Appl. Phys. B 38, 233–240 (1985)

Applied Physics B Photophysics and Laser Chemistry

Characteristics of a Hybridly Mode-Locked cw Dye Laser

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Received 17 July 1985/Accepted 6 August 1985

Abstract. We present experimental and theoretical results on the characteristics and random variations of subpicosecond pulses generated by a synchronously pumped cw dye laser with saturable absorber. The analysis of the power spectra indicates rapid fluctuations of the pulse duration of 40–50%, energy fluctuations of 3%, and a jitter of the repetition time of 0.1% corresponding to an absolute jitter of 12 ps. The latter is caused mainly by the temporal jitter of the pump laser. A mismatch of the lengths of the dye and the pump-laser cavity can result in a nonstationary mode-locking regime with a periodic change of the pulse parameters. The interpretation of the experimental results are supported by computer simulations of the pulse evolution process.

PACS: 42.55.M, 42.55.B, 42.65.G

In the recent years synchronously pumped dye lasers have been widely used for picosecond spectroscopy. Synchronous mode-locking provides the advantage of broad spectral tunability. The method is readely applicable to a number of combinations of pump and dye lasers (for a comprehensive review, see e.g. [1]). The synchronization of the pulses with respect to the external rf signal that drives the acousto-optic mode locker of the pump laser is an additional attractive feature, which offers, e.g. the possibility of dual systems, capable of providing two synchronized pulse trains [2–4], or the possibility of synchronized amplification of the pulses [5].

However, the minimum pulse duration that can be achieved with typical synchronously mode-locked dye lasers is a few picoseconds, whereas pulse durations in the subpicosecond range can be generated with passive mode-locking [6, 7]. Combination of both techniques (hybrid mode-locking) results in subpicosecond pulses [8–10]. The advantage of synchronousness is maintained, but the broad spectral tunability is largely lost.

How well the pulses are actually synchronized with respect to the rf signal is obviously an important question. Furthermore, for many spectroscopic applications, in addition to the average pulse parameters, the knowledge of random fluctuations of the laser output is essential. In particular, it is important to know fluctuations of the pulse duration because they could obscure the time resolution of the experiments. For example, it is known [11] that the second harmonic autocorrelation measurements of a train of pulses with varying duration may lead to a misjudgement of the average pulse duration.

We have investigated the properties of the pulses generated by a hybridly mode-locked cw dye laser, focussing our attention not only on the average pulse parameters, but also on random properties such as the fluctuations of the pulse energy, the pulse duration and the repetition time. The latter gives a measure of the temporal accuracy with which the pulse train can be synchronized to an external signal.

Numerical results of computer simulations of the pulse generation process based on a rate equation model are compared with some of the experimental results.

1. Experimental

The laser system used in our experiments is similar to that described in [12]. The argon-ion laser is mode-locked with the use of an acousto-optic modulator driven by a synthesized rf source with a frequency stability better than 10 Hz. To avoid thermal runaway of the modulator the acoustic resonances are locked to the synthesizer frequency with the use of an active feedback control system [13]. The argon laser pulses were measured to be 120 ps full width at half maximum (FWHM). The dye laser consisting of a standard three-mirror-folded cavity is pumped with 0.5–0.6 W average power at 515 nm. A thermal self-compensating resonator structure is used to minimize temperature-induced changes of the cavity length. Mirror vibrations and fluctuations of the dye jet are also carefully minimized to ensure good dynamical stability of the optical cavity length. Frequency tuning is done with a dielectric interference filter (wedge filter) with a bandwidth of 150 THz. The dye solution is a mixture of 3.7×10^{-3} mol/l rhodamine 6G and 3.5×10^{-5} mol/l DQOCI.

The average power of the dye laser pulse train is about 50 mW at a pump level of 500 mW. Inserting the wedge filter for wavelength tuning reduces the average power to about 30 mW.

The output coupler is a small-bandwidth, dielectric interference filter with a transmission maximum at 635 nm. The wavelength range of 615–625 nm of best mode-locking is on the short-wavelength wing of the transmission characteristic. In this range the reflectivity of the interference filter is 95–97%.

The autocorrelation function of the pulses was measured by background-free second-harmonic generation in a KDP crystal. To characterize the noise of the laser output the power spectrum of the intensity fluctuations has been measured. The signal of a fast photodiode is processed by an electronic spectrum analyzer which covers a frequency range from 0 to 1.5 GHz. The large dynamic range of about 10^{12} and the high spectral resolution of 10 Hz permits very accurate measurements of the noise.

