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Picosecond Pulse Shortening by Travelling Wave Amplified Spontaneous Emission

Zs. Bor*, S. Szatmári*, and Alexander Müller

Max-Planck-Institut für biophysikalische Chemie, Abteilung Laserphysik, D-3400 Göttingen, Fed. Rep. Germany

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Abstract. A new scheme for travelling-wave excitation of amplified spontaneous emission (ASE) employing transversal pumping is presented. ASE pulses emitted in the forward direction had a duration of 6 ps, corresponding to one half of the pump pulse duration. The spectrum was strongly structured, with individual components having a width of 0.1–1.0 Å. Essential characteristics of longitudinal and transversal excitation of travelling wave ASE are compared.

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Dye solutions even if excited only by moderate pump energies can have very high optical gain. In such case the spontaneous emission is amplified as it propagates along the pencil-shaped excited volume and forms a low divergence beam which is called *amplified spontaneous emission* (ASE). ASE has a significant effect on the operation of dye lasers and picosecond pulse amplifiers which has been treated in [1–3].

Studies of ASE excited by picosecond pulses have been carried out also [4–13]. In those works emphasis was on shortening of the ASE pulses with respect to the fluorescence lifetime, determination of various molecular absorption and relaxation parameters and the possibility of tunable ultrashort pulse generation by travelling wave ASE.

With picosecond-pulse excitation the duration of the ASE pulse was typically 3–10 times the duration of the pump pulse, which was typically 3 orders of magnitude shorter than the fluorescence lifetime of the dye molecules. Recently, Rubinov observed ASE pulses as short as the duration of the pump pulse, when self-focussing of the pump pulse occurred. However, the statistical filamentations and the intensity of the ASE being distributed in hot spots [13], are serious obstacles to practical applications.

In the present paper we show that ASE pulses of 6.1 ps duration can be generated by pumping with 12 ps pulses in a travelling wave arrangement. To our knowledge, it is the first demonstration that pulse shortening can be achieved by the use of a more favorable pumping scheme than that which was used before. We believe that this technique is a general method for picosecond-pulse shortening.

* *Permanent address:* JATE University, Dept. of Experimental Physics, H-6720 Szeged, Dóm tér 9, Hungary

Experimental Arrangement

Figure 1 shows the experimental setup, constituting a transversal pumping arrangement and replacing the

