

Travelling-Wave Amplified Spontaneous Emission Excited in a Prismatic Geometry

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Abstract. A new experimental realisation of the travelling-wave amplified spontaneous emission (TWASE) is described utilising a prismatic arrangement to get the pulse front delay in the pump beam. Rhodamine dye solutions were pumped by the amplified 25 ps long 555 nm pulses of a distributed feedback dye laser. The pulse shortening in the TWASE resulted in 12 ps output pulses with 18% energy efficiency. The observed spectrum showed numerous lines, and it contained the amplified Raman line of the pump beam. Several spots were found in the far-field zone of the generated TWASE.

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Different properties of the travelling-wave amplified spontaneous emission (TWASE) in dyes were reported in the last few years [1-4]. Here the picosecond pump pulse is running synchronously with the generated amplified spontaneous emission along the active medium, which length is more or less greater than the spatial length of the pump pulse. This method seems to be effective in pulse shortening [3] and generating ps TWASE in dyes with very low quantum yield [4]. It can be applicable in constructing tunable picosecond sources [5]. In every experiment performed at this time the pulse front delay in the cross section of the pump beam was produced by a diffraction grating [1-5]. A new experimental arrangement was proposed by Bor and Rácz [6], where a prism was attached to the dye cell, as it is shown in Fig. 1. The pulse front delay in the pump beam is derived from two combined effects: (i) the different parts of the pump beam have different path length in the prism, (ii) the group velocity dispersion of the prism causes a further delay as written in detail in [6]. Synchronization can easily be achieved by changing the angle of the incident beam. and its condition is also given in $\lceil 6 \rceil$. The aim of the present paper is to demonstrate the operation of this arrangement and to describe the spatial and spectral properties of the generated TWASE.

Experimental

Figure 1 shows the experimental arrangement of the generation of the TWASE. A XeCl excimer laser pumped picosecond dye laser system operating with 1 pps repetition rate was used as pump source. The oscillator of the dye laser system was a distributed feedback dye laser with active material of 1.5×10^{-2} M



Fig. 1. Experimental setup using prismatic arrangement for generation of the TWASE pulses

Coumarin 153 solution mixed with 1.5×10^{-4} M Rhodamine B as saturable absorber to have shorter and more stable pulses [7]. The oscillator output was amplified by a three stage amplifier. The pulse length of the amplified output was 25 ps at 555 nm with 72 µJ energy. It was polarised horizontally.

A BK 7 glass right angle prism was matched by glycerol to a 4 cm long dye cell. The incident angle of the pump beam was calculated to be 43.8° and 49.2° in the case of ethanol and methamol as solvent, respectively. Applying the Fresnel formulas the overall transmission was 73% and 76% for these solvents, respectively. This is an important advantage in contrast to the 30–60% efficiency of the grating delay element used in the previous experiments [1–5].

The pump beam was focussed onto the active medium to form a 28 mm long line by an f = 150 mm cylindrical lens. The active materials were 2×10^{-3} M Rhodamine 101, 6.3×10^{-4} M Rhodamine B, and 6.3×10^{-3} M Rhodamine 6G in ethanol. Using these concentrations the estimated absorption length was 67 µm but the real penetration depth was much lower (about 67 µm * cos82° = 9 µm), because the refraction angle inside the dye solution was about 82°. The height of the excited volume was about 120 µm.

The spectral and temporal behaviour of TWASE pulses were analysed by an SzTE-1 spectrograph (0.2 A spectral resolution) and by a Hamamatsu C979 streak camera (10 ps temporal resolution), respectively. The energy of the pulses was measured by a thermopile.

Results and Discussion

At first we can say that the results concerning to the three types of Rhodamin dyes were very similar so the results shown here are valid all of them.



Fig. 2. Dependence of the energy of the TWASE pulses on the pump energy

Using Rhodamine B solution as active material the TWASE energy is 18% of the pump one. The forwardbackward TWASE energy ratio is 64, showing the effect of the travelling-wave excitation scheme. Figure 2 shows the TWASE energy as a function of the pump energy. Above the pump energy of 15 μ J a linear relationship is found between the two quantities, which means that the gain is saturated. This curve is in accordance to the theoretical prediction of [8].

The results of the spectral measurements are similar to [1]. There are great number of spectral lines which change shot to shot, but at higher pump intensity these lines are less definite. In our experiment two broad spectral bands are obtained, centers of which move to the longer wavelength direction increasing the pump intensity. These spectral bands belong to two pulses in space as well and they can be observed in the far field as two spots (Fig. 3a). The TWASE spectrum of Rhodamine 6G is shown in the upper and lower part of Fig. 4, which was obtained by illuminating the slit of the spectrograph by one and the



Fig. 3a, b. The far-field pattern of the TWASE light of Rhodamine B a and Rhodamin 6G b, respectively

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Fig. 4. The TWASE spectrum of Rhodamine 6G. (The line of the Raman scattered light is indicated by the arrow. The two heavy lines are the Na doublet used for calibration)

other part of the far field pattern, respectively, while the photographic plate was moved vertically between the two recording.

The time duration of the forward TWASE is shorter than that of the pump pulse, it is measured to be 12 ps in accordance with [8]. The backward TWASE pulsewidth is found to be 75 ps. It is noted, that the pulses that can be separated in the far field and that belong to the two spectral bands do not come at the very same time. 4 ps time difference can be measured between them by delaying one of them using a quartz cube of 31 mm. Furthermore in some experiments the two spots devide into more parts as it is shown in Fig. 3b.

These spots in the far field were not observed in the previous travelling-wave experiments. Rubinov et al. presented similar patterns examining ultrashort fluorescence and they explained it by a selffocussing phenomena in [9]. The effects found here are not fully understood.

There is a definite line of 574 nm in the TWASE spectrum of dye Rhodamine 6G, which can be seen in Fig. 4 as well. This line does not change when metanol is used instead of ethanol as solvent, but it changes in the case of shifting the pump wavelength. The wavenumber difference between this line and that of the pump is measured to be 596 cm^{-1} . Carreira et al. published a Raman line of about 600 cm^{-1} of Rhodamine 6G in [10], so we explain the obtained spectral line as the Stokes Raman scattered light of the pump beam on the dye molecules. This scattered light is greatly amplified as it is in the band of the gain, so it is very easy to detect. This narrow band radiation can also be separated in space in the far field (see semi-circle in Fig. 3b). Similarly conical Stokes emission pattern was studied in H_2 by Yuan et al. [11].

Conclusions

(i) The effectiveness of the prismatic travelling-wave arrangement is demonstrated.

(ii) The main characteristics of the experimental observations of the TWASE are in agreement to the previous experimental and theoretical results.

(iii) Spectrally, temporally and spatially different beams are found in the TWASE pulses. These result may be due to the special experimental arrangement and their explanation needs further investigations.

(iv) Raman scattered line of the pump light on the Rhodamine 6G molecules is observed.

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