Pulse-train formation in a gain-switched polymer laser resulting from spatial gain inhomogeneity

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We present an experimental study of the dynamics of a polymer laser upon excitation with an ultrashort pump pulse. The laser is unusual in the sense that the cavity length is such that the cavity round-trip time is intermediate between the duration of the pump pulse and the spontaneous-emission time of the gain medium. The cavity is only partly filled with gain medium. The unusual time scales, in combination with the spatial gain distribution, result in the emission of a neat and regular train of short pulses. Its generation can be viewed as a special case of gain switching. A one-dimensional rate-equation model that takes the nonuniformity of the gain explicitly into account is presented. The model describes both the evolution of the inversion, which is also measured experimentally, and the dynamics of the output in great detail, and yields excellent agreement with the experimental results. A heuristic description of the arisal of the pulse train in terms of amplified spontaneous emission and gain depletion is given.

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I. INTRODUCTION

Gain switching is a well-known technique for short-pulse generation, and has been applied to a wide variety of laser systems like semiconductor lasers [1,2], Ti:sapphire lasers [3–5], and gas lasers [6]. It is accomplished by modulating the pump on a time scale short compared to the spontaneous-emission lifetime of the upper-laser level. The ensuing time-varying gain then gives rise to the production of a laser pulse that is much shorter than the spontaneous-emission lifetime, and, in some cases, even substantially shorter than the pump pulse. Synchronous pumping can be viewed as an extension of gain switching. Here, the gain medium is excited repetitively, with the excitation rate synchronized to the cavity length. Due to this repetitive gain switching, mode locking is achieved and a stable train of short pulses, considerably shorter than the pump pulses, is emitted [7,8].

Here we study the dynamics of the light emitted by a short-cavity polymer laser that is pumped by a single ultrashort pulse. The cavity dimensions of this laser are such that the cavity round-trip time is intermediate between the duration of the pump pulse, and the spontaneous-emission time of the gain medium. Additionally, the gain medium only partly fills the cavity. Such a laser shows interesting dynamics [9]. Because of the short duration of the pump pulse, the dynamics can be transparently analyzed. The pump pulse is so short that the laser evolves completely freely and gain switching provides a proper framework for a description of its dynamics.

In contrast to most gain-switched systems, our short-cavity polymer laser does not emit a single short pulse. Rather, it emits a neat and regular train of short pulses; we attribute this to a combination of the unusually ordered time scales and the inhomogeneous distribution of the gain along the cavity axis. Since gain switching still provides a useful framework for the understanding of the phenomena, we interpret the pulse train as a result of multiple gain switching.

In the present paper, we show the results of an experimental study of the pulse evolution and the dynamics of the population of the upper laser level from the very start, both below and above the laser threshold. Due to the convenient choice of cavity round-trip time, we are able to record various aspects of this pulse train, such as the rise and decay time of the individual pulses and the evolution of the train in great detail. We notice that the pulse repetition rate is given by twice the longitudinal mode spacing and that the pulse train shows odd-even staggering. All results are explained with help of a ''local'' rate-equation model, i.e., a model where the spatial distribution of gain and loss is explicitly taken into account. The rate equations are solved numerically.

II. EXPERIMENTAL SETUP

In the experiment, we employ a 25 mm long cavity consisting of a concave high reflector (50 mm radius of curvature) and a flat output coupler ($T=10\%$). Approximately at its center sits a dye cell through which the polymer solution flows continuously (see Fig. 1). The cell is slightly tilted ($\approx 10°$) relative to the cavity axis in order to avoid optical feedback from its walls. As gain medium, we use a 0.2% by weight solution of a copolymer of poly[p-phenylene vi-

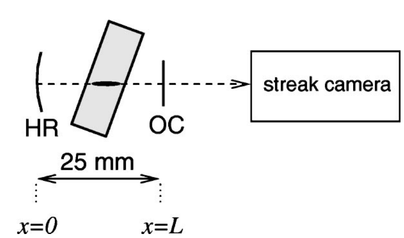

FIG. 1. Schematic of the experimental setup: The shaded area represents the dye cell, with the excitation region indicated by the oval. To avoid optical feedback from the walls of the dye cell, the latter is slightly tilted.

