Appl. Phys. B 46, 151–155 (1988)

Applied Physics B Photophysics and Laser Chemistry



# Theory of Travelling-Wave Amplified Spontaneous Emission

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Received 13 January 1988/Accepted 7 February 1988

**Abstract.** A rate-equation model for describing the travelling-wave amplified spontaneous emission pulses (TWASE) in a transversally excited travelling wave arrangement is given. 6.35 ps long ASE pulses have been obtained by 12 ps long pump pulses. The effect of pump intensity, pump-pulse duration, molecular parameters of the dyes and pump-sweep velocity on the ASE pulses is studied.

**PACS:** 42.55M

Dye solutions excited by pulsed lasers can have extremely high optical gain. If the excited volume has a relatively long and narrow shape, amplified spontaneous emission (ASE) is generated [1–3]. ASE shows laser-like properties, i.e. it has low divergence and it undergoes spectral narrowing in respect to the spontaneous emission.

Travelling-wave amplified spontaneous emission (TWASE) generated by longitudinal pump arrangement [4–6] has been studied to determine molecular parameters of dyes and to develop tunable broad-band ps probe beams for ps spectroscopy.

In order to avoid the difficulties of longitudinal pumping (i.e., self focussing and excited state absorption of pump light) a transverse travelling-wave pump arrangement has been introduced [7–9]. The key element of this setup is a diffraction grating which introduces a continuous temporal delay across the beam [10–11]. This setup turned out to be very effective. E.g. using this arrangement, TWASE was generated even in dyes having fluorescence quantum yield as low as $5\times10^{-4}$ [9].

The diffraction losses of the grating can be eliminated by using the prism setup proposed in [12].

Narrow-band tunable ps pulses can be generated by external filtering and subsequent amplification of TWASE [9, 13].

In [14] TEA $N_2$ laser was used to generate TWASE radiation.

In a recent paper by Lobentanzer [15] model calculation of TWASE was given for the IR dyes No. 5. Since this dye has 2.7 ps absorption recovery time and $5\times10^{-4}$ quantum yield, the results of calculations cannot be applied to experiments of [7, 13, 14], where dyes (Sulforhodamine B, QUI, Rhodamine 6G, Rhodamine B, Coumarine 311) having ns lifetimes and quantum yields approaching unity have been used.

In the present paper model calculations corresponding to the experimental conditions of [7] are presented. The effect of velocity mismatch between pump sweep-speed and light velocity is also discussed.

## 1. Theoretical Model

The transversally pumped TWASE is described by the following set of equations (Fig. 1)

$$\partial n(x,t)/\partial t = I_p(x,t)\sigma_p[N-n(x,t)] - n(x,t)/\tau - [I^+(x,t)+I^-(x,t)]\sigma_e n(x,t), \quad (1)$$

$$\pm\partial I^{\pm}(x,t)/\partial x + (\eta/c_0)\partial I^{\pm}(x,t)/\partial t = n(x,t)\sigma_e I^{\pm}(x,t) + n(x,t)S\Omega/\tau, \quad (2)$$

where the meanings of symbols are the following:

$N$: total concentration of dye molecules,

$n(x,t)$: the concentration of molecules in the $S_1$ vibration-rotation manifold,

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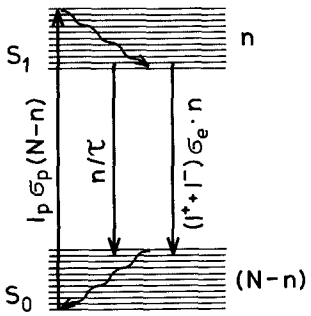


Fig. 1. Simplified energy scheme of the dye used for the rate equation model

$I^{\pm}(x,t)$: the intensity of TWASE propagating into the $+x$ and $-x$ direction, respectively,

$\tau$: fluorescence lifetime of $S_1$ state,

$\sigma_p$: absorption cross-section at the pump wavelength,

$\sigma_e$: emission cross-sections at the wavelength of TWASE,

$c_0$: speed of light in vacuum,

$\eta$: refractive index of the dye solution,

$L$: length of the excited volume.

The $S$ and $\Omega$ factors determine that part of the spontaneous emission, which propagates into the spectral and angular ranges of the TWASE, respectively.

