is high enough for all the initially present neutral molecules near the geometrical focus to be ionized during the interaction with the laser pulse. The depletion limits a further non-linear increase in the probability of inner-valence electron ionization and in the band head strength. The increase in the signal at higher pulse energies, E, is determined by the growth of the Gaussian focal volume. Consequently, at low pressure the slope of the signal is proportional to $E^{3/2}$ beyond the characteristic change of slope [17].

Since the dependence of the strongest band head of the second positive band system of N_2 on the laser energy is similar (Fig. 1a), we may adopt the explanation given above for its steep rise at low pulse energies as a sign for an increase in the peak intensity; in addition, we take the change in its slope to be due to the depletion of neutral molecules in the focal volume, although the mechanism of populating the $C^3\Pi_u$ state in N_2 is not yet unambiguously identified [12]. We note that the interpretation of the fluorescence signals as a function of the input laser energy at 0.63 Torr is consistent with the geometrical focusing of the laser pulse without any deformation, and we may refer to it as the "vacuum case".

Why does the characteristic change in slope of the band head strengths at high pressures (Fig. 1b,c) occur at a significantly lower pulse energy than in the vacuum case (Fig. 1a)? We consider two possible explanations.

First, one could again suspect depletion. This would mean that the depletion of neutral molecules at the central part of the focal volume sets in at a pulse energy lower than that in the vacuum case. However, several groups [6, 18] have measured the electron densities produced by high-power ultra-short laser pulses at atmospheric pressure. The results are in a range from $10^{12}\,cm^{-3}$ to $10^{16}\,cm^{-3}$, corresponding to probabilities of ionization of the order of 10^{-7}–10^{-3}, which are well below the depletion limit (i.e. probability of ionization ≈ 1). Thus, at high pressure the change of slope of the band head strengths cannot occur due to depletion.

The alternative explanation is based on the analysis that the rise of the band head strengths at pulse energies below the characteristic change of slope crucially depends on the increase in the laser peak intensity, as discussed above in the vacuum case. The deviation from this rise, therefore, indicates that the increase in the peak intensity slows down near the characteristic input laser energy. This explanation is also consistent with the observation of the phenomenon of intensity clamping in the interaction of femtosecond laser pulses with gases by other groups [2, 8], which implies that high laser intensities and high electron densities cannot be achieved. We note that the slopes of the band head strengths beyond their first change in slope are found to be proportional to the pulse energy, E (Fig. 1b,c). This indicates that the region of the focal volume, in which the intensity is clamped down to a maximum value, is increasing linearly.

We also observe at high pressures a second change in slope of the signals

at higher laser pulse energies, which occurs weakly at 760 Torr and is clearly marked at 400 Torr. According to our interpretation that the slope of the fluorescence signal reflects the peak intensity in the focal volume, a new intensity distribution, compared to the linear increase in a focal volume with clamped maximum intensity, has to occur with these parameters. Either the volume of maximum clamped intensity increases more quickly than linear or the peak intensity exceeds the maximum clamped value again.

In order to test the above interpretation, we have numerically solved [13] the propagation equation for the envelope function $A(z, \rho, t)$ of the electric field, $E(z, \rho, t) = A(z, \rho, t) \exp(\mathrm{i}kx - \mathrm{i}\omega t) + \mathrm{c.c.}$ The propagation equation is derived from the Maxwell's equations and includes geometrical focusing, diffraction, group-velocity dispersion, self-focusing, and plasma generation via multiphoton ionization. An initially collimated Gaussian beam (width, 250 fs; beam radius, 0.3 cm) is focused by a lens with a focal length $f = 100$ cm. We have computed the peak intensity of the pulse, given as $I_{\mathrm{peak}}(z, \rho) = \max_t \big(I(z, \rho, t)\big)$, where $I(z, \rho, t)$ is defined as the cycle-averaged magnitude of the Poynting vector.

In Figs. 2 and 3 we present results of the numerical calculations for the on-axis peak intensity, i.e. $I_{\mathrm{peak}}(z, \rho = 0)$, for the propagation of the pulse in nitrogen molecular gas at 400 Torr (Fig. 2) and 760 Torr (Fig. 3) as a function of the distance z from the lens at various input laser power, below

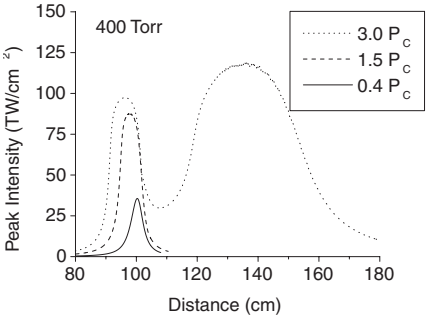

FIGURE 2 Results of numerical simulations for the on-axis peak intensity, $I_{\mathrm{peak}}(z, \rho = 0)$, as a function of the distance z from the lens of focal length $f = 100$ cm. The distributions were obtained for the propagation of laser pulses in nitrogen molecular gas at different input pulse powers below and above the critical power of self-focusing, P_{C}, at a gas pressure of 400 Torr