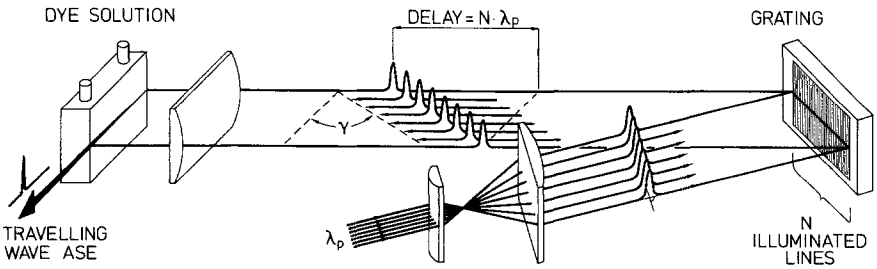

Fig. 1. Arrangement for transversal excitation of travelling wave ASE

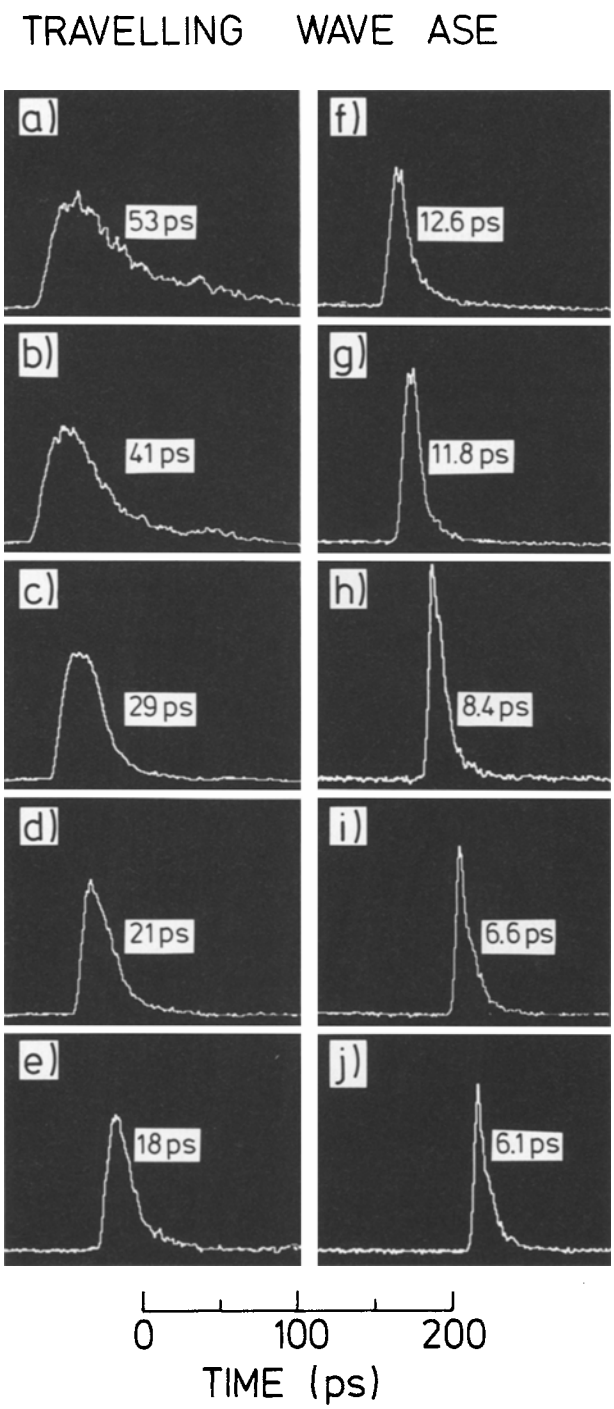

Fig. 2a–j. Travelling wave ASE pulses emitted in forward direction by a transversally excited solution of sulforhodamine B. Pump energy density increases from (a) to (j)

longitudinal arrangements which have been used so far [4–13]. The key element here is a 1200 mm^{-1} grating operating in the first order of diffraction. For each pitch of the grating an optical delay λ_p is introduced (λ_p: pump wavelength). Thus a continuous spatial delay is created across the diffracted beam [14, 15].

It is easy to show that if $\tan\gamma = n$ (n: refractive index of the dye solution; for γ cf. Fig. 1), then pump and ASE pulses are always in synchronism at any part of the excited volume, i.e. travelling wave ASE is excited.

The pump source was an EMG 501 XeCl-excimer laser-pumped distributed feedback dye laser oscillator-amplifier system [16], delivering 12 ps long pulses at 585 nm with a repetition rate of 10 pps. The energy incident on the dye cell was about 200 μJ.

Temporal Behavior of the ASE Pulses

Travelling wave ASE was readily observed with forward to backward ratios better than 50. The energy conversion efficiency of pump light to ASE was about 5%. Since the streak camera was available to us only for a very short period of time, optimization of the experimental conditions has not been attempted.

ASE pulses emitted in forward direction by a 8×10^{-3} M sulforhodamine B solution have been recorded with a HTV C1370 streak camera (Fig. 2). The length of the excited volume was 20 mm. Pump-pulse energy density was varied by shifting the cylindrical lens employed for focussing (cf. Fig. 2a–j, where j corresponds to strongest focussing). In this case the excited volume had a vertical height of about 40 μm and a penetration depth of 100 μm. The risetime of ASE pulse (j) is clearly limited by the time resolution of the streak camera which was $T(\text{instr}) = 4$ ps with the optics supplied to us. Of the same reason, the actual duration (FWHM) of the ASE pulse might even be shorter than 6.1 ps. Similar results have been obtained with a 2×10^{-4} M DODCI solution.

At the highest excitation intensity we observe a forward ASE pulse having half the width of the pump pulse (Fig. 2). ASE pulses emitted in the backward direction had durations of 300–500 ps.

Considerable shortening of the risetime of the ASE pulse with respect to that of the pump was predicted in [11]. Unfortunately, those calculations have been carried out for the case of longitudinal pumping and, therefore, the results cannot be applied directly to our experiments.

At present, we do not have a quantitative model for this effect. We think that the shortening is caused predominantly by steepening of the leading edge in the following way:

$$I_{\text{ASE}} \sim \text{Exp}(\text{const} \times \int I_{\text{pump}}(t)dt),$$

where $I_{\text{pump}}(t)$ is the temporal profile of the pump pulse and const $= f$(gain). As one can easily see, the leading edge of $I_{\text{ASE}}(t)$ can be much steeper than that of $I_{\text{pump}}(t)$, particularly, if the pump pulse rises faster than Gaussian.

The fall time of the ASE pulse is much shorter than the fluorescence lifetime of the dye molecules, since the latter is drastically decreased by stimulated emission.

Spectral Properties of the ASE Pulses

Figure 3 shows the spectrum of the ASE emitted in forward direction by the travelling wave excited solution of sulforhodamine B. It has been recorded with a spectral resolution of 0.15 Å and exhibits a considerable amount of fine structure, although this might be enhanced somewhat due to the fact that the recording was made on Polaroid film. Earlier experiments have apparently been carried out with instruments of lower resolution [6, 8, 13].

