Optics Communications 316 (2014) 61–66

Contents lists available at ScienceDirect

Optics Communications

journal homepage: www.elsevier.com/locate/optcom

Discussion

Filament-induced amplified spontaneous emission in air–hydrocarbons gas mixture

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ARTICLE INFO

Article history:
Received 15 August 2013
Received in revised form
14 October 2013
Accepted 11 November 2013
Available online 28 November 2013

Keywords:
Filament
Amplified spontaneous emission (ASE)
Fluorescence

ABSTRACT

Filament-induced amplified spontaneous emission, ASE, in air–hydrocarbons (~2%) gas mixture, CH_4, C_2H_2, and C_2H_4, was investigated by detecting fluorescence emitted from CH fragments prepared in the electronically excited $A^2\Delta$ state in the filament. The fluorescence signal recorded from the side direction of the filament was linearly proportional to the length of the filament, while the fluorescence signal emitted in the backward direction of the laser propagation increased nonlinearly with the filament length. This difference showing that the filament acted as a gain medium in which the spontaneous emission from CH was amplified (ASE). This process realized by a small amount of hydrocarbon molecular species in air can be applied to remote sensing of pollutants in air.

1. Introduction

Intense femtosecond laser pulse propagation in air shows a unique property which is called filamentation. Filament in air results from a dynamic interplay between self-focusing induced by the neutral air molecules and defocusing from the plasma produced via multiphoton/tunnel ionization of air molecules [1–7]. In recent years, filamentation has been regarded as one of the most attractive phenomena induced by femtosecond laser propagation not only by its complex and characteristic mechanisms leading to intensity clamping [8–10] and lasing action in air [11–15] but also by a variety of its applications such as lightning triggering [16–18], remote sensing [19,20], THz generation [21] and 'rain' making [22] etc.

It was recently demonstrated that many kinds of chemical species in air existing as pollutants can be detected and identified by observing the backward fluorescence from the filament produced in air [5,19,20]. Indeed, through the recent investigations by filament-induced fluorescence spectroscopy, lasing action occurring in the filament [11,12] could enhance the sensitivity of the detection of pollutants in air in atmospheric remote sensing.

The importance of this phenomenon (lasing action in filament) aroused some new work on this subject. The lasing action along the laser propagation direction, that is, in the forward direction, was observed by Yao et al. [13], who generated amplified high-order harmonics in the filaments in air using near-infrared light. On the other hand the lasing action along the direction opposite to the laser propagation direction, that is, along the backward direction, was observed by Dogariu et al. [14], who sent ultraviolet (226 nm) laser pulses through air. Also, high-energy backward lasing (with the energy of up to 300 nJ) in the atmosphere has been implemented by Traverso et al. [23]. Lasing generation in molecular nitrogen in an argon-nitrogen gas mixture in a femtosecond laser filament was observed by Kartashov et al. [24]. They showed that a mid-infrared wavelength filament-assisted nitrogen laser can be at least as efficient as its conventional discharge-pumped counterpart. In a theoretical investigation [25], the backward lasing action of N_2 and O_2 in air through the igniter-heater technique was proposed. Essentially, the intense femtosecond laser pulse (the igniter) creates a lasing-filament in air while liberating some seed electrons. A second, longer pulse (the heater) accelerates the seed electrons and initiates an electron avalanche resulting in more gain. This technique would result in the generation of a strong, coherent, counter-propagating optical pulse which provides a tool for various remote-sensing applications [25].

In the present study, we report the experimental results on the Amplified Spontaneous Emission induced by filament in air–hydrocarbons (~2%) gas mixture, CH_4, C_2H_2, and C_2H_4. By comparing the fluorescence signal of the $A^2\Delta$–$X^2\Pi$ emission band of CH radicals emitted in the direction perpendicular to the laser propagating direction, hereafter called the side direction, and that emitted in the backward direction, it was confirmed that amplification process originated from the electronically excited states of CH was created in the filament in air contaminated with a low percentage of hydrocarbon molecular species. Therefore, the idea of lasing action even in such a low percentage of air pollutant is clearly confirmed.

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0030-4018/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.optcom.2013.11.016

2. Experiment

Using a mode-locked Ti:Sapphire chirped pulse amplification (CPA) system operating at 10 Hz repetition rate, we generated transform limited pulses with a width of 58 fs FWHM (full width at half-maximum), a central wavelength at 800 nm, and a maximum pulse energy of 12 mJ/pulse after a portable compressor. In the two-pass amplifier (inside CPA system), a half-wave plate and a polarizer, installed before the gain medium, were used to change the output energy.

The schematic diagram of the experimental setup used to record the fluorescence emitted in the side direction is shown in Fig. 1 and that used to record the fluorescence emitted in the backward direction is shown in Fig. 2. In both cases, the laser beam was focused by a plano-convex lens (L3, $f=1000$ mm) into a tube filled with a gas mixture, air with 2.0% hydrocarbon molecular species, whose total pressure was 1 atm.

In order to record an image of the filament (to measure the filament's length) as well as fluorescence signal from the side direction, the fluorescence emitted from the whole filament zone was projected through a periscope, two metallic mirrors and two fused silica lenses, onto the entrance slit (2 mm width) of a spectrometer (Acton Research Corporation, Spectra Pro 500i) equipped with an intensified charged-coupled device (ICCD) camera (Princeton Instruments, PIMAX 512) as shown in Fig. 1. For recording the image of the filament, the zero-th order reflection of the grating (1200 groove/mm) was used. In recording a spectrum using the first-order reflection of the grating, the wavelength resolution of 0.3 nm was achieved when the slit width was 100 μm. The ICCD detector was operated in the gating mode that transmitted mostly the molecular fluorescence into the detector so as to minimize the background light.