The power spectra allow to identify three basic types of random output fluctuations: (i) fluctuations of the pulse energy, (ii) random variations of the repetition time (temporal jitter), and (iii) fluctuations of the pulse width. The letter is obtained by measuring the power spectrum of the second harmonic of the pulse train. Second-harmonic generation transforms changes of the pulse width of the fundamental into energy fluctuations of the second harmonic. The evaluation of these quantities from the measured power spectra will be presented elsewhere [14].

2. Model Calculations

To gain a deeper insight of the pulse evolution and the steady state pulse properties in a hybrid mode-locked dye laser we have used computer simulations based on a system of rate equations similar to that presented in [15]. However, the model has been extended to include the saturable absorber, and also the effect of spontaneous emission of the laser medium, i.e., the amplifier dye. It is essential to include spontaneous emission because there is a surprisingly pronounced effect on the pulse evolution as well as the stationary properties.

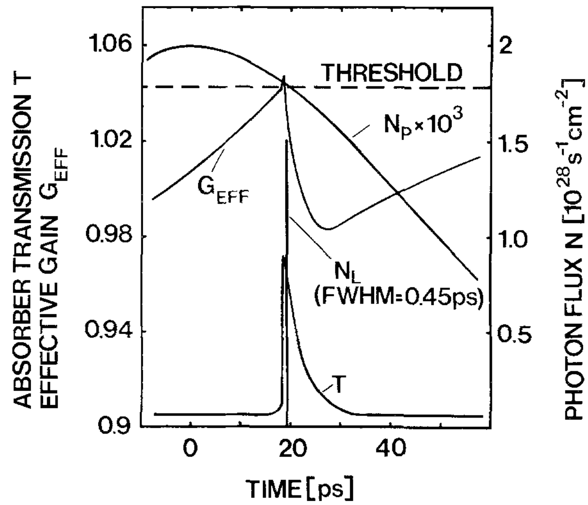

Fig. 1. Temporal behavior of the effective gain of the amplifier-absorber dye mixture, of the absorber transmission and of the photon flux N_L of the dye laser pulse during the passage of the pump pulse with the photon flux N_p. The parameters used for the numerical calculation are: peak photon flux of the Gaussian pump pulse 2.1 $10^{25}\,s^{-1}\,cm^{-2}$, pump pulse duration (FWHM) 100 ps, bandwidth of the frequency filter 200 THz, linear cavity losses 0.04, small signal absorption 0.1, relaxation time of the absorber 3 ps, ratio of the cross sections of the absorber and amplifier 1.8 and difference of the lengths of the pump and dye laser cavity 3.3 μm

The fundamentals of the pulse shaping mechanism in a hybrid laser are illustrated by Fig. 1, which shows the temporal variation of the following quantities for a typical set of laser parameters: (i) the pump pulse N_p, (ii) the effective gain G_{eff}, (iii) the transmission T of the saturable absorber, and finally (iv) the dye laser pulse N_L which because of its subpicosecond duration is represented by a straight vertical line on the time scale of the figure.

The following features of the computer simulations are noteworthy: The effective gain increases rather smoothly during the time before the threshold is reached. Once the gain exceeds the threshold, however, there is a very sharp increase of the effective gain reflecting the bleaching of the saturable absorber. As a result the laser intensity grows very rapidly, causing the gain medium to saturate within a fraction of a picosecond. The principal result of the combined gain and absorption dynamics is that the effective gain exceeds the cavity losses for just a small fraction of a picosecond, and stays below threshold for as long as several times 10^{-11} s. Thus a subpicosecond laser pulse is obtained, and the formation of a satellite pulse typical of ordinary synchronously mode-locked dye lasers is suppressed.

Another interesting feature of the hybrid system is the fact that the pulse evolution time is much longer than in a dye laser without saturable absorber. About 4000 to 5000 cavity round trips are required for reaching stationary conditions, i.e., the pulse evolution time ranges from 40 to 50 μs, more than an order of magnitude greater than the evolution time of the ordinary synchronously pumped dye laser. This property is important as regards the response of the hybrid system to pump pulse fluctuations (Sect. 3.2).

