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Experiments on the Propagation and Filamentation of Ultrashort, Intense Laser Pulses in Air

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Ultrashort (femtosecond), high-power laser pulses can exceed the threshold for nonlinear self-focusing in air resulting in extended propagation distances due to the dynamical balance between the plasma formation and the nonlinear focusing. Experiments were performed using the chirped-pulse-amplification (CPA) lasers at the Naval Research Laboratory to study the physics of self-guiding of the laser beams for extended distances and formation of multiple laser and plasma filaments. Filament dimensions and spectral content of the filaments were measured. An empirical value of the nonlinear index of refraction of air was determined by comparing the experimental profiles of the filaments with the simulation results from a fully self-consistent three-dimensional laser propagation model. The nonlinear index is found to be substantially smaller than the values reported for long (nanosecond) pulses and to agree with recent reported values for femtosecond laser pulses. The measured optical spectra of the "white" light generated in the laser propagation revealed the presence of molecular plasmas useful for chem/bio agent identification.

KEYWORDS: Intense short laser pulses, Laser atmospheric propagation, Laser filamentation

1. Introduction

Ultra-high-power lasers that can deliver intense radiation have traditionally resided in a few very large national laboratories. This is because, as the power of the laser increases, more energy is usually required and thus the size of the laser correspondingly increases. Therefore, the research of the physics associated with the intense radiation from these ultra-high-power lasers could be carried out only at these large institutions. In addition, the size and cost of the lasers severely limited the range of potential applications. This all changed during the 1980s when the chirped-pulse-amplification (CPA) method[9] of generating high-power lasers was demonstrated and popularized around the world. Instead of increasing the energy carried in a laser pulse of a fixed time duration to obtain higher power, one can produce the same laser power if one decreases the pulse duration while maintaining the

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same amount of energy in it. By utilizing ultrashort laser pulses with duration as short as few tens of femtoseconds, laser pulses with power as high as tens of terawatts can now be obtained using tabletop-sized laser systems. Research utilizing these lasers can now be performed in reasonably sized laboratories, and many potential applications are envisioned.

Many interesting phenomena are associated with the interactions of these very intense and short laser pulses with matter. In particular, the propagation of a short intense laser pulse in a gas such as air is very different from that of a long pulse or continuous wave (CW) laser pulse. For example, the high intensity of these pulses can produce nonlinear contributions to the index of refraction of the medium. The intensity of the laser pulse can also become so high that the air molecules ionize and form a plasma. The interplay between the laser pulse, the un-ionized gas, and the plasma can be very complicated and can profoundly affect the evolution of the laser pulse as it propagates through the atmosphere.

Experiments using ultrashort ($\sim$100-fs), high-intensity ($>10^{13}$-W/cm^2) laser pulses have demonstrated long-distance, self-guided atmospheric propagation,[1] air breakdown, filamentation, and white light generation. Intense, directed white light pulses have been generated and backscattered from atmospheric aerosols.[10] The generation of pulsed terahertz radiation in plasma channels formed by femtosecond pulses has also been observed and analyzed.[6,8] Although many of the observations cannot be completely explained, the experimental, theoretical, and numerical results obtained to date indicate potential applications for both passive and active remote sensing and induced electric discharges, among others. In addition, the individual micropulses in a shipboard free-electron laser (FEL) system[3] may exhibit intense, short-pulse propagation characteristics. To achieve these potential applications, it is necessary to have a comprehensive and quantitative understanding of the physical mechanisms that govern the propagation of intense, short laser pulses in air.

