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Abstract: We report on the generation of bidirectional 656 nm lasing of atomic hydrogen in a flame using 205-nm femtosecond laser pulse. The forward lasing strength is one order of magnitude stronger than the backward one.

1. Introduction

In recent years "Air Lasing", which refers to the generation of laser-like coherent emission, has attracted intense research activities (refer to [1] and references therein). In particular, its potentially revolutionizing impact on the remote-sensing field is a huge driving force for the pursuit of backward-propagating lasing concepts. As far as we know, backward lasing can mainly be achieved through two different approaches: 1) multiphoton pumping of air constituents [2-4]; 2) pumping via electron-molecule inelastic collision in femtosecond laser filamentation [5-8].

In the first scenario, the backward-propagating lasing effect has been observed in various atoms and molecules such as O, H, C, N, CO and NH_3 with nanosecond/picosecond 2- or 3-photon excitation in flames, where these species are naturally present (Chapter 1, reference [1]). In ambient air, the lasing effect of O, N, H, Ar atoms, and water molecules has recently been generated via multiphoton resonant excitation (Chapter 2, reference [1]). For N and O lasing, the signal strength can be significantly enhanced through pre-dissociation of N_2 and O_2 using another more powerful laser pulse. Interestingly, it was observed that the forward lasing strength is comparable with, or in some cases even stronger, than the backward lasing strength. The physical mechanism responsible for this phenomenon has not yet been fully settled. There are four mechanisms that can be operative: amplified spontaneous emission (ASE), which is about equally effective in both the forward and backward direction, hyper-Raman gain, which is also equally effective in both directions, four-wave mixing, which only gives rise to emission in the forward direction, and coherence-brightened superfluorescence (SF). These different mechanisms can dominate under different experimental conditions or play a role in a cooperative manner.

Recently, we obtained bidirectional 656 nm lasing from hydrogen atoms in a flame through 2-photon pumping with 205-nm femtosecond laser pulses [9]. In the flame, the hydrogen atoms are naturally present. Since hydrogen atoms are naturally present in the flame, no preceding photodissocation is needed, which is an advantage compared to O and N lasing in air lasing. Here, we report recent experimental results on hydrogen lasing in a flame. They show that the forward lasing is much stronger than the backward one.

2. Experimental description

In the experiment, the femtosecond laser system we used provides 205 nm laser pulses with duration of ~125 fs (FWHM), a maximum pulse energy of ~60 μJ at 10 Hz repetition rate. The beam diameter is about 5 mm. A simplified illustration of the experimental setup is shown in Fig. 1. Firstly, the 205 nm laser beam propagates through a bulk CaF_2 equilateral dispersive prism in order not only to get rid of emission at other wavelengths, but also to spatially separate the backward-propagating 656 nm lasing emission. After the prism, the 205 nm laser pulse with a maximum energy of ~25 μJ was focused by an $f = 40\ cm$ spherical lens into a CH_4/O_2 welding flame (equivalence ratio of ~1.0), where H atoms naturally are present. The nozzle diameter of the welding torch is 1.5 mm. The focus spot has a dimension of ~600 μm (horizontal) × ~100 μm (vertical).

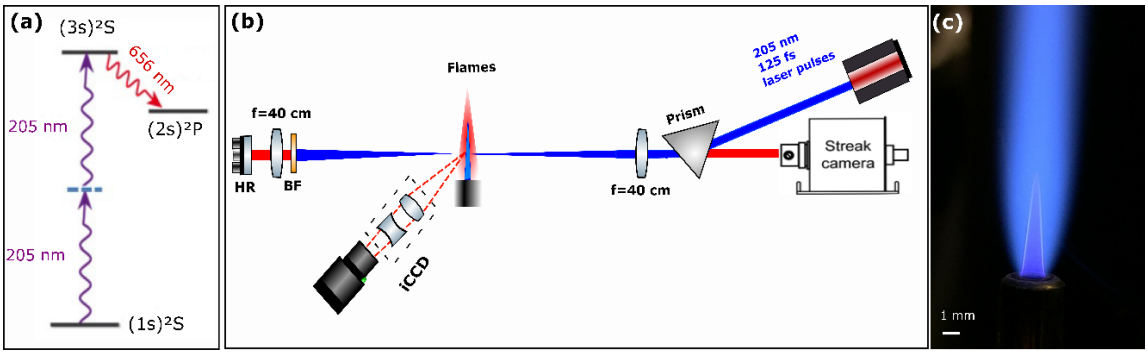

Fig. 1. (a) Energy level diagram relevant to 656 nm hydrogen lasing. (b) Schematic illustration of the experimental setup for simultaneously measuring the forward and backward 656 nm hydrogen lasing. (c) Chemiluminescence image of the CH_4/O_2 welding flame.

