Temporal and Spectral Studies of a Synchronously Pumped Dye Laser: Detailed Interpretation of Autocorrelation Measurements

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Abstract—With the combination of precise autocorrelation and spectral measurements we have established the influence of pulse substructure on the determination of pulse envelope duration of a synchronously pumped dye laser within the noise burst model of Pike and Hersher. Our results make a single sided exponential pulse shape unlikely and we present evidence supporting pulses having Gaussian or skewed Gaussian temporal profiles.

INTRODUCTION

SYNCHRONOUSLY pumped dye lasers are being used for a variety of experiments probing very fast molecular processes [1], [2]. The time resolution of such measurements is ultimately limited by the length and reproducibility of the laser pulses. In order to take full advantage of deconvolution techniques, the temporal profiles of these short pulses must be understood in detail. The needed information on the pulses and various autocorrelations is far more detailed than can be obtained by streak camera investigations.

One of the most useful methods for measuring pulse lengths is the zero-background second-harmonic technique [3]. The signal generated by this method is proportional to the autocorrelation function of the pulse intensities:

$$G(\tau) = \left\langle \int_{-\infty}^{\infty} \langle I(t) \rangle_\phi \, \langle I(t+\tau) \rangle_\phi \, dt \right\rangle_N \qquad (1)$$

where the time fluctuations at the spectral frequency are averaged in the inner brackets, and the outer brackets indicate an average over a large number of pulses. In the noise burst model, Pike and Hersher [4] have decomposed $G(\tau)$ into the product of two autocorrelation functions: one for the pulse envelope and one for the bandwidth limited substructure. We have undertaken a systematic study of these autocorrelation functions and the corresponding spectra for a variety of cavity lengths, output couplers, wavelengths, and tuning elements.

The combination of autocorrelation and spectral measurements allows reliable determination of the influence of coherence in the autocorrelation measurements. We are thus able to determine the influence of cavity detuning on the pulse envelope without the distortion imposed by coherence, and without con-

fining our observations to the "region of good mode locking." In fact, the influence of coherence is particularly insidious at close to optimum cavity length, since smooth autocorrelations can be obtained even when the pulse envelope-bandwidth product is 2–3 times the transform limit [5]. Once the coherence and envelope widths can be extracted reliably from autocorrelation measurements, it should be possible to deconvolute rise times containing both the coherent coupling contribution [6], [7] for which the appropriate time scale is determined by the coherence width, and the contribution from the incoherent portion of the molecular response with the pulse envelope.

EXPERIMENTAL

A CR12 argon ion laser was acoustooptically mode locked with a Rockland 5600 A frequency synthesizer and ENI 300L RF amplifier driving a standard mode-locking prism. The cavity of a CR590 dye laser with either a three-plate birefringent filter or wedge etalon was extended to achieve synchronous pumping. Autocorrelations were obtained by the zero background second-harmonic technique using a 2 mm thick $LiIO_3$ crystal. A microcomputer controlled stepping motor driven translation stage (Micro Controle MT160-250) was used to vary the time delay between the arrival of the two pulses in the crystal. The second-harmonic light was detected by a photomultiplier whose output was processed by a Brookdeal 9503 lock-in amplifier and voltage to frequency converter, and stored in the memory of a Tracor Northern TN 1706 multichannel analyzer (MCA). Cavity length changes were made using an Aerotech ATS300 translation stage and were measured using a dial gauge with 10 μm resolution.

Spectra of the laser pulses were taken with a Princeton Applied Research 1205A-1205D optical multichannel analyzer (OMA) and a 1 m Engis, Czerny–Turner monochromator with a 1200 l/mm holographic grating (American Holographics). The dispersion of this system in first order was about 0.019 nm/channel at 600 nm. The accumulated spectra were transferred to the MCA via the plot/print control.