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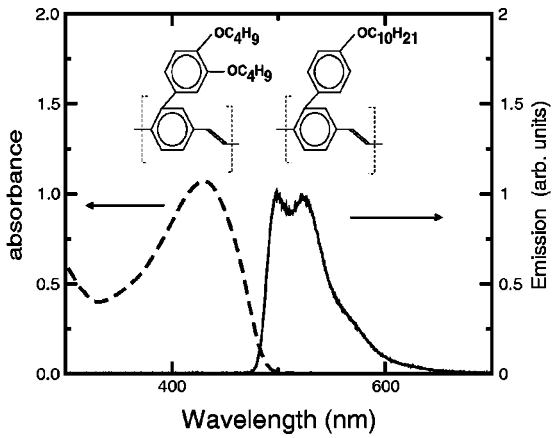

FIG. 2. Absorption and emission spectra of the dissolved copolymer. The two repeat units are shown with the spectra.

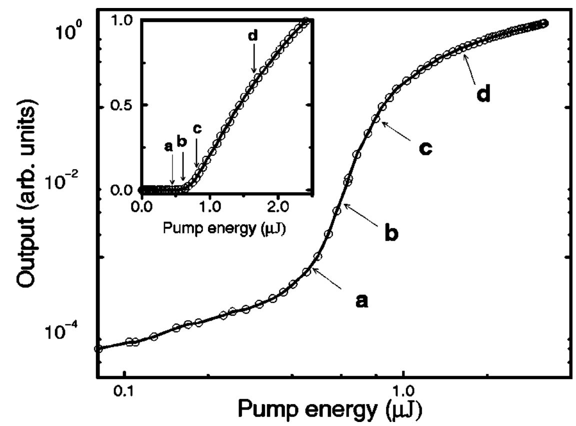

FIG. 3. Input-output curve of the polymer laser. The time-integrated light output as measured within a small solid angle, is plotted versus pump energy on a double-logarithmic scale. In the inset, the same curve is plotted on a linear scale. The pump energies at which our measurements have been performed are indicated.

nylene (PPV)]-derivatives in chlorobenzene. PPV is a popular and well-known light-emitting polymer and laser action in PPV and its derivatives has been observed both in solution and in solid state [10–13]. Details on the materials used in our studies have been published in Ref. [14]. The radiative lifetime of this material in solution is 0.86 ns; its absorption and emission spectra are shown in Fig. 2, where the polymer repeat units are also shown.

The polymer laser is side pumped with 130 fs pulses at $\lambda = 400$ nm with a repetition rate of 1 kHz. The pump pulses are generated with a Ti:sapphire regenerative amplifier system (Spectra Physics, Spitfire), seeded by a Ti:sapphire oscillator (KM-labs). The 800 nm output of the Ti:sapphire laser system is frequency doubled in β-barium borate (BBO). The blue pump light is focused into a pencil-shaped volume ($\approx 7 \times 0.05 \times 0.05$ mm) inside the polymer solution, the transverse dimension of the volume being of the same order as that of the fundamental mode of the laser resonator. The polarization of the pump is perpendicular to the cavity axis. Above threshold, the laser emits light in a narrow band ($\Delta\lambda \approx 12$ nm) centered at $\lambda = 530$ nm. We simultaneously measure the dynamics of the light emitted into lasing, and into nonlasing modes, i.e., in a sideways direction. The latter provides us with a measure of the evolution of the population of the upper-laser level [15–17]. In all experiments on the laser dynamics we employ a streak camera (Hamamatsu, C1587) providing us with a temporal resolution of approximately 10 ps.

III. EXPERIMENTAL RESULTS

For a range of pump-pulse energies, from slightly below to far above threshold, we have measured the temporal evolution of the light emitted by the polymer laser upon excitation by a *single* ultrashort pulse. These pump energies are indicated on the input-output curve that is presented in Fig. 3.

As shown by Fig. 4, a stable pulse train develops for all values of the pump [18]. On the scale shown here, the pump pulse (at $t = 0$) is infinitely short. Figure 4(a) shows the output just below threshold: initially the output grows rather smoothly but pulsations start to appear after a few cavity round trips. The output keeps growing, while the modulation depth increases and the pulses become narrower. The pulse-train envelope shows a maximum after ≈ 1.1 ns. As indicated in the figure, two pulses are emitted within one cavity round-trip time, i.e., the laser emits two interleaved pulse trains. It is observed that the envelopes of both pulse trains are quite different, in other words: the laser output exhibits odd-even staggering. These points will be discussed in more detail later in the paper.