3. Results

3.1. Energy Fluctuations

The fluctuations of the acousto-optically mode-locked argon-ion laser has been characterized in detail by Kluge et al. [12]. The fluctuations of the argon laser pumping the hybrid dye laser corresponds to the conditions described in that paper.

An example of the noise of the hybrid laser is shown in Fig. 2, which depicts a 1 MHz section of a typical spectrum with the first order component at $\nu_1 = 79.801$ MHz. At the bottom of the sharp spike a strong broad noise band can be distinguished with a width of 400 kHz (FWHM). The power ratio of the maxima of the noise band and the spike is found to be constant for all frequency components of the spectrum, and therefore the 400 kHz noise band is identified to be caused by energy fluctuations. A comparison with the power spectrum of the argon laser [12] indicates that the energy noise is due to intrinsic fluctuations of the dye laser rather than being induced by fluctuations of the pump. From the estimate of the spectral area a r.m.s. value of the energy fluctuations of $\Delta E_p/E_p = 3\%$ is obtained. The 400 kHz frequency width gives a correlation time of 0.4 μs. The observed fluctuations of the pulse energy are slightly stronger than those of the synchronously pumped dye laser without absorber ($\Delta E_p/E_p = 1.5\%$ [12]).

3.2. Temporal Jitter

When the spectrum analyzer is operated with higher resolution a second noise feature with a frequency width $\Delta\nu = 300$ Hz can be resolved (Fig. 3). The ν^2-dependence of this band is clear evidence of a distinct temporal jitter of the dye-laser pulses. The r.m.s. value $\Delta T/T = 0.1\%$ corresponds to an absolute jitter of $\Delta T = 12$ ps with a correlation time of 0.5 ms. As the pump laser shows temporal jitter [12] with very similar features the observed dye laser jitter is probably

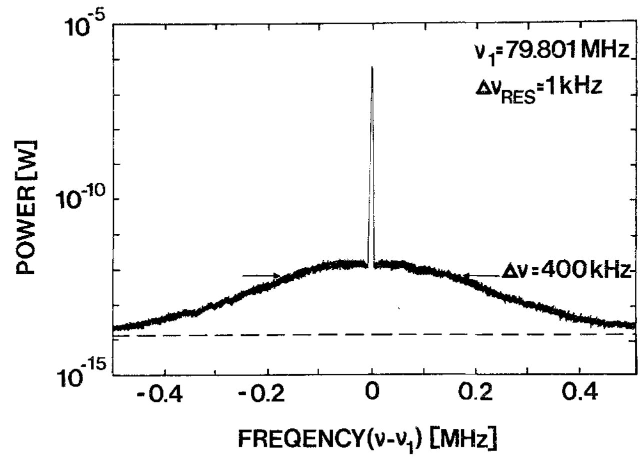

Fig. 2. Section of the power spectrum of the first frequency component with a resolution bandwidth of $\Delta\nu_{\mathrm{Res}} = 1$ kHz. The dashed line denotes the instrumental noise level

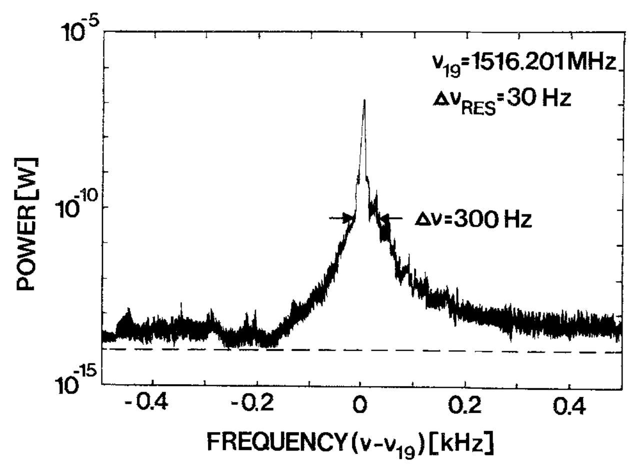

Fig. 3. Section of the power spectrum of the 19-th frequency component

caused by the pump source. However, whereas in the case of a purely synchronously pumped dye laser [12] the temporal jitter of the pump and dye laser pulse are strictly correlated, only a partial correlation exists in the hybrid laser. This is evident from cross-correlation measurements between the pulses of two hybridly mode-locked dye lasers pumped by the same argon laser. The relative jitter of 10 ps between the two dye lasers deduced from the cross-correlation measurements would correspond to an absolute jitter of 7 ps, if the temporal jitter of both dye lasers is assumed to be uncorrelated. The comparison with the larger jitter value of 12 ps obtained by means of the spectrum analyzer indicates that the temporal jitter between the two dye lasers and, consequently, between the dye lasers and the argon laser is only partially correlated.