The autocorrelation traces and spectra were both transferred from the MCA memory to a Digital Equipment VAX computer where curve fitting was accomplished by methods described by Bevington [9]. The general functional form used was

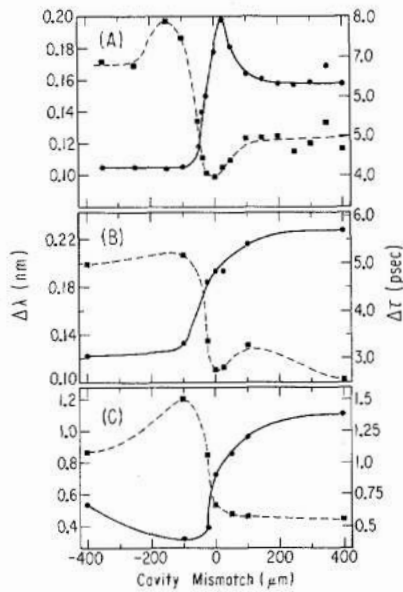$$G(\tau) = AG_1(\tau) [1 + DG_2(\tau)] + F \qquad (2)$$

where

Fig. 1. Spectral width $\Delta\lambda$ (●——●) and substructure width $\Delta\tau_N$ (■---■) versus cavity length mismatch for the (a) 70 percent output coupler and BRF, (b) the 55 percent output coupler and BRF, and (c) the 55 percent output coupler and wedge etalon. Note the change in the vertical scale in (c).

$$G_1(\tau) = e^{-(x-c)^2/2B^2} \tag{3}$$

or

$$\left[1 + \left(\frac{x-c}{B}\right)^2\right]^{-1} \tag{4}$$

or

$$e^{-(x-c)/B} \tag{5}$$

and

$$G_2(\tau) = e^{-(x-c)^2/2E^2}. \tag{6}$$

The term in brackets in (2) corresponds to the autocorrelation of the pulse substructure, while G_1 corresponds to the autocorrelation of the pulse envelope. The same fitting program with D fixed at 0 was used to fit simple Gaussian, Lorentzian, or exponential functions to the observed spectra or the second halves of the necessarily symmetric autocorrelations.

RESULTS

A. Spectra

Fig. 1 shows the spectral full width at half maximum for a three plate birefringent filter (BRF) with 70 and 55 percent transmitting output couplers, and for a wedge etalon (WE) with 55 percent output coupler. The spectral widths were obtained by Gaussian fits to the data. The position of the spectral maximum was independent of cavity length, but the spectral half-width was a strong function of cavity length in all cases. In all three plots short cavities lead to narrow spectra and there is a rapid increase in spectral bandwidth by roughly a factor of two as the cavity is lengthened towards the length of exact match with the pump laser. With the 55 percent T output coupler the spectral width increases more slowly and attains a
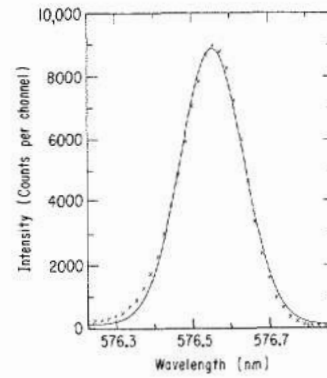

Fig. 2. Spectrum of the laser operating at optimum cavity length with 70 percent output coupler and BRF along with the best fit to a Gaussian function.

steady value as the cavity is lengthened further. Lengthening beyond the optimum with the 70 percent output coupler, on the other hand, causes the spectrum to narrow again. Presumably this difference arises because the laser is being pumped much closer to threshold with the 70 percent output coupler. For optimum length cavities the wedge gives a spectrum about five times as wide as the three plate BRF. We will see, however, that this difference is not reflected in the ratio of the pulse durations obtained with the two tuning elements.