2. Propagation of Intense, Short Laser Pulses in Air

The propagation of intense, short laser pulses in the atmosphere involves a variety of linear and nonlinear optical processes. The nonlinear processes are the consequences of the high intensity of the laser pulse. Processes affecting the laser spot size include diffraction, nonlinear self-focusing, ionization, and plasma defocusing. In addition, self-phase modulation, stimulated Raman scattering, and plasma formation also contribute to considerable spectral broadening and white light generation by the laser pulse. On the other hand, the ultrashortness of the laser pulse also needs to be taken into consideration because the physics governing the atmospheric propagation of short, intense laser pulses can be very different from that of long laser pulses. For example, the Raman instability associated with the excitation of molecular rotational modes, which can disrupt the long-distance propagation of long, e.g., nanosecond, pulses may not be as disruptive for laser pulses that are shorter than the characteristic picosecond period of the rotational mode. The implication of this observation is that the nonlinear refractive index n_2 of air could be a function of the laser pulse length.[4,7] A 100-fs pulse could have an effective nonlinear refractive index several times smaller than that of a picosecond pulse. The inherently large spectral bandwidth of a short pulse also renders it more susceptible to dispersion effects in the atmosphere. Finally, the broad spectrum of the short laser pulse could affect the absorption characteristic of the laser in the atmosphere. In conventional narrow-bandwidth, long-pulse lasers that are used in laser radar (LIDAR) applications, the laser line can be positioned in between absorption lines to minimize attenuation in the atmosphere. However, the broad spectrum of a short pulse could be overlapping several individual absorption lines, and this could affect the thermal blooming process, which is a sensitive function of the absorption rate. These effects could be important for proposed shipboard FEL systems.[3]

Perhaps the most prominent phenomenon observed when a high-power laser beam propagates in air is the formation of self-guided laser filaments. When no external focusing is provided, the wave nature of the light emitted from a laser will naturally diffract, and the laser beam will continuously diverge and increase in size. However, the refractive index of air varies with the intensity of the laser in such a way that the higher intensity portion of a laser pulse encounters a higher value of the refractive index. As the refractive index is a measure of the ratio of the speed of light in vacuum to that in the medium under consideration, a higher index of refraction signifies a slower speed of propagation for that portion of the laser pulse, and the laser pulse will converge (focus) onto this lower velocity portion. This is analogous to the propagation of light inside an optical fiber where the core of the fiber has a higher index of refraction. The higher intensity core portion of a laser pulse now also encounters a higher index of refraction, and so it will be guided just like the light traveling down an optical fiber.

The condition for which such self-focusing can occur is governed by the initial laser power in the pulse. When the laser power reaches a threshold value, $P_{\mathrm{NL}} = \lambda_0^2/(2\pi n_0 n_2)$ (Ref. 2), where λ_0 is the laser wavelength and n_0 is the linear refractive index,the nonlinear self-focusing effect can overcome the diffractive divergence of the laser beam, and an ideal laser beam will remain at a constant size forever. For air, the conventionally known value for this critical power is about 3 GW. If the laser power is above this critical value, the laser beam will focus, and theoretically it will continue to decrease in size until a catastrophic collapse is reached.

Fortunately, at high enough intensities, the air will break down and a plasma will be formed. One of the optical properties of plasma is that it has a negative contribution to the index of refraction. Since more plasma is formed where the laser intensity is high, the refractive index is smaller near the core of the laser beam. This is exactly the opposite of the nonlinear contribution to the refractive index before the ionization occurs. The two opposing effects can, in certain circumstances, balance each other and result in a long-lived, noncollapsing filament. More filaments could be formed if the laser power is many times higher than the critical power for self-focusing. These filaments can propagate extended distances, much longer than would be allowed if diffraction effect alone is considered. An example is shown in Fig. 1. It shows the scanned gray-scaled image of the burnt pattern produced on thermally sensitive paper by a 3.56-TW laser pulse from the 1.054-μm-wavelength, 400-fs tabletop terawatt (T^3) laser (described below) at the Naval Research Laboratory (NRL) after propagating for 10 m in the laboratory. Many tens of filaments are clearly visible. The initial laser beam size is 4 cm in diameter, and the individual filaments have diameters of about 200 μm. At this small size, the propagation distance for which the filament diameter will expand by 41.4% due to diffraction (known as the Rayleigh range) is only ~3 cm. The combined effects of nonlinear focusing and plasma formation have kept the filament from diverging for very much longer than was expected. Also, at the small diameter size of these filaments, the laser intensities are in the range of 10^{13}–10^{14} W/cm^2. At such intensities, almost all solid or liquid media will break down and be damaged. The intense field can also generate secondary radiation that can disrupt the operation of many electronic devices. Therefore, these filaments are suitable for applications that involve sensor damage or electronic countermeasure processes.

