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On Noise and Fluctuations in a Synchronously Mode-Locked cw Laser System

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Abstract. A single-shot and synchronously-scanned streak camera, autocorrelation and noise spectrum analysing techniques are utilized to study the output characteristics of synchronously mode-locked cw lasers. Four main conclusions are drawn: (i) the pulse train from a synchronously-pumped dye laser reveals, besides phase jitter, considerable pulse shape fluctuations; (ii) autocorrelation measurements may be highly misleading when actual pulse shapes are considered; (iii) both the phase jitter and pulse shape fluctuations of the dye laser output are caused by the phase fluctuations of the pumping ion laser pulse train; (iv) the phase jitter of the ion laser proceeds from the fluctuations in the cavity round-trip time with a characteristic time of about 5 μs. Under optimum conditions the rms noise of the dye laser output was 2% and the phase jitter with respect to the rf sine drive of the acousto-optical mode-locker, ~30 ps. A qualitative explanation fo the noise properties is given.

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The synchronously mode-locked cw dye laser has emerged as a versatile source of ultrashort light pulses for transient spectroscopy. As compared to other picosecond light sources it affords high average power, a wide tuning range, a sufficiently short pulse duration and a moderate peak power. The repetitive nature of the mode-locked output enables powerful methods of signal integrating and averaging to be involved, provided the lasing process is stable enough. With this in mind the stability requirements have been considered [1–4] and a number of stabilization schemes have been proposed [1, 5–7], which demonstrate different success in noise reduction. The discussions and proposed solutions usually rely on the dye laser pulse width determined from an autocorrelation measurement. However, the latter is a highly averaged quantity and may lead to serious errors in interpreting actual pulse shapes [8]. Noise properties, that show up in an autocorrelation function, have been discussed [9–13] and a conclusion about the inherent substructure of dye laser pulses has been reached. Another series of experiments of importance here has based on the synchroscan streak camera [14, 15] measurements showing a considerable temporal jitter of the synchronously mode-locked dye laser pulse train with respect to the locking frequency of the pumping ion laser. This has forced one to synchronize the streak camera directly to the pulse train to better the time resolution. However, the result (~10 ps [16, 17])[1] still greatly exceeds the actual pulsewidth of ~1 ps, as determined from an autocorrelation or a single-shot streak camera measurement. This is in a remarkable contrast with the results of the measurements using passively mode-locked lasers [19]. Hence, the elucidation the noise properties of an actively mode-locked cw laser is important for further progress in various synchroscan streak camera applications.

In this paper[2], we report on some characteristics of a synchronously-pumped mode-locked cw laser system, taking advantage of direct single-shot and synchronously-scanned streak camera measurements

[1] Only recently a certain success in long-term synchronization was achieved, showing an apparatus function of 6 ps FWHM [18]

[2] Preliminary reports on the subject were delivered on the 2nd Soviet-France Symposium on Construction of Optical Apparatus [10] and on the "Lasers-81" Conference [20]

as compared with second-order autocorrelation and some other indirect noise analyzing techniques. Our main attention is focused on the phase jitter of the pulse train and the pulse shape fluctuations. Independently, the same kind of problems were recently examined in [21] by measuring the power spectrum of the laser intensity. Some of the important qualitative results coincide in the two papers.

1. Experimental

The laser system consists of an acousto-optically mode-locked Kr^+ laser (Spectra-Physics Mod. 171-01, 4.5% output coupler), and an Oxazine I dye laser (SP Mod. 375, 16% output coupler) with an extended cavity to achieve synchronous pumping. A two-plate birefringent filter or a wedged etalon served as a tuning element. The mode-locker (SP Mod. 342) was driven by an amplified rf output of a stable (better than 10^{-7}) frequency synthesizer (Soviet G4-73). The typical output characteristics of the system were the following: locking frequency 2×40.87 MHz; average output power of the Kr^+ laser 700 mW ($\lambda = 674.1$ nm, tube current $25 \div 35$ A), pulse duration <80 ps; average output power of the dye laser 150 mW, pulse duration 3 ps (with a two-plate Lyot' filter in the cavity).