Ryan *et al.* [10] in their study of a synchronously pumped prism tuned dye laser found a similar spectral broadening as the cavity of their laser was lengthened. They, however, also observed significant spectral shifts as the cavity length was changed. Ryan *et al.* [10] suggest that such shifts were probably due to partial compensation for cavity length changes by the laser taking a different path through the prism. This explanation is corroborated by our observation of no spectral shift in a laser where no such path change can compensate for a length mismatch.

The spectra obtained with the three plate BRF, for all cavity lengths, were much better fit by a Gaussian function than by a Lorentzian function. Fig. 2 shows a typical spectrum and fitted curve for the birefringent filter. The wedge etalon yielded spectral shapes having a poorer fit to a Gaussian. Real time display of the spectrum showed instability of the position of maximum wavelength. The instability was less than one half the spectral half-width. This instability was not present with the birefringent filter. Since spectra were recorded using >10^6 pulses, it is not straightforward to assume that our spectra are representative of a single pulse. We will show later, however, that in many cases the individual laser pulses could have had a narrower spectrum only by violation of the bandwidth limit. Under these circumstances it seems reasonable to take the spectral shape as if it were due to a single pulse.

II. AUTOCORRELATION WIDTHS

Fig. 1 also contains representative measurements of the FWHM of $G_2(\tau)(\Delta\tau_n)$ obtained by fits of autocorrelation measurements using (2)-(6). The fit of the coherence spike to a Gaussian function is excellent, as would be expected from the Gaussian spectral shape. The pulse envelope portion of the autocorrelation does not fit to either a Gaussian or Lorentzian
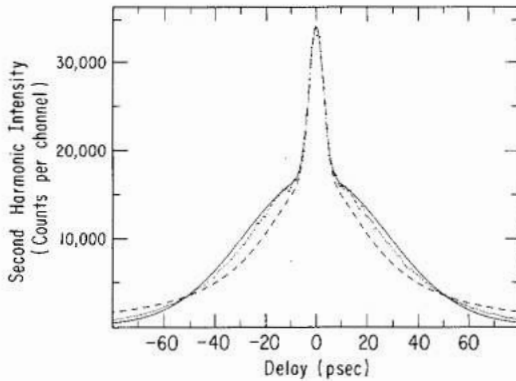
Fig. 3. Autocorrelation trace with the dye laser cavity 250 μm too long, the BRF, and 70 percent output coupler. The fitted curves are defined by (2) with a Gaussian (——) and Lorentzian (---) envelope profile.
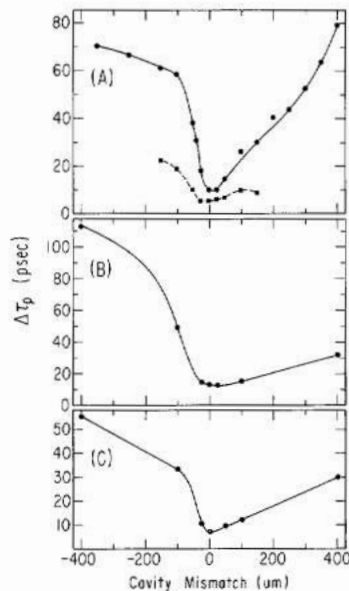

Fig. 4. Pulse envelope width $\Delta\tau_p$ versus cavity length mismatch for (a) the 70 percent output coupler and BRF, (b) the 55 percent output coupler and BRF, and (c) the 55 percent output coupler and wedge etalon. For comparison, the FWHM of the autocorrelation $\Delta\tau$ is included in (a).