$\Omega$ can be calculated as $\Omega = d^2 \cdot \pi / 4L^2$, where $d$ is the penetration depth of pumping into the dye solution. $d$ was approximated as $d = 1/(N \cdot \sigma_p)$. (This expression does not take into consideration the bleaching of absorption, which at the highest pump intensities was 25%).

In the above calculations, it was supposed that the penetration depth of pumping and the vertical size of the excited volume [Ref. 7, Fig. 1] are the same.

For $t - \eta x/c < 4T_0$ the pump intensity $I_p(x,t)$ is 0. For other cases it is regarded to be Gaussian in time with pulse duration (FWHM) $T_0$, uniform along the dye cell and sweeping with a speed of $c/\eta$ along the dye cell into the direction of the $x$ axis:

$$I_p(x,t) = I_{p0} \exp\left( -4 \ln 2 \frac{(t - \eta x/c - 4T_0)^2}{T_0^2} \right) \quad (3)$$

if $t - \eta x/c \geqq 0$, otherwise $I_p(x,t) = 0$.

In this model, the following approximations are used:

The effects of triplet states and higher excited states are neglected.

The non-radiative processes within each manifold are regarded to be much faster than the shortest pump-pulse duration (12 ps) and the expected shortest TWASE pulse durations (6 ps).

The reduction of pump intensity in the transverse direction (into the dye medium) due to absorption is neglected.

The polarization dependence of absorption and emission cross-sections, directional distribution and rotational relaxation of the dye molecules are neglected.

The spectral dependence of the emission cross-section in the spectral range of TWASE is neglected.

The above approximations have been successfully used in theories describing ASE [1–2], the behaviour of ps amplifiers [3] and the temporal behaviour of transient effects in pulsed dye lasers [16–19].

We used these approximations not only to simplify the calculations but also to see clearly the effect of travelling wave excitation by itself, without any side effect taking place in the dye molecule.

The numerical values of the parameters corresponded to the experiments described in [7] and to the molecular parameters of sulforhodamine B: $N = 6 \times 10^{17}$ cm$^{-3}$, $\sigma_p = 10^{-16}$ cm$^2$, $\sigma_e = 1.4 \times 10^{-16}$ cm$^2$, $\tau = 4 \times 10^{-9}$ s, $\eta = 1.36$, $L = 2$ cm, $\Omega = 5.5 \times 10^{-5}$, $S = 0.1$, $c_0 = 3 \times 10^{10}$ cm/s.

The following initial and boundary conditions have been used: $n(x,0) = 0$, $I^+(0,t) = 0$, $I^-(L,t) = 0$. The equations have been solved using the method of first-order finite differences.

## 2. Results and Discussion

### 2.1. Dependence on Pump Intensity

Figure 2 shows the calculated TWASE pulses on the output plane. The pulses are normalized to their peak intensities. On the output plane ($x = L$) the pump pulse has its maximum at $t = 138.6$ ps, which is equal to the transit time of light through the 2 cm long dye cell plus $4 \cdot T_0$, see [3]).

The pulses are asymmetric, having faster rise time and longer fall time. This is very similar to the experimental observations [Ref. 7, Fig. 2].

Figure 3 shows the durations (FWHM) of the pulses as a function of the peak intensity of the pump pulses.

Figure 4 exhibits the peak power of the output pulses for different pump intensities.

For pump intensities exceeding $1.4 \times 10^{26}$ cm$^{-2}$ s$^{-1}$ the TWASE pulse duration is 6.35 ps. This value is called saturated pulse duration ($T_{sat}$). When the pump intensity is increased, no further