The explanation of the partial correlation of the jitter in the hybrid laser versus the full correlation in the or-

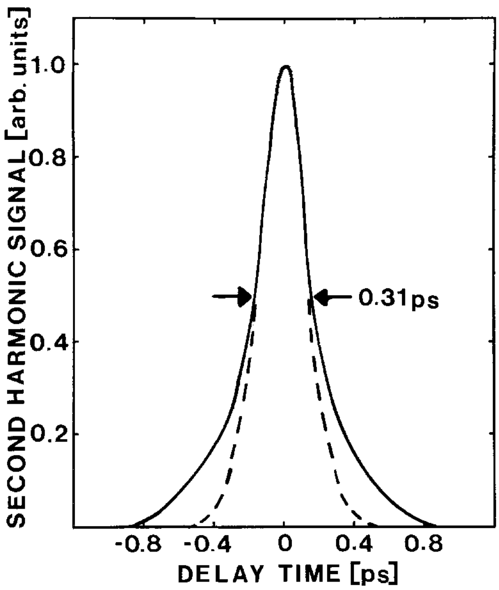

Fig. 4. Autocorrelation trace of the pulse train (solid line) with a width (FWHM) of 0.31 ps. The dashed line represents the autocorrelation function of a sech^2-shaped pulse with the same width

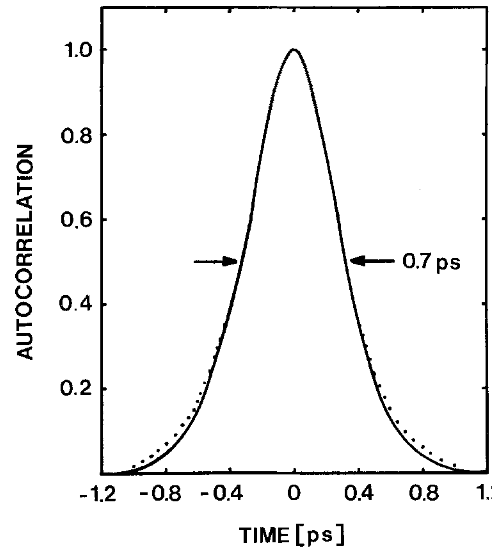

Fig. 5. Autocorrelation function of a steady state dye laser pulse obtained by the numerical calculations with a slope of the wings similar to the experimental curve of Fig. 4, but with a width of 0.7 ps. The parameter used are the same as for Fig. 1. The dots represent the autocorrelation function of a sech^2-shaped pulse with the same width

dinary synchronously pumped laser without absorber could be the following. Computer calculations and also direct experimental observation show that the characteristic pulse evolution time of the hybrid laser is much larger: it takes about 4000 to 5000 round trips for the pulses to become stationary, whereas in the dye laser without saturable absorber it takes 200 to 300 passes [16] to adjust to changes of the pump pulses. We believe that the slower response of the hybrid laser prevents the establishment of full correlations of the temporal jitter.

3.3. Pulse Duration

When the laser is run with the interference filter as output coupler the duration of the pulses is significantly shorter than that obtained with an ordinary output mirror for which the lasing wavelength is close to the central wavelength of the reflectivity band. Such a behavior could be explained as being due to a self-phase modulation of the pulses which can arise from the nonlinear interaction with the dye medium. Self-phase modulation occurs, for example, if the lasing wavelength differs from the center wavelengths of the absorption or emission bands of the dyes [17]. To obtain pulses with shortest durations it is necessary to compensate the phase modulation of the pulse (chirp) by means of elements with suitable group velocity dispersion [18]. One possibility is to use the group velocity dispersion of a dielectric mirror. The dispersive effects of the mirror are expected to be strongest at the edges of the reflectivity band [19, 20]. Thus we interpret our observation, that the shortest pulses are obtained with the interference filter, as being due to a compensation of the pulse chirp by the group velocity dispersion of the interference filter. Similar observations have been made by others, who used the negative group-velocity dispersion of a four-prism sequence [21]. A more detailed description of the pulse chirp of a hybridly mode-locked dye laser will be given elsewhere [22].