Since the length of the filament generated by the output beam of the compressor was longer than the opening of the side window, a telescope was used to increase the beam diameter. By this way we reached to the filament lengths shorter than the opening of the side window which made it possible to collect all of fluorescence signal in the side direction. Besides that, in the current experiment for all different mixture of hydrocarbons and air, the results were achieved in single filament regime. Thus experiment was done in a special range of the pump power to prevent the beam from breaking up to several numbers of filaments (multiple filaments regime). Although more amplification of spontaneous emission would have been achieved by the longer filament, these two factors (diameter of the side window and formation of multiple filaments) restricted us to reach to longer filament length.

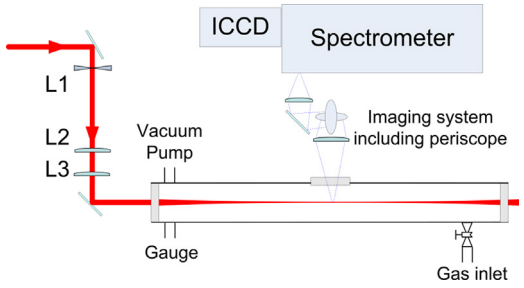

Fig. 1. Experimental setup for recording the image of the laser-induced filament as well as the side fluorescence intensity. The focal lengths of the two plano-convex lenses (L2 and L3) are +1000 mm and the focal length of the plano-concave lens (L1) is −200 or −250 mm.

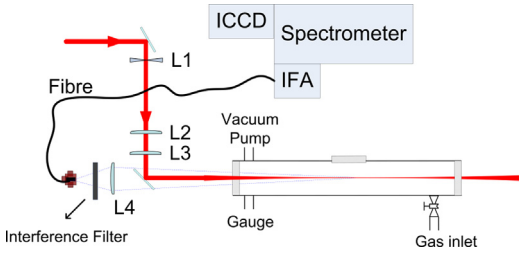

Fig. 2. Experimental setup for recording the backward fluorescence of the laser induced filament. The specifications of the optical components L1–L3 are the same as in Fig. 1. IFA stands for the imaging fiber adaptor.

The backward fluorescence was collected with the fused silica lens (L4 in Fig. 2), whose focal length is f(L4)=200 mm onto the entrance end of a 3 m long fibre bundle (Princeton Instruments, Model LG-455-020), and was guided to the entrance slit of the spectrometer through an Imaging Fiber Adaptor (IFA in Fig. 2). In addition, an interference filter that transmits 431 nm light with the spectral bandwidth of 2.5 nm (FWHM) is placed in front of the fiber head to avoid introducing the strong white light into the fiber.

The sample gas tube was first evacuated to a background pressure of 4×10^{-2} Torr, and then, the air (98%) and hydrocarbon (2%) were mixed in the gas tube so that the total pressure became 760 Torr. The concentration of 2% was chosen because the lower flammable limits of the CH_4, C_2H_2, and C_2H_4 in air are 4.4%, 2.5% and 2.7%, respectively [26].

3. Results and discussion

In Fig. 3, we show the spectrum of the filament-induced fluorescence of air (black) and that of air–acetylene (2%) gas mixture (red) which was recorded from the side. The fluorescence spectra for the mixture of air with ethylene and air with methane were found to be almost the same as the one for the mixture of air with acetylene. The fluorescence signal appearing at around 431 nm corresponds to the $A^2\Delta$–$X^2\Pi$ (0,0) transition of CH fragments [27]. The origin of each fluorescence signal was the subject of many investigations. The fluorescence mechanism of hydrocarbons and nitrogen in strong laser field were discussed in [28–30]. The fluorescence mechanism in hydrocarbon was described as a neutral dissociation through superexcited states [28]. First, highly neutral excited states above the ionization potential, superexcited states, were populated through multiphoton excitation. Then the superexcited states could decay into different channels.

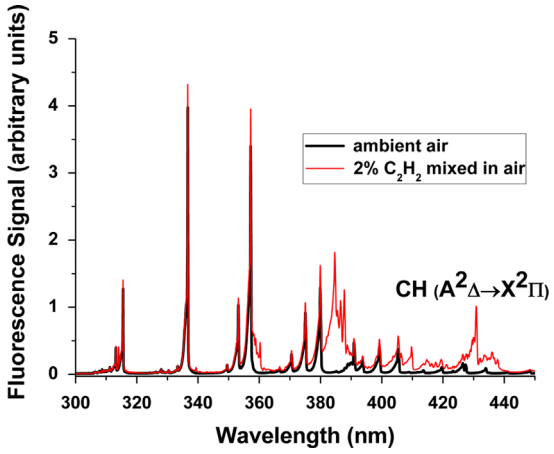

Fig. 3. The fluorescence emission spectrum from the laser-induced filament in ambient air (black) and that from the laser-induced filament in 2% acetylene in air (red). The $A^2\Delta$–$X^2\Pi$ (0,0) transition of CH fragments can be seen at around 431 nm. More precise spectroscopic assignments of the emission spectrum in the wavelength region above can be found in Refs. [28,29]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)