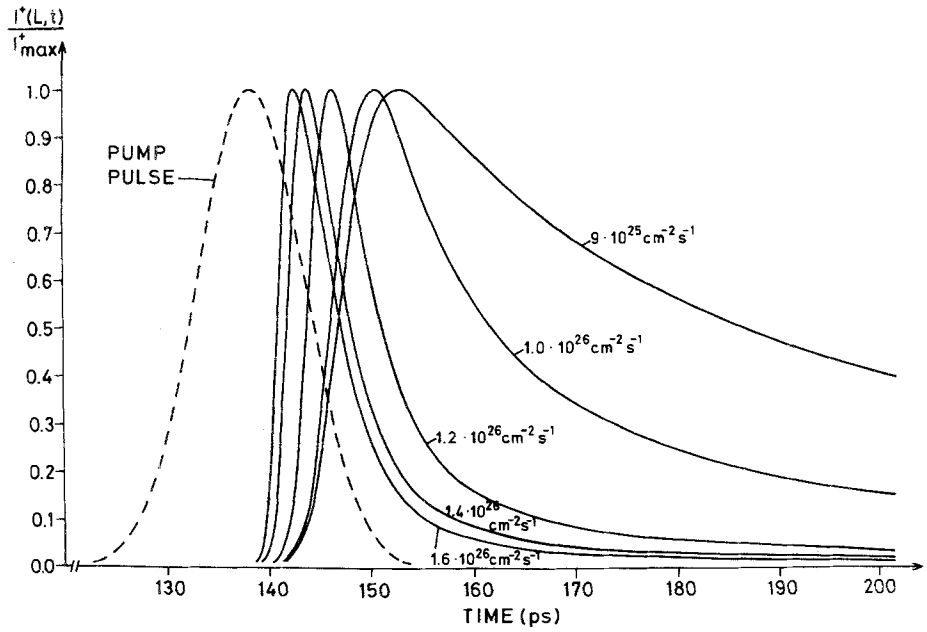


Fig. 2. TWASE pulses for different pumping levels. The pump pulse shape is also shown

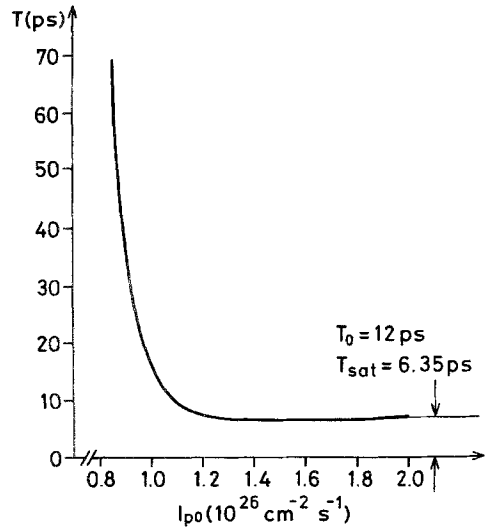


Fig. 3. Duration of the TWASE pulses (FWHM) propagating into the right direction for different pumping intensities

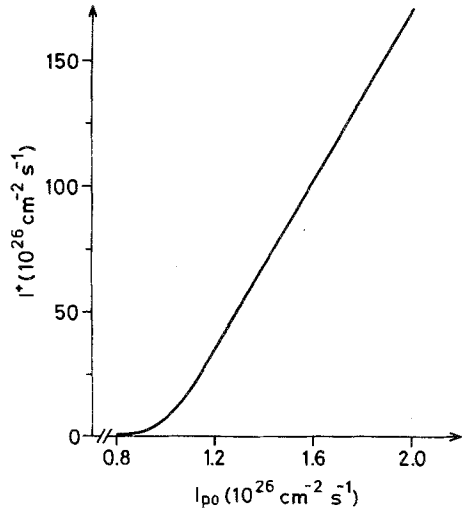


Fig. 4. Dependence of output peak power for different pump intensities

pulse shortening occurs. $T_{sat} = 6.35$ ps is in good agreement with the measured value of 6 ps [7].

The pulse shape of the saturated pulses is practically independent of the pump power (Fig. 5).

On increasing the pump intensities, the computation time was rapidly increasing. For pump intensities exceeding $2 \times 10^{26}$ cm$^{-2}$s$^{-1}$, the calculations have not been carried out. [The reason for that is the following. Neglecting the effect of stimulated emission for the pump intensity $2 \times 10^{26}$ cm$^{-2}$s$^{-1}$ from (1) we obtain $n = 1.53 \times 10^{17}$ cm$^{-3}$, which gives a small-signal single-pass gain of $4.03 \times 10^{18}$. Such extremely high gain requires very small steps of integration leading to large computation times.]