An example of an autocorrelation trace of the pulses of the hybrid laser is depicted in Fig. 4 (solid curve). In this case the dye laser was operated without a frequency tuning element, and the full width at half maximum (FWHM) is 310 fs. The width and shape of the autocorrelations changes drastically with the cavity length, the lasing wavelength (Sect. 3.4), and with the absorber concentration. If the concentration is decreased the autocorrelation traces become broader and show a more or less pronounced coherence spike indicating random substructures of the pulse.

The autocorrelation width gives a pulse duration of 210 fs if a sech^2-shaped pulse is assumed. However, the comparison with the autocorrelation of a sech^2-pulse (dashed curve in Fig. 4) shows that the experimental curve exhibits much broader wings. Also for comparison, an autocorrelation function calculated from our computer simulation of the hybrid laser is shown in Fig. 5. It turns out that the calculated autocorrelation can be approximated fairly well by an autocorrelation of a sech^2-pulse, although the actual calculated pulse shape is slightly asymmetric and not a $sech^2$-pulse. The wings of the experimental and the calculated autocorrelation fit rather well, whereas the width of the theoretical curve is 700 fs, much greater than the measured autocorrelation half width.

It has been shown [11] that one way to explain the inconsistency between the measured half width and wings of the autocorrelation function is to assume a statistical variation of the pulse duration. Following this concept, we have fitted the experimental data with an averaged autocorrelation obtained by averaging over an exponential probability distribution of pulse durations. In Fig. 6 we compare the autocorrelation averaged over an exponential distribution of $sech^2$-pulses (solid line) with the experimental data (dotted line). It is seen that a fairly good agreement is obtained. The average pulse duration of the statistical model is 260 fs, slightly greater than the value of 210 fs deduced from the measured autocorrelation, and the r.m.s. deviation is 55%.

The statistical interpretation of the experimental autocorrelation functions invoking fluctuations of the pulse duration is supported by the power spectra of the second harmonic of the pulse train. An example is depicted in Fig. 7. The comparison with the corresponding spectrum of the fundamental (Fig. 2) reveals a drastic increase of the noise. From the power spectrum we find that the r.m.s. value of the fluctuations of the second harmonic energy is 40–50% compared with a value of 6% that would be expected as a result of the energy noise of the fundamental. One is lead to conclude that substantial fluctuations of the pulse durations occur. If we neglect the small effect of the fluctuations of the fundamental the r.m.s. value of the pulse duration fluctuations is directly given by the fluctuations of the second harmonic energy, i.e., $\Delta t_p/t_p = 40$–50%.

This value agrees well with the r.m.s. deviation of 55% which has been used for the fit of the autocorrelation data using the statistical model.

We believe that these fluctuations of the pulse duration are typical for the hybrid dye laser with a dye mixture and a linear cavity. For instance, a closer inspection of the autocorrelation trace of the hybrid dye laser depicted in [23] with a rhodamine 101-rhodmine B mixture as amplifier-absorber combination reveals similar broad, exponentially shaped wings of the autocorrelations.

A fully satisfactory explanation of the observed fluctuations of the pulse duration is not available as yet. Nevertheless, our experimental observation suggest a plausible interpretation. For example, when the concentration of the saturable absorber is decreased, one obtains autocorrelation functions with a pronounced central spike – sometimes called coherence spike – on

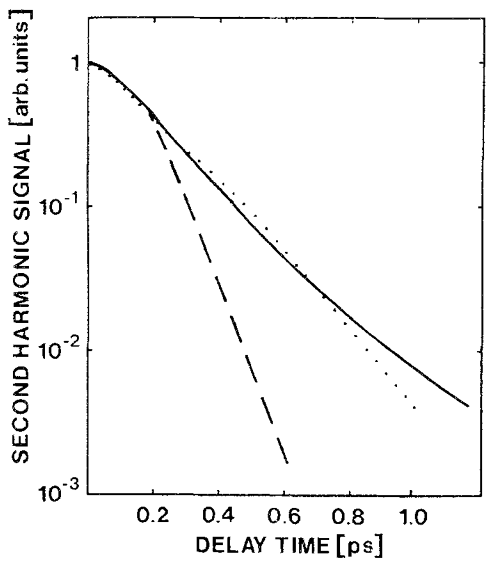

Fig. 6. Logarithmic plot of the fit of the autocorrelation function of a $sech^2$-shaped pulse averaged with an exponential distribution of the pulse durations (solid line) to the experimental trace of Fig. 3 (dotted curve). The dashed line shows the autocorrelation of a single $sech^2$-shaped pulse