The initial power of the laser pulse shown in Fig. 1 was about a thousand times more than the critical self-focusing power. From the theory of filamentation instability, the maximum

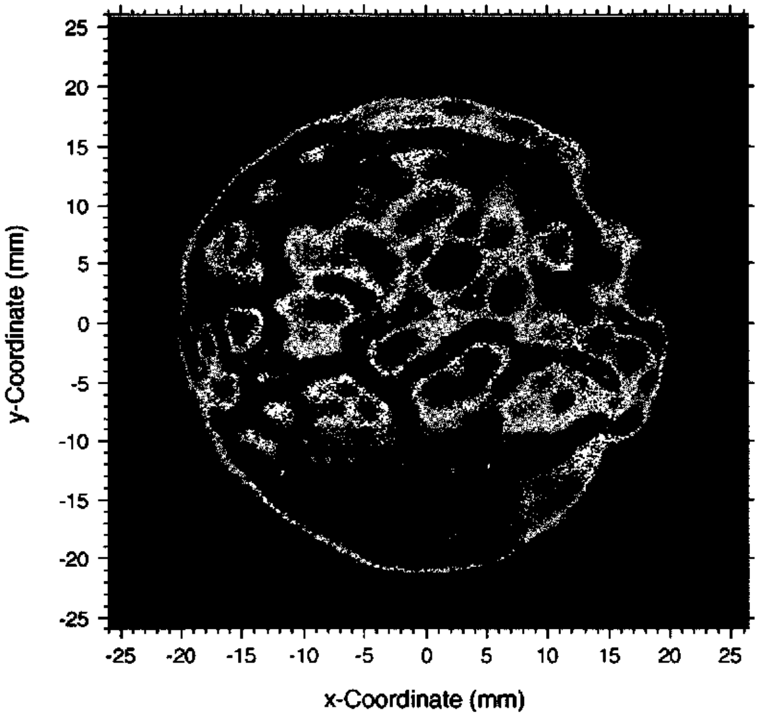

Fig. 1. False color image of self-guided filaments in a 3.56-TW laser pulse produced by the T^3 laser after propagating 10 m in the laboratory. The beam diameter is 4 cm. The pattern of the filament distribution is correlated with the initially nonuniform transverse beam profile.

growth rate occurs when the laser power within the filament is roughly equal to P_{NL} (Ref. 7). This might suggest that many hundreds of filaments could be created under some favorable conditions. The comparatively low number of filaments seen in the experiment demonstrated that the physics of filament formation probably is more involved than simple growth rate considerations. An important consideration among the many complications is the correct evaluation of the critical self-focusing power. Since the critical power was calculated from the nonlinear refractive index of air, one needs to measure the value of the nonlinear index for short, intense laser pulse propagation. A search of the literature reveals that indeed the nonlinear refractive index of air had been measured primarily with optical methods that involved pulses much longer than a picosecond. However, there is strong experimental and theoretical evidence that the standard long pulse value for the nonlinear refractive index for air is not applicable to the self-focusing of femtosecond laser pulses.[4,7] These issues are discussed in more detail in Sec. 4.

An indirect way of obtaining the value of the nonlinear refractive index of air is to compare experimental results of filamentation with numerical results from a theoretical model that includes most of the relevant physics. The NRL air propagation simulation code models atmospheric laser pulse propagation effects with a system of three-dimensional, nonlinear equations that include diffraction, group velocity, and higher order dispersion, stimulated molecular Raman scattering, photoionization, nonlinear bound electron effects, ionization energy depletion, and propagation in a spatially varying atmosphere.[7] The coupled set of equations that was derived for the laser amplitude and electron density is used to analyze a number of physical processes, such as optical/plasma filamentation, pulse compression, nonlinear focusing, and white light generation.