The single-shot streak camera measurements were performed on a commercially available Soviet "Agat-SF" camera with a time resolution (FWHM) of $\leqq 8$ ps. The synchroscan system [17] was based on a Soviet UMI-93M image converter tube driven by an amplified and frequency-doubled reference signal from the mode-locker driver. Data recording from the streak camera output screen was performed with an optical multichannel analyzer (OSA-500 with a SIT vidicon) operating on-line with an EC 1010 computer for data processing. To achieve short integration times the laser beam was reflected by a rotating mirror across a slit in front of the streak camera.

Autocorrelation traces were run on a non-collinear SHG autocorrelator with angle-tuned $LiIO_3$ crystal. The noise level of the laser output (and its second harmonic) intensity was measured by using a fast (ns response time) photodiode connected to dc or ac (sensitive in a broad frequency range of 10 Hz to 5 MHz) voltmeters. The noise was characterized by the ratio of the rms ac and dc signals.

2. Results

2.1. The Dye Laser

A characteristic set of synchronously-scanned streak camera traces of the dye laser output, depending on the time during which recurrent pulses are integrated on the streak camera screen, is given in Fig. 1. From the upper curve a long-term temporal jitter (random variations of the pulse repetition time) of <80 ps with respect to the rf drive of the mode-locker is evident in this particular case. The pump pulses were <80 ps long and showed no satellites, nevertheless the jitter could not be reduced by the dye laser adjustments only[3]. Shorter integration times reduced the jitter and allowed one to establish the time scale during which the pulse phase is almost fixed (the phase-jitter correlation time), of about 5 μs, corresponding to ~400 cavity round-trips. The latter is slightly more than the estimated pulse build-up time in a synchronously-pumped dye laser [22].

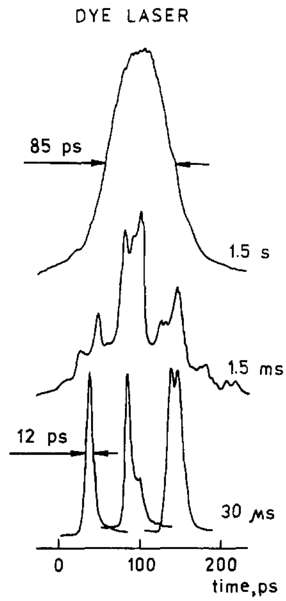

Fig. 1. Synchroscan-streak-camera traces of the dye laser output depending on the integration time. In case of a 30 μs integration time different traces are given to demonstrate that even at such short times only about 30% traces have an "ideal" shape with FWHM, which is close to the static resolution

The measurement of intensity fluctuations with a fast photodiode, as explained in Sect. 1, indicated 2–5% rms noise at the fundamental frequency which only slightly exceeds the corresponding ion laser noise (Sect. 2.3). At the same time the noise level of the second harmonic reached 5–35%, indicating substantial fluctuations of the pulse shape, as was also noted in [3]. The fluctuations of the second harmonic depend crucially on the ion laser adjustments and, as a rule, are weaker when working on the low-frequency side of the mode-locker acoustic resonance.

In Fig. 2 single-shot streak camera traces of the dye laser pulses are given together with the corresponding

[3] At best the long-term jitter could be reduced to ~30 ps judgeing by the FWHM of the synchroscan streak camera trace (integration time ≳ 1 s) as a result of careful optimization of the ion laser (Sect. 2.3)