The other frames of Fig. 4, containing the results for higher and higher pump energies, show a basically similar behavior, although the initial growth of the signal is less easily observed. The pulse structure, however, is much more fully developed, i.e., the modulation depth is substantially larger. Overall one notices that further above threshold, the

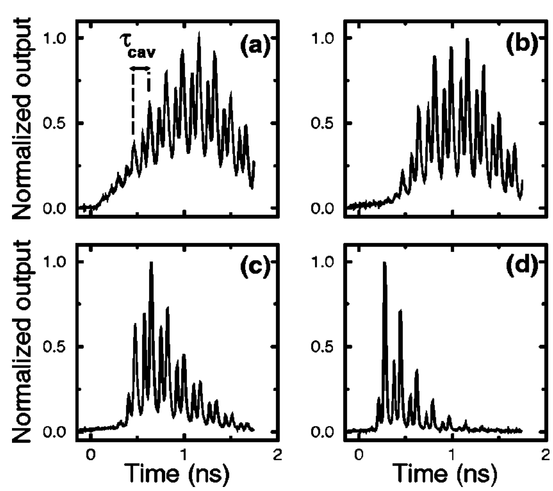

FIG. 4. Temporal dynamics of the polymer laser output for pump energies ranging from below threshold to far above threshold, as indicated in Fig. 3. Pump parameters: $r = 0.77$, $r = 1$, $r = 1.3$, $r = 2.8$, for graphs (a)–(d), respectively. The pump parameter is defined as the pump energy normalized to the experimentally determined threshold value of the pump energy.

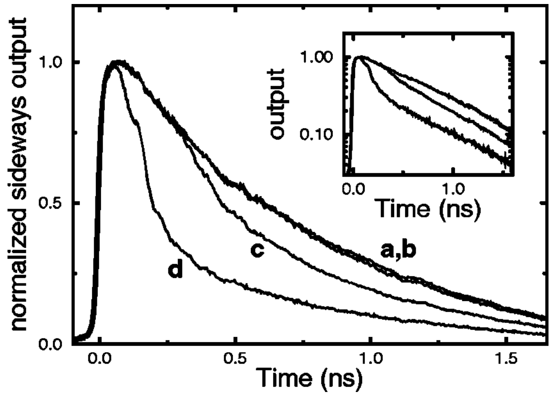

FIG. 5. Population decay, measured by means of sideways emitted light. The indices a–d correspond to the pump energies indicated in Figs. 3 and 4. The inset shows the results on a logarithmic scale.

highest peak occurs earlier and earlier. Below, we will show that this is caused by gain depletion.

Figure 5 shows the evolution of the sideways emitted photoluminescence for the same values of the pump strengths as in Fig. 4. Curve (a) shows the results for the case where the laser is operated below threshold. As can be seen from the inset, where the curves are plotted on a logarithmic scale, the population decays exponentially in this case, as represented by the lowest straight line. The slope of this line yields the photoluminescence lifetime of the material. Curves (b) through (d) show the dynamics of the upper-laser level population for pump strengths ranging from slightly above to approximately three times threshold, i.e., through the range where gain depletion becomes important. This is best illustrated in curve (c): initially, the population of the upper-laser level decays by spontaneous emission only. However, the intracavity intensity grows, and after approximately 0.3 ns, stimulated emission leads to substantial depletion of the inversion. In curve (d), the gain depletion sets in almost immediately; here the population of the upper-laser level is seen to decrease in a stepwise fashion. These steps are correlated with the transit of the optical pulses through the gain medium.

IV. RATE EQUATIONS

As a laser material, a dissolved light-emitting polymer shows very similar behavior to that of a laser dye in solution [10,12,13]. We therefore assume the gain to be homogeneously broadened, the intraband relaxation to be very rapid, and the lower-laser level to be empty [19]. Hence, the inversion is given by the population of the upper-laser level. We thus model our laser as an homogeneously broadened ideal four-level laser [13]. Furthermore, we use a one-dimensional description where we separate the intracavity photon density in two fluxes that travel in opposite directions through the cavity. This approach has been fruitfully applied to other systems where the gain and/or loss are inhomogeneously distributed [20,21]. Examples are systems based on traveling-wave amplified spontaneous emission [19,22–24] and semiconductor lasers where the spontaneous-emission noise is described in such terms [25].