3. Experimental Setup

Two tabletop terawatt CPA laser systems, the T^3 laser and the Ti:sapphire femtosecond laser (TFL), are used for the experiments on air propagation. The T^3 laser employs titanium-doped sapphire (Ti:sapphire) in the laser oscillator and regenerative amplifier and neodymium-doped glass (Nd:glass) in the power amplifiers. The lasing wavelength is in the infrared at 1054 nm. It can generate laser pulses 400 fs long with 5 J of laser energy per pulse at a peak power of >10 TW. For the experiments described here, laser pulses of ~ 1 J of energy per pulse were used at a repetition of 3 min per shot. The NRL TFL is based entirely on Ti:sapphire technology, and its lasing wavelength is at 810 nm. It can be rep-rated at 10 Hz. The laser pulse width can be as short as 50 fs, but for the experiments described here, the laser pulse length used was 100 fs with ~ 30 mJ of energy in each pulse.

The experimental setup is shown Fig. 2. The output of the T^3 laser has an approximately flattop transverse profile with a diameter of 40 mm. For the experiments described here, the T^3 laser was operated at close to full power to maintain stability of the laser parameters. As discussed in Sec. 2 and shown in Fig. 1, many filaments would be formed at such high power. To generate only a single or a few filaments, the energy of the laser pulse was reduced by sending it through an aperture 8.5 mm in diameter. The resulting power of the laser pulse was ~ 100 GW or less and was measured by monitoring the energy of the laser pulse with a calibrated photodiode. A plano–convex lens with a focal length of 12 m was used in some of the experiments to slightly focus the laser beam with a relatively large F-number of $\sim 1{,}200$. Filamentation of the focused or collimated beam (when no lens was used) at a

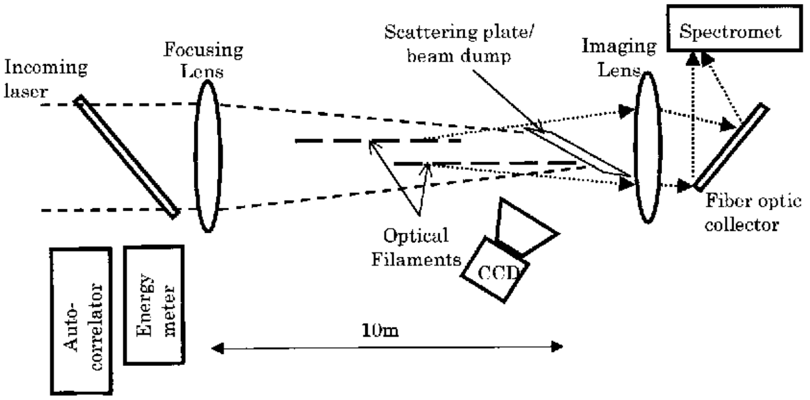

Fig. 2. Experimental setup of the NRL ultrashort, intense laser propagation experiment. Laser pulse length and energy are monitored before the focusing lens. Filament structures are imaged with a CCD camera from a white scattering surface. Broadband radiation is collected with an optical fiber and analyzed with spectrometers. The intense laser filaments are stopped with a laser beam dump.

distance of 10 m from the lens was observed using a white scattering surface oriented at a large grazing incident angle of ~75 deg. The elongated footprint of the laser and its filaments on the scattering surface reduced the laser intensity to below the breakdown threshold of the surface. The laser image was recorded with a charge-coupled device (CCD) camera. Direct observation of the filaments was also obtained by intercepting the laser beam and its filaments with a thermally sensitive paper (burn paper), and the resultant burnt pattern was digitally scanned for data processing. The dynamic range of the burn paper is poor and can serve only as an indicator of the presence of the filaments.

A second set of experiments was carried out using the TFL laser at maximum peak powers of ~300 GW. The TFL laser has a Gaussian transverse beam profile with a diameter of ~8 mm. No focusing lens was used. The filaments were allowed to propagate for 7 m and then collected with a laser beam dump 1 cm in diameter. The broadband radiation generated by the filaments was observed to emit into a cone with a larger divergence than the laser and therefore not intercepted by the laser beam dump. The radiation was imaged with a 40-cm-focal-length lens into an optical fiber. The spectrum was analyzed with a set of three fiber-optic grating spectrometers manufactured by Ocean Optics, Inc. The spectral ranges of the spectrometers are 250–330, 300–440, and 410–520 nm. The spectral resolution of the spectrometers is better than 0.5 nm.