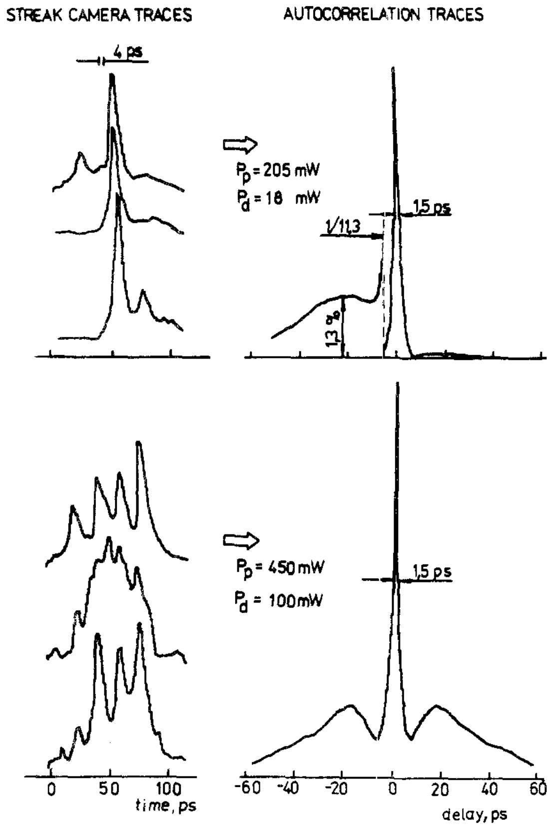

Fig. 2. Single-shot streak camera traces of the dye-laser pulses and corresponding autocorrelation functions. P_p and P_d are the average output powers of the ion and dye laser, respectively. The width of the streak camera traces is determined by the camera resolution

autocorrelation functions. The results of a series of measurements can be summarized in the following: (i) the fluctuations are more severe at higher pump levels; a substantial reduction of the pumping power or increase of the output coupler transmission results in cleaner pulses at the expence of the average power; (ii) the pulses tend to have satellites instead of chaotic substructure (see also [11]); (iii) at lower gain-to-loss ratio, apart from a single pulse, pulses with only one satellite pulse could frequently be detected (both leading or trailing) with about 20% intensity in the satellite and the distance between the pulses varying between 10–50 ps; (iv) even a weak (less than 1%) background in the autocorrelation function may give evidence of "dirty" pulses or a satellite, whereas "a satellite" in the autocorrelation function may result from a more complicated substructure. These results, as reported in [10], are in good agreement with [11, 13] and support the conclusion of [23] that intracavity power density is the critical parameter.

In [9, 12] the "noise burst" model [24] was employed to interpret the autocorrelation function of the dye laser output. The experiment was well described provided a constant background term was added. We suggest that the observed background (see also [2, 3, 25]) is caused by fluctuating (both in time and intensity) satellite pulses which may appear correlated, as evidenced by the characteristic shape of the autocorelation function in Fig. 2. Note that the background (as well as the wings) of any autocorrelation function is usually "noisier" than the central part – an argument supporting the previous statement.

2.2. Correlation Between Dye and Ion Laser Pulses

To relate the dye and ion laser noises we performed simultaneous single-shot streak camera measurements of pulse trains, as depicted in Fig. 3. Kr^+ laser pulses had 72 ± 9 ps FWHM (900 mW average power), whereas the dye pulse duration was less than the camera resolution, as shown in Fig. 2. The rms variation in the timing of the dye-laser pulses with respect to the pump pulses in the pulse trains was about 9 ps (determined from more than 30 independent traces). The more exact determination of this value is difficult due to simultaneous Kr^+ laser pulse shape fluctuations. Nevertheless, our figure does not contradict the results of [26], where two dye lasers were pumped by a single ion laser and the cross-correlation function indicated a jitter of less than 10 ps, nor of [16], where a synchronously-scanned streak camera was synchronized to the pulse train of the ion laser yielding a resolution of 9 ps for the dye laser output. This suggests that the pulse trains of dye and ion lasers are well locked to each other and the main part of the jitter proceeds from the pumping ion laser. The same conclusion is reached by comparing the intensity fluctuations of the lasers, which look highly correlated when monitored with a fast photodiode and a double-beam oscilloscope, as well as by measuring the power