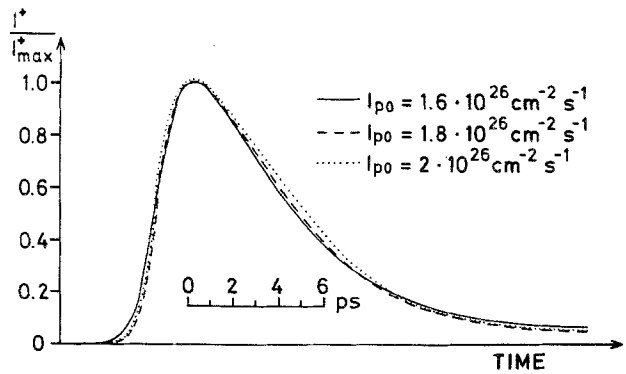


Fig. 5. The normalised TWASE pulse shapes for pump intensities exceeding $1.4 \times 10^{26}$ cm$^{-2}$s$^{-1}$ are practically unchanged

Equations (1, 2) take into account the reverse ASE, i.e. ASE fluxes propagating into the $-x$ direction, too. This is not the case in [15]. For pump intensities, when saturation occurs, i.e. for $I_{p0} > 1.4 \times 10^{26}$ cm$^{-2}$s$^{-1}$, the duration of reverse ASE is around 25 ps. The forward-to-backward energy ratio is 5.5. The shortening of the reverse ASE is caused by the fast depopulating effect of the TWASE propagating into the forward direction.

### 2.2. *The Effect of Mismatch of Excitation-Sweep Velocity on the Parameters of* TWASE

The travelling-wave condition is exactly fulfilled, if the pumping pulse sweeps over the dye cell with a speed of $c_0/\eta$ [7]. This happens in the model $c = c_0$, see (3).

Figure 6 shows the TWASE pulse durations for the sweep-velocity range $c/c_0 = 2/3 - 3/2$. According to this, the pulse duration has a minimum value at $c = c_0$.

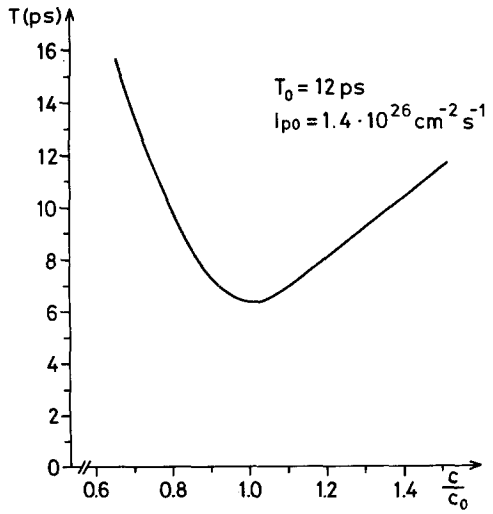


Fig. 6. Duration of the TWASE pulse for different pump sweep-velocities. $c/c_0$ is the measure of the mismatch between the speed of pumping and propagation speed of the light in the dye solution

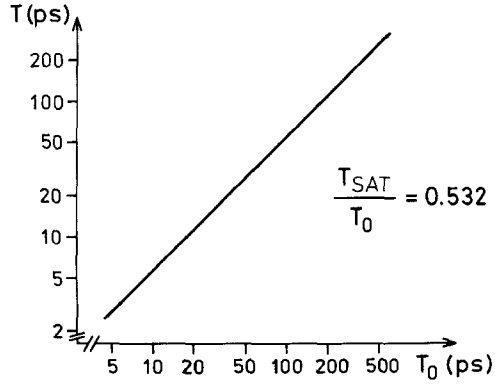


Fig. 7. TWASE pulse duration as a function of pump pulse duration

Since experimentally it is easy to control both $\eta$ and $\tan\gamma$ [7] with an accuracy of 5%, from Fig. 6 it can be concluded that travelling-wave pumping condition can be easily fulfilled experimentally, with such precision, that no noticeable broadening occurs.

### 2.3. *The Effect of Other Parameters on* TWASE

1) Equations (1, 2) depend on the product of $I_p \cdot \sigma_p$ and not on these parameters separately.