4. Experimental Results

The gray-scaled profile of two filaments formed when an apertured T^3 laser pulse was focused with a 12-m-focal-length lens at a propagation distance of 10 m is shown in Fig. 3. The laser energy in the pulse at the 8.5-mm aperture was 32 mJ, and therefore the initial laser power as determined by its pulse width of 400 fs was 80 GW. When the laser power was varied by changing the energy per pulse, it was found that the threshold power of formation of a single filament was 30 GW. The critical power of nonlinear self-focusing, P_{NL}, can be estimated to be 4.4 GW using the value of $n_2 = 4.2 \times 10^{-19}$ cm^2/W as published in the literature for nanosecond laser pulses at the laser wavelength of 1053 nm (Ref. 5). The apparently large discrepancy between the experimentally determined threshold power for

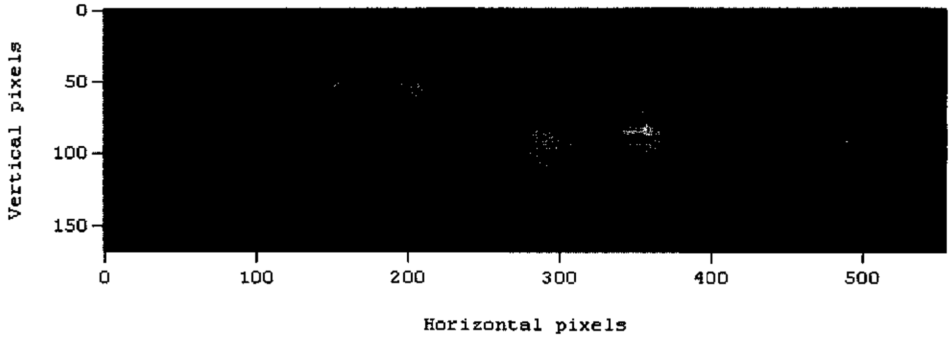

Fig. 3. Image of two filaments formed by a 400-fs laser pulse at a peak power of 80 GW. The image is elongated in the horizontal direction because of the grazing incident angle of 75 deg of the surface. The calibration in the vertical direction is 18 μm to a pixel. The intensity is recorded in a.u.

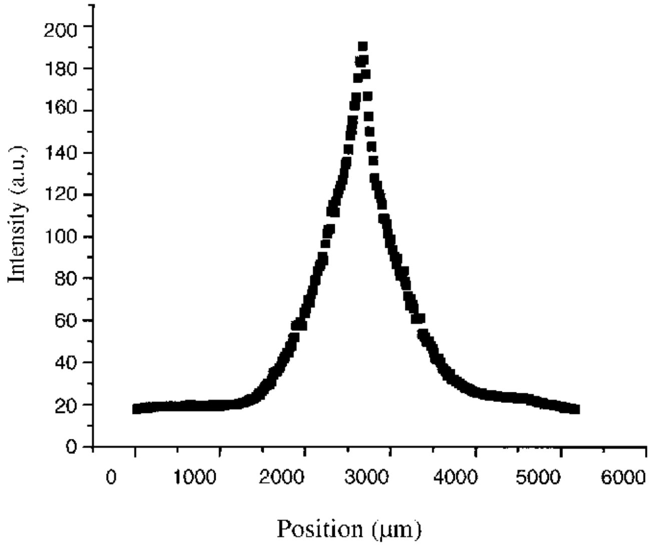

Fig. 4. Vertical lineout of the lower right filament in Fig. 3. Gaussian fit to the central filament gives a half-width of 120 μm.