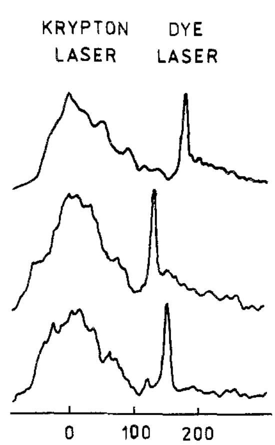

Fig. 3. Simultaneous single-shot streak camera traces of the ion and dye laser. Three traces from more than 30 are given to demonstrate the gratest observed variation in the timing of the dye laser pulse with respect to the pump pulse

spectrum of the laser intensity by using an electronic spectrum analyser [21][4]. Therefore one can almost neglect the influence of the internal noise of the dye laser owing to jet instabilities, dust particles, etc. on the mutual jitter of the pulse trains of the pumping ion laser and dye laser.

2.3. The Kr^+ *Laser*

It was shown in Sect. 2.2 that the mode-locked output of the Kr^+ laser yielded pulses of FWHM $\Delta t = 72 \pm 9$ ps measured with a single-shot streak camera (Fig. 3). The synchroscan streak camera measurements (Fig. 4) confirmed this value and at longer integration times revealed a considerable jitter highly resembling the behaviour of the dye laser. The autocorrelation function of the ion laser output intensity had a FWHM of about 120 ps also in rough accordance with the Δt value. The autocorrelation function showed no satellites under optimum conditions, neither were the wings "noisier" or fluctuating as in the case of the dye laser.

The ion laser output showed $\leqq 3\%$ rms noise at the fundamental frequency. The noise level of the frequency-doubled output was twice that of the fundamental one indicating, together with the streak camera results in Fig. 3, that there are only moderate pulse shape fluctuations present.

The spectral width of the mode-locked Kr^+ laser-emission line was $0.20\,cm^{-1}$ ($0.16\,cm^{-1}$ on free running), testifying nearly band-width-limited operation (for Gaussian pulse shapes). Hence, we can conclude that the mode-locked Kr^+ ion laser emits a train of well-defined picosecond pulses that exhibit rather weak intensity fluctuations due to residual rectifier noise but a considerable phase jitter, $30 \div 80$ ps (depending on the alignment), with respect to the 40.81 MHz mode-locker rf drive.

To elucidate the possible sources of the jitter we measured the time-shift between the pulse train and the rf sine as a function of locking frequency, cavity length detuning and plasma tube current. The following linear approximations hold: a 1 ps delay of the pulse train corresponds to a 3.7 Hz increases of the locking frequency or to a 0.73 µm shortening of the cavity length or to a 13 mA decrease of the tube current. On actual operation the tube current fluctuations seem to be the most reasonable source of the observed jitter.

[4] Although the qualitative conclusion made in [21] coincides with ours, there is an essential quantitative discrepancy between the estimated values of the jitter correlation times, which are about 10^2 times longer in [21]. The reason of this difference is not clear yet

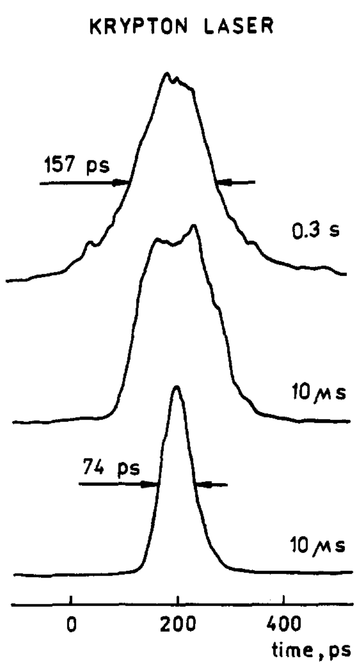

Fig. 4. Synchroscan streak camera traces of the Kr^+ laser output depending on the integration time. The two traces at the bottom at equal integration time demonstrate that the phase-jitter correlation time should be shorter than 10 µs