For the evolution of the inversion density $N(x,t)$ and for the left- and right-traveling photon densities $[n^-(x,t)$ and $n^+(x,t)$, respectively$]$ we write the following rate equations (in analogy with Ref. [19]):

$$\frac{\partial N(x,t)}{\partial t} = -\gamma N(x,t) - c\sigma\eta N(x,t)[n^+(x,t)+n^-(x,t)],$$

$$\frac{\partial n^+(x,t)}{\partial x} + \frac{\eta}{c}\frac{\partial n^+(x,t)}{\partial t} = \sigma N(x,t) n^+(x,t) + \frac{\gamma\beta\eta}{c} N(x,t),$$

$$-\frac{\partial n^-(x,t)}{\partial x} + \frac{\eta}{c}\frac{\partial n^-(x,t)}{\partial t} = \sigma N(x,t) n^-(x,t) + \frac{\gamma\beta\eta}{c} N(x,t). \quad (1)$$

These rate equations are defined on the interval $x=0$ through L, where L is the length of the cavity. In Eq. (1), γ is the spontaneous-emission rate, σ denotes the stimulated-emission cross section, η the refractive index of the polymer solution, c the speed of light in vacuum, and β the fraction of spontaneous photons emitted in the lasing mode. Note that Eq. (1) does not contain the usual pump term. The pump pulse is so short in comparison to all other relevant time scales that its effect is well represented by setting the value of the inversion density distribution $N(x,0)$ at $t=0$. Also, again in contrast to common practice, our rate equations do not include a term describing the cavity losses. In our laser, the dominant losses are not homogeneously distributed over the cavity, as is usually assumed, rather, they are localized at the output coupler and at the windows of the flow cell. The localized losses are taken into account via the conditions by which the left- and right-traveling photon densities are coupled at the cavity mirrors

$$n^+(0,t) = T_{\mathrm{cell}}^2\, n^-(0,t), \quad (2)$$

$$n^-(L,t) = R_{\mathrm{OC}} T_{\mathrm{cell}}^2\, n^+(L,t). \quad (3)$$

Here, $L=25$ mm represents the distance between the mirrors, $R_{\mathrm{OC}}=0.9$ the power reflection coefficient of the output coupler (for the high reflector, we assume $R_{\mathrm{HR}}=1$), while $T_{\mathrm{cell}}\approx 0.85$ is the power transmission coefficient of a single window of the flow cell. This number is rather low because of diffraction losses at the edges of the flow cell.

We numerically solve the set of coupled rate equations presented in Eq. (1), combined with the coupling relations of Eqs. (2) and (3). As initial conditions we take $n^{\pm}(x,0)=0$ and $N(x,0)=N_0\exp[-(x-x_0)^2/\sigma^2]$. As indicated, the initial inversion is assumed to have a Gaussian distribution (since the gain medium is transversely pumped) with a full width at half maximum equal to about $\frac{1}{3}$ of the cavity length ($\sigma = L/6\sqrt{\ln 2}$). We position the gain medium close to the center of the cavity ($x_0 \approx L/2$). For our simulation, we use the following values for the numerical coefficients: $\gamma = 1.5\cdot 10^9\ \mathrm{s}^{-1}$, $\eta=1.5$, $c=3\cdot 10^8$ m/s, $\sigma=10^{-16}\ \mathrm{cm}^2$,

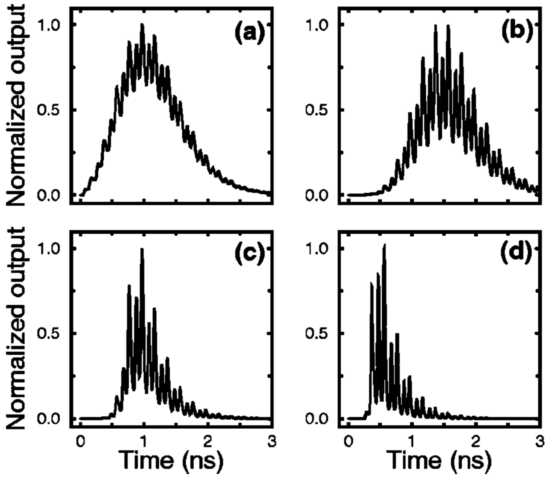

FIG. 6. Simulation of the dynamics at various pump energies from just below threshold to five times above threshold.

(order of magnitude of conventional laser dye) and $\beta = 10^{-6}$. To solve the rate equations, we use an Euler-type method, dividing the cavity in small steps, and approximating the differentials by finite steps ($\Delta x = L/100$ and $\Delta t = \eta \Delta x/c$).

The results for the photon flux $cn^{+}(L,t)$ at the output coupler are shown in Fig. 6 for several values of the peak inversion density N_0, normalized to its value at the laser threshold. The latter is found from an input-output curve that is calculated from the same rate equations. In full agreement with the experimental results of Fig. 4, the calculated photon flux exhibits a pulsed structure that develops after a few round trips. The calculated flux shows the same periodicity as in the experiment and exhibits the odd-even staggering of the pulse heights. The excellent agreement of the results of the model with the experimental data indicates that our model is sufficiently detailed. Hence, one may conclude that our model accurately describes the coupling between the left- and right-traveling solutions and that additional couplings (e.g., grating formation) are rightfully neglected. The accuracy of the numerical method has been verified by showing that the same results are obtained when the stepsize is reduced.