filament formation and the theoretical critical power can be attributed to several factors. The theoretical critical power could be underestimated as shown below because the nonlinear refractive index of air takes on a substantially different value for femtosecond laser pulses. Second, P_{NL} is the laser power required only to counteract diffraction, and it takes more power to pinch the laser beam down to a filament. Third, the power residing in the filament is only a fraction of the total power in the pulse. Filamentation is an instability developed from the nonuniformity of the initial laser transverse profile. While the entire laser beam is undergoing the so-called "whole beam" self-focusing, since the total power is greater than P_{NL}, perturbations grow into filaments with the highest growth rate if the power in the filament is approximately P_{NL} (Ref. 7). A lineout of the lower right filament in Fig. 3 is shown in Fig. 4. It shows the filament riding on a triangular pedestal [extending to ~120 arbitrary units (a.u.) in height and ~2 mm wide at the base] of laser radiation of the whole beam. A Gaussian fitting of the filament above the pedestal for intensity values of >120 a.u. was performed. The Gaussian half-width of the filament is determined to be ~120 μm. It is apparent that a substantial amount of power can be residing in the pedestal. The actual power of the laser pulse and its filaments after propagating 10 m has not been measured because the filaments can reach an intensity of $>10^{13}$ w/cm^2, which is more than sufficient to damage any filters and power/energy meters.

More quantitative understanding of the propagation physics of ultrashort, intense laser pulses in air can be obtained when experimental results are benchmarked with numerical simulations using realistic models of propagation physics. An experiment was performed using the T^3 laser to generate filaments with a known initial condition that could be simulated with the NRL air propagation code.

The NRL air propagation code[7] solves a set of nonlinear, propagation equations for the laser envelope and the surrounding medium. All of the dominant physical processes required to simulate the experiment are included, e.g., diffraction, group velocity and higher order

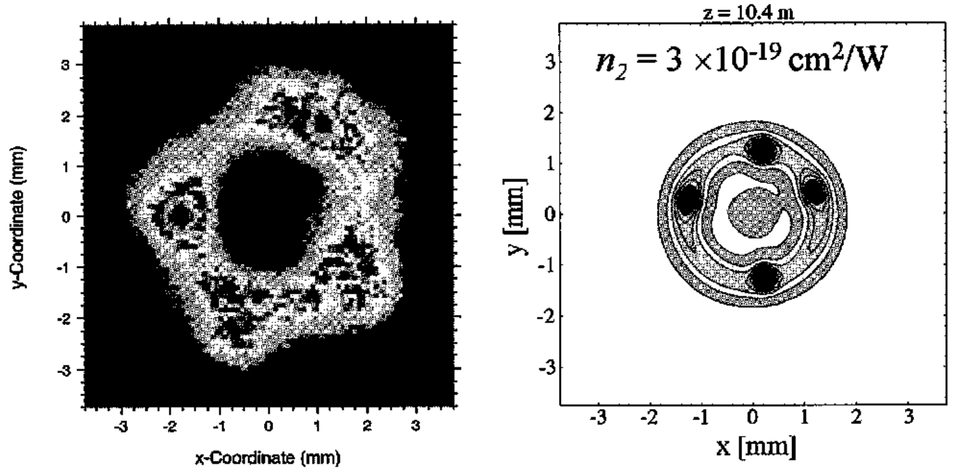

Fig. 5. Formation of filaments from an apertured 400-fs laser beam with peak power of 108 GW at a distance of 10 m. The experimental result is shown on the left, with the simulation result on the right. For the simulation to obtain the best match with the experiment for the same propagation distance and number of filaments, the value of the nonlinear refractive index is found to be 3×10^{-19} cm^2/W, which is 30% less than the conventional value for long pulses.

dispersion, nonlinear self-focusing (optical Kerr effect), and stimulated Raman scattering, which also contributes an effective nonlinear index. For these simulations, the nonlinear index due to the Raman process was taken to be equal to the nonlinear index of the Kerr effect, as is appropriate for ultrashort (~100-fs) pulses.[4]

The circular aperture imposed on the initial laser beam created a well-defined top-hat transverse profile suitable as an input to the simulation code. As the shaped laser beam propagates through the atmosphere in the absence of a focusing lens, normal diffraction reshapes the profile into a donut-shaped form. Nonlinear effects enhance the fluctuations in the intensity around this donut shape, and filaments are formed. At a distance of 10 m, four distinct filaments are formed as shown in the scanned gray-scaled image of burn paper pattern on the left in Fig. 5. The experimental laser parameters of 400-fs pulse length and peak power of 108 GW are imported into the simulation code, and the results are compared to the experiment.