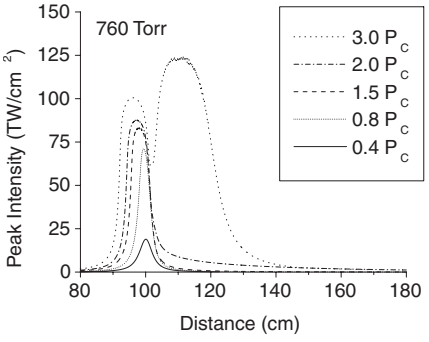

FIGURE 3 Same as Fig. 2, but at atmospheric gas pressure

and above the critical power of self-focusing, P_{C} (see, e.g., [19]). The peak intensity distribution changes its shape from an undisturbed Gaussian distribution around the geometrical focus at $z = 100$ cm at low input power ($0.4 P_{\mathrm{C}}$, solid line) to a broader distribution with the maximum occurring before geometrical focus due to self-focusing at higher input powers. We further note that the increase in the maximum of the on-axis peak intensity reduces for input laser powers near and above P_{C}. This confirms the occurrence of intensity clamping in the pulse at intermediate and atmospheric pressure, as observed in the fluorescence spectra (Fig. 1b,c). Further, the results of the calculations imply a strong relation between the effect of self-focusing and the phenomena of intensity clamping, in agreement with earlier theoretical and experimental studies [1–4, 8–10, 13].

Another feature occurs at the highest input laser power of $3 P_C$ (Figs. 2 and 3, dotted line). A second maximum in the on-axis peak intensity distribution appears at distances beyond the geometrical focus, $z = 100$ cm. This is consistent with the prediction [4, 11, 13] and the observation [4, 12] of a re-focusing of the laser pulse at high laser energies. The peak due to re-focusing is broader than the peak due to self-focusing, and the maximum of the re-focusing peak even slightly exceeds that of the self-focusing peak. Note that the width of the re-focusing peak is considerably larger at 400 Torr (Fig. 2) than at atmospheric pressure (Fig. 3). These results of the numerical calculations are in qualitative agreement with our observation of a second change (increase) in the slope of the band head strengths (Fig. 1b,c). A quantitative comparison between experiment and theory would require additional integrations over the temporal and spatial intensity distributions. However, since the fluorescence signals depend strongly on the peak intensity, the present numerical results on the on-axis peak intensity are sufficient to give a qualitative interpretation of the major changes in the slopes of the signals. Thus, we identify the first change in slope in the observed fluorescence spectra at high gas pressures to be due to intensity clamping and the subsequent second change to be due to re-focusing of the laser pulse.

We finally note that the characteristic first change in slope in the band head strengths offers the opportunity of a precise measurement of the input laser pulse energy responsible for intensity clamping at high gas pressures, given by the crossing point of two linear fits (Fig. 1). We present the laser energy for this change in slope as a function of the inverse pressure in Fig. 4. It can be seen that below 100 Torr the change in slope occurs at a constant pulse energy of about 8.8 mJ. This implies that at these pressures the change is due to the depletion effect, which does not depend on the gas pressure. For high pressures we observe a deviation from this constant value, indicating the presence of intensity clamping. We note that the gradient of the straight line in the high-pressure regime is equal to unity; thus, the characteristic laser energy for intensity clamping is found to depend linearly on the inverse pressure. This result implies again a close relation between the effects of self-focusing

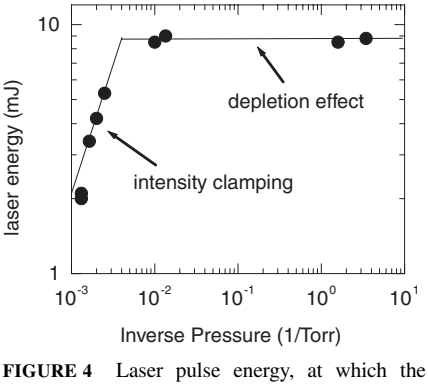

FIGURE 4 Laser pulse energy, at which the characteristic change in slope in the band head strengths occurs, as a function of the inverse gas pressure. Note the linear dependence of the characteristic energy on the inverse pressure at higher pressures, indicating intensity clamping, while at low pressures its value remains constant due to the depletion effect

and intensity clamping, since the critical power for self-focusing is known to depend linearly on the inverse pressure too (e.g. [1]).

4 Conclusion

We have measured the fluorescence spectrum of nitrogen molecular gas interacting with a short focused Ti:sapphire laser pulse at different gas pressures and pulse energies. Characteristic changes in the slope of the signals as a function of the laser energy are identified as being due to intensity clamping and re-focusing of the laser pulse at high pressure. The analysis of the experimental data is found to be in qualitative agreement with results of numerical simulations, obtained by using a pulse propagation model. The pulse energy for intensity clamping as a function of the gas pressure is presented.

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