2) When increasing $\sigma_e$ by a factor of $M$ and decreasing the pumping rate $I_p \cdot \sigma_p$ by the same factor, no noticeable change in the temporal shape of pulses is observed.

3) The value of $\tau$ does not affect the results as long as $\tau \gg T_0$ holds.

4) The solutions of equations are practically independent of the numerical value of $\Omega$. Solving the equations with two values of $\Omega$, differing by a factor of 10, we did not observe significant change. This also approves the validity of the approximation $d = 1/(N \cdot \sigma_p)$ given in Sect. 1.

5) The saturated pulse duration (Fig. 3) is proportional to the pump-pulse duration. The calculated ratio of this durations is 0.532.

This holds as long as $T_{\text{pump}} \ll \tau$ is fulfilled.

6) From 1)–5) it follows that TWASE can be generated by all typical laser dyes, and the main properties are expected to be the same as for sulforhodamine B.

It should be noticed that the above results are different from the predictions of [15], where symmetric pulses were observed both theoretically and experimentally. This is probably due mainly to the difference in fluorescence lifetime between sulforhodamine B (4 ns) and dye No. 5 (2.7 ps). Due to this, in our experiments $\tau \gg T_0$, while $\tau \ll T_0$ in [15].

Therefore, in [15], the pump pulse creates a moving Gaussian "population-wave", while in our case a moving "population-wave", having the form of the integral of the Gaussian pump pulse, is formed.

We solved (1–3) with the molecular parameters for dye No. 5, i.e. $\sigma_p = 4 \times 10^{-16}$ cm$^2$, $N = 5 \times 10^{17}$ cm$^{-3}$, $\sigma_e = 5 \times 10^{-16}$ cm$^2$, $T_0 = 4.5$ ps [9]. The TWASE pulse duration as a function of pump intensity calculated by our simple model is similar to that was given in [15]. For $1.5 \times 10^{26}$ cm$^{-2}$s$^{-1}$ pump intensity we obtained 1.7 ps long pulses having only a very little asymmetry (risetime from the half-peak intensity to the peak is 10% shorter than the fall time to the same level). This is about half of the experimental result of [9], where using 4.5 ps long Gaussian pump pulses 3.5 ps long symmetric TWASE pulses have been obtained.

In [14] 45–55 ps long TWASE pulses were generated by using the 700 ps long pump pulses from a

TEA $N_2$ laser. Our model calculation gave under the experimental conditions of [14] 370 ps long TWASE pulses, which is about 7–8 times longer than the measured ones.

First we thought that self-absorption of TWASE radiation by the dye molecules might be responsible for the pulse shortening, therefore self-absorption was also included in (1–3). Although pulse shortening was observed in the solutions, it cannot be the main effect, because pulse shortening was experimentally observed with coumarine 311 dye which has practically no self absorption.

In [9], in some experiments, the pump intensity distribution showed a gradient along the dye cell. We made calculations where $I_{p0}$ in (3) was replaced by $0.4 \cdot I_{p0}(1+3x/L)$, which ensured a four-fold linear increase in intensity along the dye cell, while keeping the total pump power unchanged. However the calculations showed no change in the TWASE pulse shape.

Besides, our calculation predicted that under those experimental conditions [14] there was practically no difference between travelling-wave and non-travelling-wave pumping. This is quite plausible, considering the 700 ps long pump-pulse duration and 2 cm long dye cell [14].

Since the data presented in [14] give convincing experimental evidence of short ASE pulse generation, we suppose that there are some other effects not included in (1–3) and not mentioned in [14], either, which contribute to the observed pulse shortening.

## 3. Conclusions

A simple model describing the behaviour of transversally excited TWASE is given.

In order to see clearly the effect of travelling-wave excitation, all side effects (such as intramolecular relaxations, effects of higher excited states, triplet states, reabsorption, orientation distribution and relaxation of molecules, transverse dependence of population etc.) are excluded from the analysis.

The effects of pump intensity, pump-pulse duration, velocity mismatch between pump sweep and light speed are studied.

The calculations are in good agreement with the results of [7–9, 15].

*Acknowledgements.* This work has been supported by the 359/84 Foundation of the Hungarian Academy of Sciences.

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