Additional insight into the laser dynamics is obtained from the simulation of the evolution of the inversion density in the center of the gain medium: $N(L/2,t)$. The results for this quantitiy are shown in Fig. 7 for the same four values of the pump energy as in Fig. 6. In curves (a) and (b), corresponding to the pulse trains of Figs. 6(a,b) the inversion density decays in a purely exponential fashion, i.e., the gain is not depleted by the stimulated-emission process. Initially, curve (c) also follows exponential decay. However, after ≈ 0.6 ns, the reduction of the inversion accelerates very rapidly. This point corresponds to the onset of gain depletion. In curve (d), the inversion decays very rapidly almost from the start. Here, the initial gain is so high that gain depletion sets in immediately. In curves (c) and (d), one notices that the reduction of the inversion has steplike features. These steps can be associated with the passage of the short optical pulse through the gain medium.

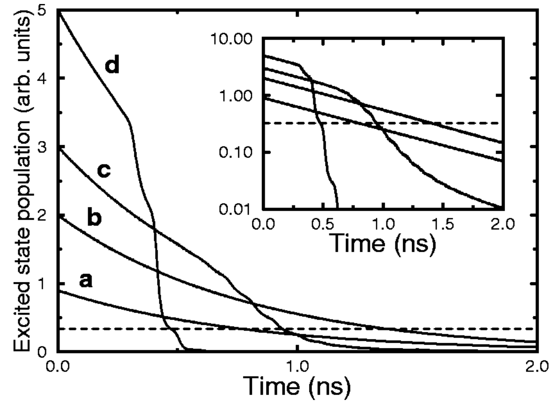

FIG. 7. Calculation of the population decay corresponding to the dynamics shown in Fig. 6. Note the onset of gain saturation and the steplike decay of the inversion in curves (c) and (d). The dashed line indicates the population of the excited state at which the gain is equal to the losses. The cross points of the ''loss line'' and the decay curves indicate the time at which maximum output is reached (compare to Fig. 6).

The horizontal line in Fig. 7 indicates the equivalent cavity loss. It is now easy to understand the overall features of the pulse trains in Fig. 6. The pulse height will grow as long as the inversion is larger than the value indicated by this line. And, obviously, the pulse height will decrease (rapidly) when the inversion has dropped below this line. The intersection of the various curves in Fig. 7 with the ''loss line'' sets the position of the maximum in each pulse train. As we will demonstrate below, optical gain is an essential ingredient for the generation of the strongly modulated output. Since the inversion density in Fig. 7 always starts above the loss line, net gain is present in all cases, and consequently, pulse sharpening is observed for all curves in Fig. 6

Here one clearly sees that the common threshold condition formulated as ''the gain is equal to the loss'' is ill defined for this system. Because of its time dependence, the gain equals the loss only at a specific time t_e. For $t < t_e$ the gain exceeds the loss, resulting in amplification of the intracavity photon flux, while for $t > t_e$ the system is lossy. As argued above, under these conditions, pulses will arise due to stimulated emission. However, when the initial inversion is not too large, the stimulated-emission process may come to an end before a powerful coherent beam if formed. Consequently, the time-integrated laser output may indicate that the laser is below threshold, even though, initially, the gain exceeded the loss, tempting one to conclude that the laser is above threshold. The lack of clarity in the threshold definition for this type of system, is succinctly discussed in Ref. [4].

Based on its appearance, one may be tempted to interpret the pulse train emitted by our laser as an example of a relaxation oscillation (spiking). However, relaxation oscillations in lasers develop because of depletion and subsequent recovery of the population of the upper-laser level [26]. In our system, the pump is switched off immediately; hence this population cannot recover. It is thus inappropriate to view the pulse train as a relaxation oscillation. As we will show below, an appropriate interpretation for the generation and

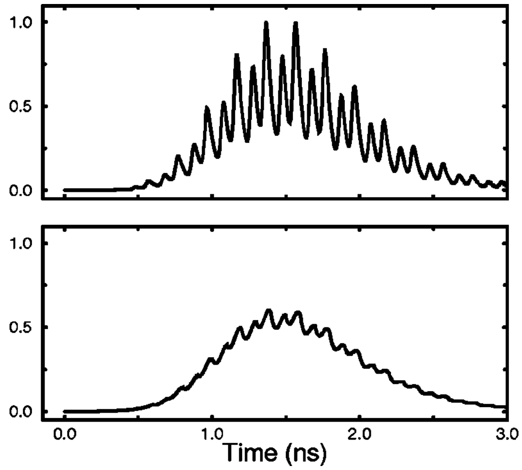

FIG. 8. Comparison between the evolution as shown in Fig. 6(b) (upper frame) and a laser that is pumped with an excitation stripe that is four times longer (lower frame). The number of excited molecules is the same in both cases.