The 656 nm lasing emission is generated in both forward and backward direction. In the backward direction, a streak camera was used to measure the lasing signal. The forward lasing emission was collected in a symmetrical configuration and reflected back into the streak camera. In this way, the forward and backward lasing signals can be simultaneously measured. In the side direction, an intensified CCD camera was positioned to measure the spatial distribution of hydrogen atoms through capturing the laser-induced 656 nm fluorescence.

3. Results and Discussion

The first experiment was conducted to measure the spatial profiles by using the "focus mode" of the streak camera, in which case it basically works as a conventional CCD camera. The results are shown in Fig. 2 (a)-(b). The divergence of the forward lasing beam is few times larger than that of the backward lasing beam. Also, the intensity of the forward lasing pulse is apparently much stronger than the backward one. We integrated the signal within a rectangular region on the camera chip (same area for both cases) and plotted the correlation between the forward 656 nm lasing signal and the backward one, as shown in Fig. 2(c). A linear fitting to the data suggests a slope of ~10.

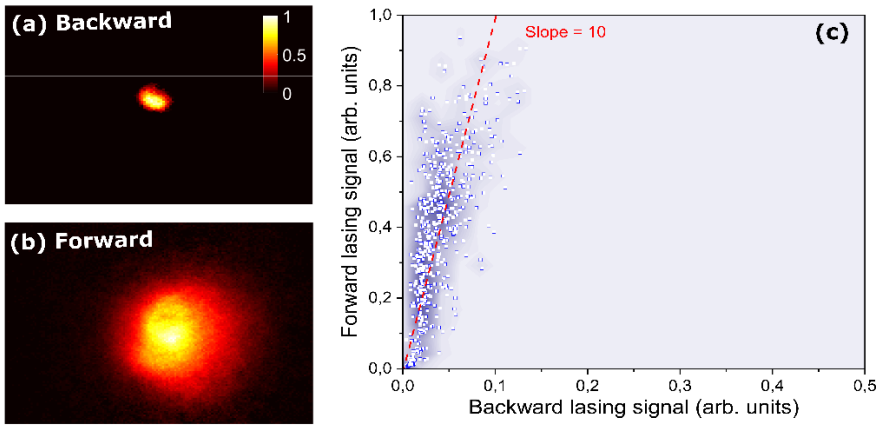

Fig. 2. (a)-(b) Spatial profiles of the backward and forward lasing beam captured by the streak camera operating in focus mode (i.e. no streak applied). (c) Scatter plot of the correlation between the forward and backward 656 nm lasing signals (normalized), where the blue dots represent data recorded for 660 shots. Linear fitting to the data gives rise to a slope of ~10.

A similar observation, i.e. that the forward lasing is more divergent and much stronger than the backward one, was also made in experiments with 337 nm lasing of neutral nitrogen molecules. For the 337-nm lasing, the pumping process is enabled via electron-molecule inelastic collisions through femtosecond laser filamentation, which is not resonant [8]. It was observed that the forward 337 nm lasing strength is about 3 orders of magnitude stronger than the backward one. This difference was attributed to the traveling excitation nature of the pumping process, given that the effective gain lifetime (~15 ps) is much shorter than the propagation time over a 3-cm-long excitation volume. In our experiments, the effective gain length was approximately 2 mm, which is 20 times larger than the transverse size. Thus, the gain medium can be regarded as being pencil-shaped, and we suspect that traveling wave excitation might play an essential role for generating a forward 656 nm lasing signal that is one order of magnitude stronger than the backward lasing signal. We also notice that the backward 845 nm oxygen lasing signal, pumped by 50 fs 226 nm laser pulses after predissociation, was reported to be about an order of magnitude stronger than the forward one [10], i.e. opposite to our case. It was interpreted as being due to destructive interference between hyper-Raman gain and four-wave-mixing. In order to fully understand the physical mechanism of femtosecond laser-pumped atomic lasing, both further experiments and numerical simulations are needed.

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