TABLE I
PULSE PARAMETERS AT VARIOUS LASER CONDITIONS

LASER CONDITIONS		$\Delta\nu(s^{-1})$	Δt_N(ps)	$\Delta t_N \Delta\nu$	Δt_p(ps)	$\Delta t_p \Delta\nu$
70% BRF	+400μm	1.40×10^{11}	3.30	0.462	55.77	7.81
	+150μm	1.42×10^{11}	3.49	0.496	21.13	3.00
	0μm	1.57×10^{11}	2.81	0.441	7.09	1.11
	-100μm	9.29×10^{10}	5.40	0.502	41.20	3.83
	-350μm	9.29×10^{10}	4.86	0.451	49.73	4.62
55% BRF	+400μm	2.12×10^{11}	1.80	0.382	22.21	4.71
	+100μm	2.01×10^{11}	2.32	0.466	10.56	2.12
	0μm	1.80×10^{11}	1.95	0.351	8.81	1.59
	-100μm	1.24×10^{11}	3.65	0.453	34.79	4.31
	-400μm	1.14×10^{11}	3.52	0.401	80.42	9.17
55% WE	+400μm	9.99×10^{11}	0.424	0.424	20.96	2.09
	+100μm	8.66×10^{11}	0.460	0.398	8.66	7.50
	0μm	6.56×10^{11}	0.590	0.387	4.96	3.25
	-100μm	2.88×10^{11}	1.06	0.305	23.58	6.79
	-400μm	4.85×10^{11}	0.820	0.398	39.25	19.0

0.06 (Gaussian transform limit within experimental error) and remains constant even for autocorrelations obtained from cavities near optimum length, where the contributions from $G_1(\tau)$ and $G_2(\tau)$ cannot be visually discerned. The average time bandwidth product for the pulse substructure is calculated using all the points displayed in Fig. 1 (which includes those of Table I). The corresponding product for the pulse envelope $\Delta t_p \Delta\nu$ is a strong function of cavity length with the minimum value corresponding to a Gaussian pulse roughly 2.5 times wider than transform limit. (This deviation from transform limited behavior is, of course, implicit in the use of (2) to describe the autocorrelations [4].) Deviations from the transform limit are much larger with the wedge than the three plate BRF. Then minimum envelope width for the wedge etalon (55 percent output coupler) is only 60 percent narrower than for the BRF despite the five-fold difference in the transform limited pulsewidth.

Referring to (2), the value of D[the ratio of heights of $G_1(\tau)$ and $G_2(\tau)$] should be unity. We found $D = 1.0 \pm 0.1$, but occasionally D values of slightly less than one yielded an unambiguous improvement in the fit. This corresponds to a coherence spike of slightly reduced height and could indicate that the substructure width measured is slightly wider than for the actual pulse.

DISCUSSION

Laser Pulse Shape

Unambiguous determination of the pulse shape from autocorrelation measurements is not possible, since the autocorrelation is necessarily symmetric, while the pulse itself may not be. The presence of the coherence spike (for pulses longer than the transform limit) can also cause difficulty in determination of the shape of the envelope portion of the autocorrelation. Inclusion of detailed spectral width and shape measurements greatly assists in the interpretation of the autocorrelation shapes. In particular, the influence of coherence (pulse substructure) is described very well by the model of Pike and Hersher [4]. By use of (2), the pulse envelope autocorrelation can be obtained unambiguously, even where the structure of

function very well, but lies somewhere between the two. We have also tried fitting our pulse envelopes to sech^2 functions which have been suggested by models of passively mode-locked systems [11], but find the quality of these fits indistinguishable from the quality of those for a Gaussian. Fig. 3 shows a typical autocorrelation trace and the best fit Gaussian and Lorentzian envelopes. Fig. 4(a)-(c) shows the dependence of the pulse envelope width [FWHM of $G_1(\tau)$] on cavity length for the conditions in Fig. 1. The widths are those for a fit to a Gaussian shape, although Lorenztian pulses would yield the same general result.