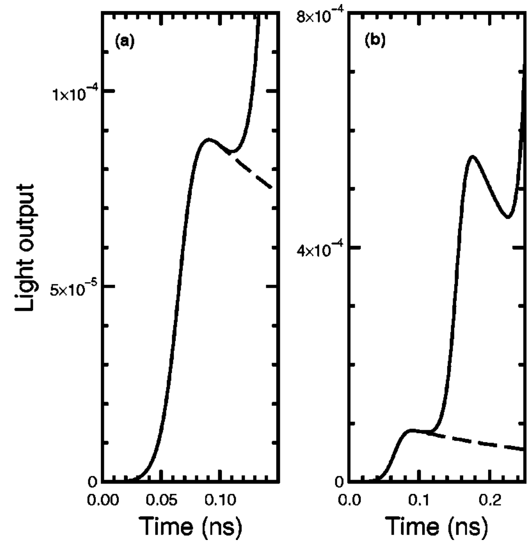

FIG. 9. Detail of the initiation of the pulse train. Frame (a) shows only the first pulse while frame (b) shows both the first and second pulse. The dashed line shows the decay of the output if no mirrors would have been present.

sharpening of the output pulses can be given in terms of gain switching.

Usually, gain switching is achieved by modulating the gain (or loss) in a time short compared to the spontaneous lifetime of the laser medium. After such a switch, the intracavity intensity builds up by amplified spontaneous emission. The subsequent depletion of the gain brings the pulse to an end, resulting in a short output pulse. The dynamics of the gain thus lie at the heart of the emission of such a short pulse. In our laser, the light experiences not only a variation of the gain in the temporal domain, but also in the spatial domain, because the laser medium is localized at the center of the cavity. This spatial variation causes the light to experience an additional modulation of the gain. On the time scale of the spontaneous lifetime, the light thus *repetitively* experiences gain switching, although the medium is only excited once.

In order to stress the importance of the spatial distribution of the inversion in the cavity, we compare the calculated dynamics of Fig. 6(b) to a calculation of the pulse evolution under excitation with a four-times wider pump spot inside the same laser cavity (with the same number of excited species inside). Figure 8 shows the laser output for both excitation spots. The upper frame shows the results of Fig. 6(b), while the output for the four-times larger pump spot is presented in the lower frame. For the larger pump spot, the output clearly exhibits a much weaker modulation than for the smaller pump spot, yet the overall shape is the same. This shows that the spatial distribution of the gain has a major impact on the detailed temporal output of our laser. The regime of normal gain switching can be found by filling the cavity uniformly with gain. In that case, the modulation disappears almost completely [27], but the overall shape is conserved. This shows that the pulse-train *envelope* of our laser represents an example of normal gain switching.

V. HEURISTIC DESCRIPTION

An heuristic description of the pulse train is obtained by analyzing the light intensity at the output coupler during the first round trip after the pump pulse, as shown in Fig. 9, an enlargement of Fig. 6(b). In the left frame, one sees the first pulse that is coupled out of the cavity, containing the light that, immediately after excitation, propagates in the direction of the output coupler. During the rising slope of the pulse, the signal grows because an ever larger part of the gain medium contributes to the light intensity at the output coupler. This continues until the full length of the gain medium contributes. Beyond that point, the signal starts to decrease because the inversion is decaying over the full length of the gain medium. The dashed line shows how the light output would continue to decay if no mirrors would be present. Due to the mirrors, however, additional light arrives at the output coupler after some time, light that originally was emitted towards the high reflector. Because of the delay between the arrival of the retroreflected light and the roll-over of the light that arrived first at the output coupler, the output signal will exhibit a small dip. This dip occurs because the cavity is not completely filled with gain, the photoluminescence lifetime is finite, and because the pump pulse is so short [28].