3. Discussion

Basing on the outlined observations we present a qualitative explanation of the lasing properties of a synchronously-pumped laser system under real (noisy) operating conditions. Let us first consider an ideal system: the ion laser emits a train of well-defined pulses exactly in synchronism with the mode-locker sine drive. The pulses are further shortened in the dye laser that emits a synchronized train of single pulses of variable duration depending on the cavity mismatch. Under optimum conditions single pulses of 1–5 ps duration are emitted. This corresponds to the steady-state regime as considered in a number of theoretical papers ([27, 28] and others) based on the self-reproducibility requirement. In a real system, however, fluctuations in the ion laser modulate the output intensity and, what is even more important, bring to phase fluctuations of the pulse train. Under these circumstances the dye laser is pumped by a pulse train whose temporal jitter by far exceeds the characteristic pulse durations of the dye laser. This very fact modifies crucially the output properties of the dye laser. High gain in the dye laser and broad time profile of the gain (compared to the duration of the dye laser pulses) favour the formation of satellite pulses, if some change in the pumping pulse train occurs. (As shown in [22], the dye laser pulse can be formed on a few roundtrips, whereas its steady-state shape and position are reached substantially later.) At the same time "old" pulses may still see considerable gain and therefore die out slowly or, if jitter is sufficiently fast, live till they are favoured again. From this we understand also, that at higher pump levels the multiple pulse operation can occur more easily and several pulses can survive for a longer time, as observed experimentally.

The above construction fairly well explain the observed experimental results, the jitter properties and their origin as well as the multiple-pulse operation of the dye laser. It is noteworthy that the phase-locking of the streak camera deflection directly to the dye (ion) laser pulse train does yield a better time resolution, though, owing to fluctuating pulse shapes, it cannot (and does not [16–18]) eliminate the phase jitter effect completely. At the same time this enables one to understand why the streak camera working in synchronizm with the passively mode-locked dye laser, where the pulse train jitter caused by the pumping laser is absent, ensures, as a rule, a better time resolution [19].

The primary cause of the observed jitter of the ion laser pulse train is the fluctuations in the tube current which modulate the refractive index of the active medium. The latter alters the cavity round-trip time, which in turn equals to a detuning of the cavity with respect to the rf drive of the mode-locker. Such a fluctuating detuning leads to substantial changes in the pulse timing, i.e. jitter. We note that the extremely critical dependence observed experimentally (see the end of Sect. 2.3) is also well described by the theory [29]. An important consequence of the above is that a higher Q-quality of the ion laser should reduce the jitter, as lower saturated gain results in a weaker mode pulling [30]. The experiment proved it twofold: a better alignment reduced the jitter, whereas replacing the 4% output coupler by a 7% one increased the jitter by 30%. A quantitative description of the mode-locked ion laser will be published elsewhere [31].

In conclusion, we have shown (see also [21]) that the main noise properties of a synchronously mode-locked system – jitter and pulse shape fluctuations of the dye laser – are caused by the phase fluctuations of the pumping ion laser pulse train. So far the deteriorated dye output has been avoided by limiting the pumping power and increasing the output coupling of the dye laser, i.e. by reducing average output powers. This has enabled reaching an acceptable noise level in pump and probe measurements and achieving a moderate 10 ps resolution in synchroscan streak camera systems. To allow further progress in this field (e.g. $\sim$1 ps resolution in synchroscan cameras) a substantial improvement of the ion laser stability and, first of all, the improvement of power-supply stability is required. A combined active-passive mode-locking of the dye laser should also give a positive effect. Such kind of efforts are now under way in our laboratory.