The reciprocal nature of $\Delta\tau_N$ and $\Delta\lambda$ is clearly apparent in Fig. 1; for a more detailed comparison, Table I lists values of $\Delta\nu$, Δt_N, Δt_p, $\Delta t_N \Delta\nu$, and $\Delta t_p \Delta\nu$ for several experimental conditions

$$[\sqrt{2}\Delta t_N = \text{FWHM } G_2(\tau) = \Delta\tau_N, \sqrt{2}\Delta t_p$$
$$= \text{FWHM } G_1(\tau) = \Delta\tau_p].$$

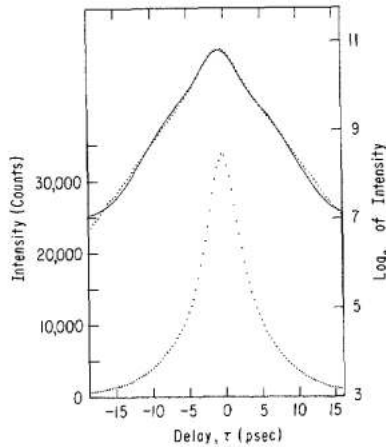It is particularly encouraging that the product $\Delta\tau_N \Delta\nu$ is 0.43 $\pm$

Fig. 5. Autocorrelation trace for near optimal cavity, the BRF, and 70 percent output coupler. (a) Semilogarithmic plot of data (· · ·) and fit to (2) (———). (b) Linear plot of data.

$G(\tau)$ is not apparent, i.e., at, or close to, optimum cavity lengths. We believe that the results presented in Fig. 4 represent the first experimental determination of the cavity length dependence pulse envelope autocorrelation width corrected for the effects of coherence. In Fig. 4(a), we also show the FWHM of $G(\tau)$ [see (1)] as a function of cavity length over the region where $G(\tau)$ appears as a smooth function. This function is clearly a shallower function of cavity length than $\Delta\tau_p$ and also leads to a considerable underestimate of the actual pulse duration. The variation in pulse envelope with cavity length is qualitatively quite similar to the calculations of Kim *et al.* [12]. The minimum is significantly sharper for the 70 percent output coupler than for 55 percent. Generally, our envelope width does not increase as rapidly for short cavities as in the calculations of Kim *et al.* [12]. A second difference is, as shown by our previous work [5], the minimum envelope duration occurs for cavities in exact synchrony with the pump laser, rather than for slightly longer cavities [12].

Fig. 5 shows a semilogarithmic plot of an autocorrelation for optimal cavity length, along with a fit to (2) with G_1 and G_2 both Gaussian.

The excellent fit of autocorrelations from close to optimum cavity lengths to functions of the form $\exp(-\alpha|x|)$ has led several authors to hypothesize that their pulses are bandwidth limited single-sided exponentials [13]–[15]. Our spectral data rule out this possibility for our pulses—which are generated in the same manner. There are two pieces of evidence that lead to this conclusion. First, a bandwidth limited pulse with single-sided or double-sided exponential temporal profile has a Lorentzian spectral profile, and such a spectral shape is not observed. Second, the time bandwidth product for an exponential pulse is almost one order of magnitude smaller than that observed for our pulse, if we assume exponential shape. We are thus confident that synchronously pumped dye lasers do not give bandwidth limit single-sided exponentials, and that by making this assumption many authors have underestimated their pulsewidths by about a factor of three. For example, a symmetric exponential $G(\tau)$ with FWHM of 2 ps would correspond to a Δt_p of about 2.7 ps rather than 1 ps.

In arriving at these conclusions we have considered other possible explanations for the shape and width of the laser spectra. There is always the possibility that the laser pulses could be chirped or that the central wavelength of the pulses could vary greatly over the time scale of our spectral measurements. Such processes would not necessarily show up in the autocorrelation functions [4]. If the pulses were exponential, it would be possible to obtain spectra wider than predicted by the bandwidth limit and possibly even of a Gaussian shape rather than the expected Lorentzian one. We feel that this is not a satisfactory explanation for our observation, that under all conditions the coherence peak is Gaussian. Under conditions of cavity mismatch where the coherence spike is most prominent, it is clear that the spectrum should be Gaussian and cannot be significantly narrower than observed. This agreement between spectrum and Fourier transform of coherence puts rather narrow limits on the degree to which the pulses could be chirped or jump about in wavelength (at least at cavity mismatch). A catastrophic narrowing in the spectra of individual pulses (accompanied by the necessary wavelength instability) at perfect length matching could result in our observed spectra, but the consistent quality of our fits to the coherence spike for all cavity lengths would not support such an explanation. A more rigorous demonstration could be accomplished by taking the spectrum of a single pulse along with a "complete" autocorrelation as described by Diels [16].