The small dip is responsible for the development of the strongly modulated output that appears after some round trips through the cavity. This is illustrated by the results shown in the right frame of Fig. 9, where the first and second pulse are shown together [note the different scale as compared to frame (a)]. From the height difference between the first and second peaks, one sees that the light has experienced gain during its second pass through the gain medium. One also sees that the dip following the second pulse is considerably deeper than the first dip. This can be understood when we see the second pulse as a nonlinearly amplified version of the first pulse: With a decaying inversion, the front of the light traveling through the gain medium experiences more amplification than the tail. As a result, a sharpened peak is emitted [26].

The emission of the subsequent peaks is now easily described since we have entered the regime where stimulated emission dominates. The evolution of the pulse train is now very similar to that in a regenerative amplifier [30] with the first (and second) peak acting as a seed. Repetitive nonlinear amplification results in the strongly modulated output. Note that the spontaneous emission lifetime (0.86 ns) exceeds the cavity round-trip time (0.17 ns), a necessary requirement for having gain during several round trips.

As discussed earlier, there are two essentially separate but interleaved pulse trains that emerge from the cavity resulting in a pulse repetition rate twice the longitudinal mode spacing [31]. Obviously, the pulse trains experience coupling since they feed from the same gain. Except from this fact, these pulse trains are largely independent. This becomes clear when, in the calculation, one moves the gain medium along the cavity axis, to a position somewhat closer to the high reflector than to the output coupler (distance ratio 9:11). In that situation the calculated pulse train shows the odd-even staggering as observed in the experiment. In terms of the two interleaved pulse trains, the odd-even staggering is very simply explained: The odd-numbered pulses have their origin in light that, directly after excitation, travels towards the output coupler, returning to the gain medium at a time 1.1 L/c. The even-numbered pulses, however, grow out of the light that, initially, moves towards the high reflector, making a second pass through the gain medium at a time 0.9 L/c. The time of second passage is thus different for the two light pulses. Since the gain continuously diminuishes, the pulse that arrives earliest will, at each round trip, experience a larger gain as compared to the other. The small asymmetry has already been included in the model presented above (Fig. 6), reproducing the considerable amplitude odd-even staggering of the pulses in Fig. 4. Figure 10 serves to illustrate the asymmetry by marking the time of passage of the pulses through the gain medium during subsequent round trips through the cavity. Also indicated in Fig. 10 is the evolution of the gain for the case that gain depletion does not come into play [cf. Figs. 6(a,b)]. The gain asymmetry will give rise to a substantial asymmetry in the pulse heights of the two trains since the amplification depends exponentially on the gain.

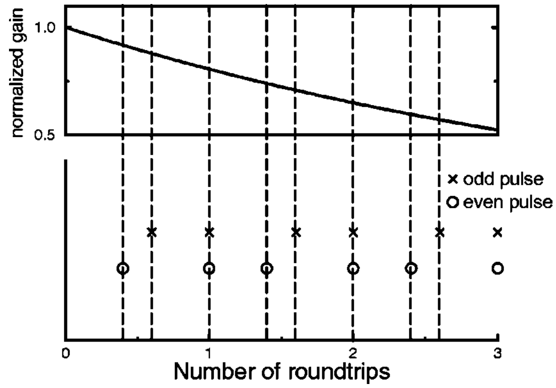

FIG. 10. Visualization of the moments of passage through the gain medium for the odd (cross) and even (circle) pulses. Because of the asymmetric position of the gain medium inside the cavity, the odd and even pulses do not always pass through the gain medium at exactly the same moment. Each $(m+\frac{1}{2})$ round trip $(m=0,1,2\ldots,)$ the even pulse passes the gain medium a bit earlier than the odd pulse. Since the gain is continuously decaying (as shown by the curve in the upper half of the figure), the even pulse experiences more gain than the odd pulse during this passage. This effect gives rise to the odd-even staggering of the pulse heights.

VI. CONCLUSION

We have performed a detailed experimental time-domain study of the output of a short-cavity polymer laser upon excitation by a single femtosecond pump pulse. In the experiment, the gain medium only partially fills the cavity and the duration of the pump pulse is very much shorter than the cavity round-trip time, which, in turn, is shorter than the sponteneous emission lifetime of the gain medium. Although the gain medium is excited only once, a whole train of pulses is emitted. This behavior occurs because the spatial distribution of the gain medium makes the light to experience repetitive gain switching. We show that our results can be well reproduced by a one-dimensional rate-equation model that takes the nonuniformity of the cavity explicitly into account. This model exhibits the buildup and decay of the pulse train, the odd-even staggering of the pulses when the gain medium is positioned somewhat asymmetrically in the laser cavity, and allows us to calculate the decay of the inversion. We find excellent agreement with the experiment.