In these simulations, filamentation is induced by seeding the apertured pulse with white noise with a relative amplitude of 3%. The number of filaments observed after propagating ~10 m in air is a function of the chosen value of the nonlinear (Kerr) index n_2. The nonlinear refractive index of air is varied in the simulation runs. It was found that, in order to match the experimental result for the same number of filaments at the same distance, the simulation had to employ a nonlinear refractive index ~30% less than the conventional value. The simulation result is shown in the gray-scaled image on the right in Fig. 5. This value of $n_2 = 3 \times 10^{-19}$ cm^2/W agrees with recent results[4] obtained by measuring the self-phase modulation induced red shift in the radiation spectrum emitted by femtosecond laser pulses propagating in air. The propagation distance in that experiment was relatively short, and no filaments were formed. Our measured value is essentially an effective nonlinear refractive

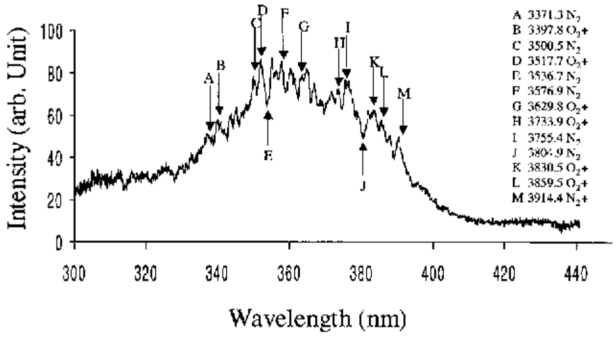

Fig. 6. Broadband radiation spectrum in the UV region from a 100-fs, 300-GW laser pulse propagating for 7 m in air. Spectral line structures are identified to be electronic transitions of neutral and ionic species of the oxygen and nitrogen molecules in air.

index with relevance to the formation of filaments by femtosecond, intense laser pulses propagating long distances in air.

Another interesting phenomenon arising from the propagation of short, intense laser pulses in air is the generation of broadband radiation, often referred to as "white light" or "supercontinuum." This radiation is the consequence of the nonlinear self-phase modulation effect that is caused by the rapid variation in the index of refraction from the front to the back of the laser pulse. Nonlinear generation of optical frequencies outside the original laser linewidth can lead to as much as 100% broadening. As the laser wavelength is in the infrared, the broadened spectrum can extend into the ultraviolet (UV) and far infrared. A portion of the spectrum of the radiation collected after the laser pulse from the 810-nm-wavelength TFL laser has propagated for about 7 m is shown in Fig. 6. It shows that radiation was produced in the UV, and many of the spectral features have been identified with the neutral or ionic species of the oxygen and nitrogen molecules. These features indicate that the molecules in the air where the laser has traversed can be excited, and it offers the potential application of these ultra-short-pulse lasers for identification and detection of chemical and biological molecules from various airborne pollutants or compounds. Substantial spectral broadening is also routinely observed in simulations with the NRL air propagation code.

5. Conclusions

Experimental, theoretical, and numerical studies have been performed on the propagation and interactions of ultrashort, intense laser pulses in air. Filamentation of the laser pulse and the generation of broadband radiation in the UV region were observed. Through the benchmark process between experimental and numerical model calculations, we have gained valuable knowledge of the underlying principles and performed the measurement of the nonlinear refractive index of air for femtosecond, terawatt laser pulses. There are many applications including the propagation physics involved in the shipboard FEL system and short-pulse remote sensing in the standoff detection of airborne pollutants or chemical/biological compounds. Further understanding can be achieved with experimental and theoretical/numerical studies of fundamental physics such as the onset of various nonlinear processes as a function of the laser characteristics of the intense laser pulses.

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