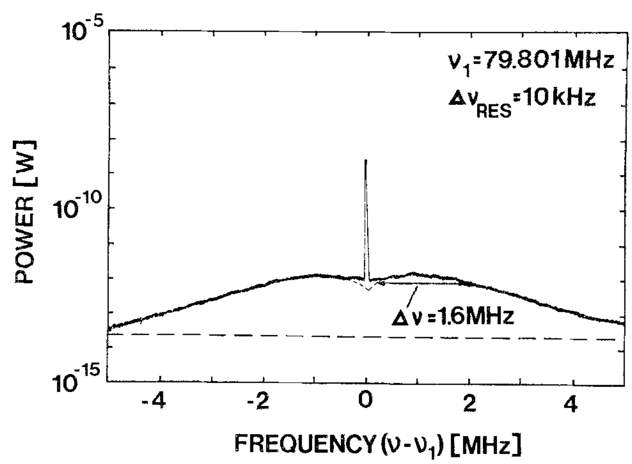

Fig. 7. Section of the power spectrum of the first frequency component of the second harmonic of the pulse train

top of some broad feature with broadened wings. This type of autocorrelation function which is characteristic of a random substructure of the pulses is typically observed in ordinary synchronously pumped dye lasers [24]. On the other hand, substructure-type autocorrelations are not observed in synchronously pumped dye lasers operating in a colliding pulse mode with a ring laser configuration having separate absorber and amplifier dyes [25]. In this case the autocorrelation functions of the pulses can be very well described with the assumption of a simple pulse shape, e.g. a $sech^2$-shaped pulse indicating that fluctuations of the pulse structure do not play a role. These observations suggest that effective saturable absorption is crucial for the complete elimination of the structural fluctuations.

In our case the laser operates with a mixture of absorber and amplifier dye rather than separate dyes, and it is not possible to adjust the saturation regimes independently by using different beam areas in the amplifier and absorber. We suggest that the fluctuations of the pulse duration can be regarded as remanents of the strongly fluctuating pulse substructure which is not fully eliminated in a linear laser configuration with common absorber and amplifier.

3.4. Cavity Length and Wavelength Dependence of the Pulse Durations

The pulse duration depends strongly on the matching of the cavity lengths of the pump and dye laser. From curve 2 of Fig. 8 it is evident that a stability of the cavity length better than 1 μm is required to maintain operation at the minimum pulse duration. This is a ten times better accuracy than that required for a synchronously pumped dye laser without absorber. Figures 9 and 10 show the autocorrelation traces of the pulses at a cavity mismatch $\Delta L = 0$ and $\Delta L = 3.5$ μm, respectively. These autocorrelation traces represent pulses with a greater duration than the example of Fig. 4, but the shapes of the autocorrelation traces are essentially identical. The longer pulse durations of the examples discussed in this subsection are due to the fact that these measurements were carried out on a dye laser with a tuning element in the cavity. With the filter broader pulses are obtained presumably because of bandwidth narrowing or additional group velocity dispersion caused by this element.

Two examples of the calculated variation of the pulse duration with cavity length are depicted in Fig. 8 (curves 1 and 3). The measured variation of the pulse duration with cavity length in this range (curve 2) is reasonably well described by the theoretical model. The dashed part of the calculated curves denotes the range where the pulses in the computer simulations do not reach a steady state but exhibit periodic changes.

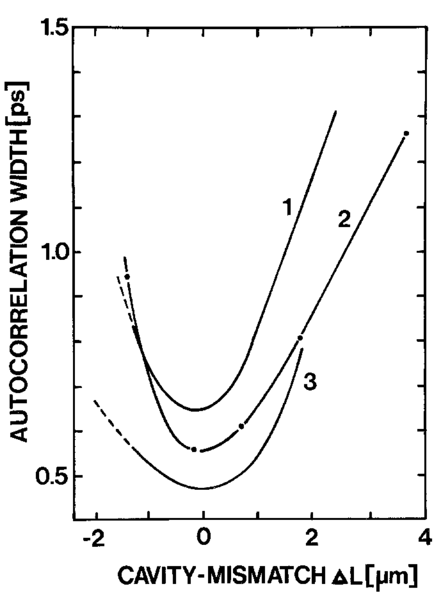

Fig. 8. Autocorrelation width (FWHM) vs cavity-mismatch ΔL. The origin $\Delta L = 0$ has been chosen arbitrarily. Curve 2 shows the experimental dependence curve 1 and 3 the numerically calculated one for a ratio of the saturation energies of 1.8 and 2.1, respectively. The dashed part of curves 1 and 3 denote the range where the pulse parameters show an oscillatory behavior