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[1] J. AuYeung, Appl. Phys. Lett. **38**, 308 (1981).
[2] S. Schuster and H. Haug, J. Opt. Soc. Am. B **13**, 1605 (1996).
[3] P.F. Moulton, J. Opt. Soc. Am. B **3**, 125 (1986).
[4] J.M. Eggleston, L.G. DeShazer, and K.W. Kangas, IEEE J. Quantum Electron. **24**, 1009 (1988).
[5] A. Deleva, Z.Y. Peshev, Z.I. Aneva, B.K. Kaprielov, E. Vidolova-Angelova, and D. Angelov, J. Mod. Opt. **47**, 793 (2000).
[6] L.W. Casperson, J. Appl. Phys. **47**, 4555 (1976).
[7] J. C. Diels and W. Rudolph, *Ultrashort Laser Pulse Phenomena* (Acadamic Press, San Diego, 1996).
[8] G.H.C. New, Rep. Prog. Phys. **46**, 877 (1983).
[9] S.A. van den Berg, R.H. van Schoonderwoerd den Bezemer, H.F.M. Schoo, G.W. 't Hooft, and E.R. Eliel, Opt. Lett. **24**, 1847 (1999).
[10] D. Moses, Appl. Phys. Lett. **60**, 3215 (1992).
[11] N. Tessler, G.J. Denton, and R.H. Friend, Nature (London) **382**, 695 (1996).
[12] U. Lemmer, A. Haugeneder, C. Kallinger, and J. Feldmann, in *Semiconducting Polymers*, edited by G. Hadziioannou and P.

F. van Hutten (Wiley-VCH, Weinheim, 2000).
[13] G. Kranzelbinder and G. Leising, Rep. Prog. Phys. **63**, 729 (2000).
[14] H. Spreitzer, H. Becker, E. Kluge, H.S.W. Kreuder, R. Demandt, and H.F.M. Schoo, Adv. Mater. **10**, 1340 (1998).
[15] D.T. Schaafsma and D.H. Christensen, Phys. Rev. B **54**, 14 618 (1996).
[16] R. Ulbrich and M. Pilkuhn, Appl. Phys. Lett. **16**, 516 (1970).
[17] L. Csillag, M. Janossy, and T. Salamon, Phys. Lett. **26A**, 436 (1968).
[18] To obtain a good signal-to-noise ratio we average the signal over several seconds. As a check we compare these results with single-shot measurements and find no significant differences.
[19] J. Klebniczki, Z. Bor, and G. Szabó, Appl. Phys. B: Photophys. Laser Chem. **46**, 151 (1988).
[20] U. Ganiel, A. Hardy, G. Neumann, and D. Treves, IEEE J. Quantum Electron. **11**, 881 (1975).
[21] M. Azadeh and L.W. Casperson, J. Appl. Phys. **83**, 2399 (1998).
[22] M. Mack, Appl. Phys. Lett. **15**, 166 (1969).
[23] C. Lin, T.K. Gustafson, and A. Dienes, Opt. Commun. **8**, 210 (1973).
[24] J. Hebling and J. Kuhl, Opt. Lett. **14**, 278 (1989).
[25] W.A. Hamel and J.P. Woerdman, Phys. Rev. A **40**, 2785 (1989).
[26] A. E. Siegman, *Lasers* (University Science Books, Sausalito, California, 1986).
[27] Note that the ouput of the laser is also slightly modulated when the gain is homogeneously distributed over the cavity. In that limit, the modulation is the result of the high losses that are localized at the mirrors, inducing an inhomogeneous photon distribution in the cavity (Refs. [6,25]).
[28] It is easily seen that the two light signals will not be distinguishable when the cavity is completely filled. In that case, no ''pulse front'' can be assigned and the photon density in the cavity is essentially homogeneous. The same argument applies when the pump pulse has a duration larger or equal to half the cavity round-trip time (Ref. [29]).
[29] D.J. Bradley and A. Durrant, Phys. Lett. A **27**, 73 (1968).
[30] Y.Y. Hwang, C.H. Lee, and J. Wang, Appl. Opt. **36**, 7802 (1997).
[31] Double or multiple pulsing is also known to occur in mode-locked lasers (Refs. [32,33]), the origin of the multiple pulsing being different in that case.
[32] M. Lai, J. Nicholson, and W. Rudolph, Opt. Commun. **142**, 45 (1997).
[33] M.F.H. Tarroja, M. Sharafi, and L.W. Casperson, J. Opt. Soc. Am. B **6**, 1564 (1989).