It might be that this structure is related to the statistical distribution of some "seed" spontaneous emission which could start from a very low level, because we estimate that small signal single pass gain in forward direction could be extremely high – up to 10^{20} for an excitation pulse of infinitely short duration – under our experimental conditions. In addition, nonlinear optical effects could enhance the contrast of the lines. The occurrence of such effects is likely because the output power density of ASE is in the GW/cm^2 range.

The widths of the individual spectral lines are about 0.5–1.0 Å corresponding to transform limited pulses of 5–10 ps duration.

Comparison of Transversal and Longitudinal Excitation of Travelling Wave ASE

1) In the case of *longitudinal* pumping the small signal single pass gain cannot exceed a value given by

$$\exp(\sigma_e/\sigma_p)$$

if the pump energy density is lower than the saturation energy density

$$E_s = h\nu/\sigma_p$$

($h\nu_p$: energy of the pump photons, σ_e: stimulated emission cross section of the dye at the emission wavelength, σ_p: absorption cross section of the dye at the pump wavelength).

σ_e and σ_p are typically of the same order of magnitude, therefore, the gain is usually insufficient for the observation of ASE. Only when the pump energy density was increased to about $250 \times E_s$(!) could one observe good ASE [10, 11]. In this particular case the concentration of the dye solution was chosen so as to give a small signal transmission of about $10^{-25} - 10^{-13}$ at the pump wavelength. At lower dye concentrations the number of available molecules was too small to produce high enough gain even in the case that all molecules were excited to the lasing level [10]. At higher concentrations the pump pulse was absorbed in a thin layer at the beginning of the sample, and a travelling wave effect was not to be expected [10]. Besides, when the pumping energy density is $250 \times E_s$, practically all molecules at the beginning of the sample are in the excited state and thus a major fraction of the exciting photons is lost by excited state absorption.

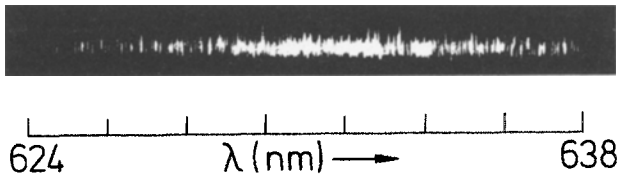

Fig. 3. Spectrum of travelling wave ASE of sulforhodamine B. Spectral resolution was 0.15 Å

In the case of *transversal* pumping (Fig. 1) small signal single pass gain can exceed 10^8 even if only 2% of the molecules are in the excited state, assuming a dye concentration of 8×10^{-3} M/l, dye cell length 20 mm, and $\sigma_e = 10^{-16}$ cm^2. Thus excited-state absorption of the pump photons is negligible and the pump energy is used more efficiently than in the *longitudinal* arrangement. Moreover, the selection of the dye concentration is not as critical as for *longitudinal* pumping.

2) For *longitudinal* pumping with high pump-energy density self-focussing can easily occur [13]. This is an unwanted effect which is not to be expected in the case of *transversal* pumping because the energy densities are much lower.

3) In the case of *longitudinal* pumping the group velocities of pump and ASE pulses are equal. – This is true if the dispersion of the refractive index of the dye solution and the group velocity delay of the pump pulse due to the preferential absorption of the leading front of the pulse are negligibly small.

In contrast, for *transversal* pumping the relative speeds of ASE and pump pulses can easily be varied by changing the angle γ (Fig. 1). Small deviations from the relationship $\tan\gamma = n$ could have a strong influence upon the duration and shape of the ASE pulses. Control of the angle might, therefore, establish a method of pulse tailoring.

4) Spatial inhomogeneities of the pump beam can cause serious experimental difficulties in the case of *longitudinal* pumping, while the requirements in this respect are reduced for the *transversal* arrangement since some kind of averaging occurs in one direction.

We believe that, in our experiments, the shortening of the ASE pulse with respect to the pump pulse has been achieved with much lower pump energies as compared to other work [8, 10, 11, 13], because of the above listed advantages of *transversal* pumping.

Conclusions

A new scheme for travelling wave excitation of ASE has been studied. It consists of a transversal pumping arrangement offering significant advantages over longitudinal arrangements.

The duration of the ultrashort laser pulse was reduced by a factor of two by travelling wave excitation of ASE. It is very likely that the wavelength of ASE pulses can be adjusted with an accuracy of at least ± 5 nm by using different dye solutions. Travelling wave ASE appears to be very well suited for generation of relatively broad probe continua in picosecond spectroscopy experiments using mode-locked neodymium lasers.

The application of the travelling wave transversal excitation could probably be used in a picosecond pulse amplifier pumped by a mode-locked Nd : YAG laser [15, 17]. The expected advantages arise from the fact that by proper synchronization of the pump pulse with the pulse to be amplified broadening due to preferential amplification of the leading part of the pulse [3] could be avoided. Besides, higher small signal gain per stage can be obtained, since single pass gain reduction by ASE [3] is expected to be much smaller than in the case of stationary excitation.

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