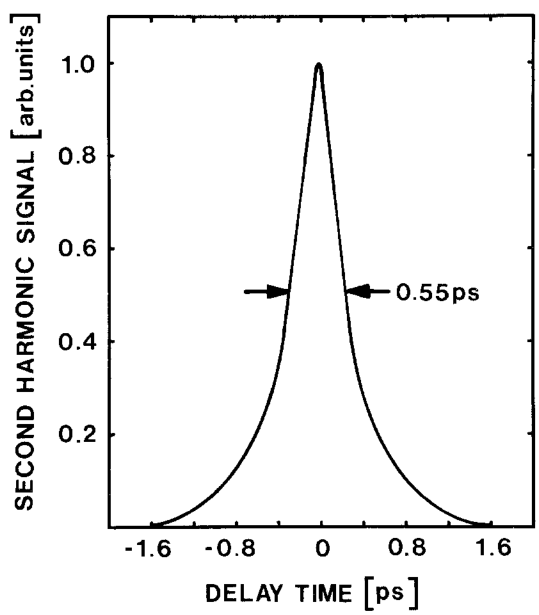

Fig. 9. Autocorrelation trace of the pulses for a cavity mismatch $\Delta L = 0$

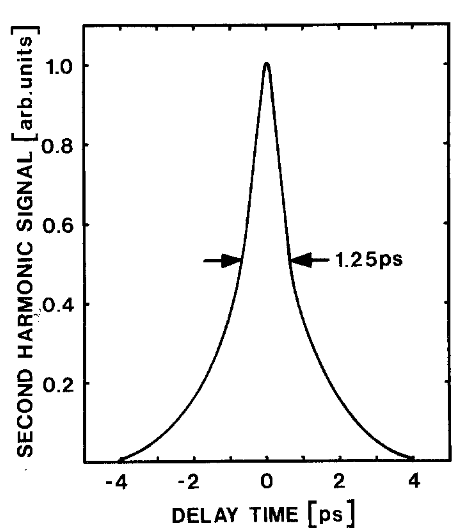

Fig. 10. Autocorrelation trace for a cavity mismatch $\Delta L = 3.5$ μm

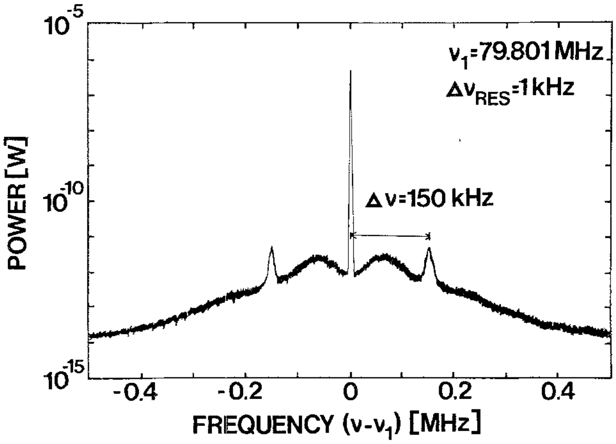

Fig. 11. Section of the power spectrum of the first frequency component of the pulse train from the dye laser when the cavity length is too short

Some experimental evidence supporting this observation has been obtained from the power spectra. Figure 11 depicts the power spectrum of the pulse train when the cavity is too short. Two sharp frequency peaks located symmetrically with respect to the central peak are seen. This indicates an oscillation of the pulse energy with a frequency given by the separation between the central and the side peaks. This oscillatory behavior of the pulses is believed to be due to the fact that for an excessively large cavity mismatch the pulse shaping by the amplifier and absorber cannot fully compensate the temporal shift of the dye-laser pulse with respect to the pump pulse. As a result, the interaction of the dye-laser pulse with the gain medium takes place at successively later times as more round trips are completed, and the dye laser pulse experiences decreasing gain. After some time a point is reached where the gain is not sufficiently depleted after the passage of the pulse. The gain remains above threshold and a second pulse can be generated. Thus, the first pulse dies out and the second pulse takes over. This process is then repeated with the new pulse. The repetition cycle depends on the cavity length mismatch, typical values being 5–10 µs. These values agree well with the repetition cycles found in the computer simulations.