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References

1. A.I. Ferguson, J.N. Eckstein, T.W. Hänsch: J. Appl. Phys. **49**, 5389 (1978)
2. R.K. Jain, C.P. Auschnitt: Opt. Lett. **2**, 117 (1978)
3. S.R. Rotman, C.B. Roxlo, D. Bebelaar, M.M. Salour: Appl. Phys. Lett. **36**, 886 (1980)
4. J.P. Ryan, L.S. Goldberg, D.J. Bradley: Opt. Commun. **27**, 127 (1978)
5. B. Couillaud, A. Ducasse, L. Sarger, D. Bosher: Appl. Phys. Lett. **36**, 1 (1980)
6. H. Klann, J. Kuhl, D. von der Linde: Opt. Commun. **38**, 390 (1981)
7. A.I. Ferguson, R.A. Taylor: Opt. Commun. **41**, 271 (1982)
8. E.W. Stryland: Opt. Commun. **31**, 92 (1979)
9. D.M. McDonald, J.L. Rossel, G.R. Fleming: IEEE J. QE-**17**, 1134 (1981)
10. A.O. Anijalg, E.B. Berik, A.A. Vill, P.L. Kukk, V.T. Mihkelsoo, P.M. Saari, A.M. Freiberg, K.E. Timpmann: Proc. 2nd Soviet-France Symposium of Construction of Optical Apparatus (Moscow, 16–21 March 1981) p. 13
11. S.L. Shapiro, R.R. Cavanagh, J.C. Stephenson: Opt. Lett. **6**, 470 (1981)
12. D.P. Millar, A.H. Zewail: Chem. Phys. **72**, 381 (1982)
13. C. Kalpuozos, G.A. Kenney-Wallace, P.M. Kroger, E. Quitevis, S.C. Wallace: In *Picosecond Phenomena* III, ed. by K.B. Eisenthal, R.M. Hochstrasser, W. Kaiser, A. Laubereau, Springer Ser. Chem. Phys. **23** (Springer, Berlin, Heidelberg 1982) p. 221
14. M.C. Adams, W. Sibbett, D.J. Bradley: Opt. Commun. **26**, 273 (1978)
15. A. Freiberg, A. Raidaru, A. Anijalg, K. Timpmann, P. Kukk, P. Saari: Eesti NSV Tead. Akad. Toim. Füüs. Matem. **29**, 187 (1980)
16. P.G. May, W. Sibbett, K. Smith, J.R. Taylor, J.P. Willson: Opt. Commun. **42**, 285 (1982)
17. A.O. Anijalg, K.E. Timpmann, A.M. Freiberg: Pis'ma Zhurn. Tekh. Fiz. **8**, 1461 (1982)
18. J. Aaviksoo, A. Freiberg, S. Savikhin, G.F. Stelmakh, M.P. Tsvirko: Chem. Phys. Lett. **111**, 275 (1984)
19. W. Sibbett, W.E. Sleat, J.R. Taylor: In *Picosecond Chemistry and Biology* (Science Reviews, Northwood, Middlesex, 1984) p. 197
20. J. Aaviksoo, A. Anijalg, A. Freiberg, Ü. Mets, P. Saari, K. Timpmann: Techn. Digest of the Intern. Conf. Lasers '81 (New Orleans, 14–18 Dec. 1981) p. 97
21. J. Kluge, D. Wiechert, D. von der Linde: Opt. Commun. **51**, 271 (1984)
22. J. Hermann, U. Motschmann: Opt. Commun. **40**, 379 (1982)
23. J. Kluge, D. Wiechert, D. von der Linde: Opt. Commun. **45**, 278 (1983)
24. H.A. Pike, M. Hershel: J. Appl. Phys. **41**, 4562 (1970)
25. V. Sundström, T. Gillbro: Appl. Phys. **24**, 233 (1981)
26. R.K. Jain, J.P. Heritage: Appl. Phys. Lett. **32**, 41 (1978)
27. N.J. Frigo, T. Daly, H. Mahr: IEEE J. QE-**13**, 101 (1977)
28. D.M. Kim, J. Kuhl, R. Lambrich, D. von der Linde: Opt. Commun. **27**, 123 (1978)
29. O.P. McDuff, S.E. Harris: IEEE J. QE-**3**, 101 (1967)
30. W.E. Lamb: Phys. Rev. **134**, A1429 (1964)
31. J. Aaviksoo: Proc. ESSR Acad. Sc. Phys. Math. (to be published)