The measured variation of the autocorrelation width with the wavelength are shown by curve 1 in Fig. 12. The wavelength was tuned by means of a wedge filter. The minimum duration occurs at $\lambda = 620$ nm, whereas at $\lambda = 615$ nm the pulse width is much greater.

Because of the wavelength dependence of the absorption and emission cross sections, σ_a and σ_e, a change of the wavelength is equivalent to a change of the ratio of the saturation energies of the saturable absorber and the amplifier dye. For comparison, curve 2 in Fig. 12 represents the numerically calculated dependence of

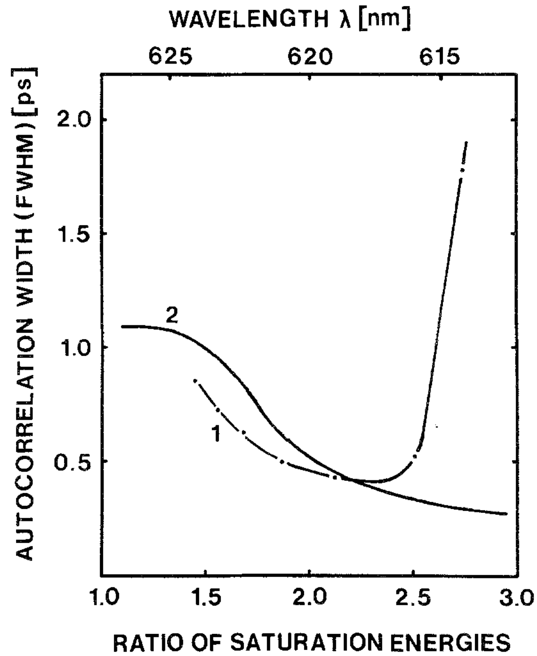

Fig. 12. Wavelength-dependence of the autocorrelation widths (curve 1) and the theoretical dependence on the ratio of the saturation energies σ_a/σ_e (curve 2). The relation between the wavelength and the ratio of the saturation energy has been obtained from the wavelength dependence of the emission cross-section σ_e of rhodamine 6G and of the absorption cross-section σ_a of DQOCI

the pulse duration on the ratio of the saturation energies. It is obvious that in the range $1 \leqq s = \sigma_e/\sigma_a \leqq 3$ the pulse width decreases with increasing s but for $s > 3$ no further pulse shortening is expected.

This disagreement between the experimental and the calculated variation of the pulse duration is expected to be due to the fact that the theoretical model neglects (i) the phase modulation of the pulse which may occur both in the absorber and the amplifier dye, and (ii) the group velocity dispersion of the cavity.

4. Conclusions

From measurements of the autocorrelation functions and the power spectrum of the laser intensity we have obtained detailed information about fluctuations of the pulses generated in a synchronously pumped dye laser system with a saturable absorber. The numerical simulation has enabled us to interprete some of the experimentally observed properties of the pulses.

With a mixture of rhodamine 6G and DQOCI we obtain pulses with a mean duration of 0.26 ps, when no bandwidth-limiting frequency filter is used. The energy fluctuations are 2–3% (r.m.s.) with a correlation time of about 0.4 µs. The temporal jitter is 0.1% (r.m.s.) which corresponds to about 12 ps on an absolute time scale.

The temporal jitter is caused by the jitter of the pump laser and has a correlation time of 0.5 ms. Cross-correlation measurements between the pulses of two hybrid dye lasers pumped by the same argon-ion laser have indicated that the temporal jitter between the dye and pump laser is only partially correlated.
From a detailed analysis of the autocorrelation traces and from the power spectra of the second harmonic strong fluctuations of the pulse duration of 40–50% (r.m.s.) have been established. A consequence of this result is that the actual average pulse duration is significantly longer than the duration deduced from the width of the autocorrelation trace.
An interesting experimentally observed feature which is confirmed by the computer simulations is the periodic change of the pulse properties when the cavity length of the dye laser falls short of a certain lower limit. This indicates a periodic process where one pulse dies out and, simultaneously, a new pulse grows up.

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