The remaining question is the origin of the exponential shape of $G(\tau)$ for a perfectly matched cavity. The envelope shapes obtained through (2) for closely matched cavities, although much shorter than for mismatched cavities, are essentially the same shape at all cavity lengths and are roughly Gaussian. There is little difference in the quality of the fit to (2) for close match or large mismatch even though the envelope and coherence spike are clearly not exponential at large mismatch. In a previous paper [5] we suggested that the exponential shape simply arose from the form of (2) with $\Delta t_p \sim 2\text{-}3\ \Delta t_n$. The present results strongly support this conclusion.

Recently, van Stryland [15] has pointed out the importance of remembering that (1) contains as ensemble average over the nearly 40 million pulses used to collect each data point. Some of the pulses are likely to be longer than others, for example, those occurring shortly after lasing has been interrupted by a bubble in the dye jet or a similar brief trauma. It is possible to generate almost any shape of autocorrelation function by summing the appropriate distribution of Gaussian functions of different widths. van Stryland obtains a symmetric exponential by using a rather large distribution. He ignores, however, the presence of the coherence spike, the inclusion of which obviates the necessity for such a large distribution. As noted earlier [5], even the sum of two Gaussian fits very well (see Fig. 5). The deviation of our fit from the experimental curve in Fig. 5 is probably because we have used only a single Gaussian in fitting the envelope function. Following van Stryland's suggestion [17], a modest distribution of pulse durations will produce the required exponentials. On the other hand, a distribution of exponential or Lorentzian pulses is not capable of reproducing the observed data for mix matched cavities. We conclude that the pulses, however skewed, resemble Gaussian functions.

We have also looked at the autocorrelation function obtained from the optical Kerr effect in CS_2 [18]. This autocorrelation contains the information of the third-order autocorrelation function

$$G^{(3)}(\tau) = \left\langle \int_{\infty}^{\infty} \langle I(t) \rangle_\phi^2 \, \langle I(t-\tau) \rangle_\phi \, dt \right\rangle_N$$

convoluted with the rotational correlation function of CS_2. This third-order autocorrelation function is sensitive to pulse asymmetry, but our traces are symmetric. We feel that this indicates that our pulses are skewed only slightly if at all. Insertion of a solid etalon of finesse 3 and free spectral range 15 cm^{-1} narrowed the spectrum to about 0.25 Å at 570 nm. The resultant autocorrelations at optimum cavity matching fit extremely well to a single Gaussian function corresponding to bandwidth limited pulses.

Deconvolution of Molecular Response Times

In pump-probe spectroscopy [6] where the probe and probe pulses derive from the same ultrashort light pulse, the observed molecular response is the sum of two components: the convolution of the molecular response with the pump and probe pulse envelopes and the coherent coupling contribution [6]–[8]. These two contributions will have different characteristic time scales related to $G_1(\tau)$ and $G_2(\tau)$, respectively, if the pulse is not transform limited.

SUMMARY

The Pike and Hersher model [4] of ultrashort light pulses provides an excellent description of the pulses from a synchronously pumped dye laser. The spectrum and substructure of such pulses are definitely Gaussian. The pulse envelope is not a single-sided exponential but is most likely a Gaussian or skewed Gaussian. We do not intend to distinguish between Gaussian and sech^2 shapes, the latter having been predicted for passive mode-locked systems [11]. They are qualitatively similar and the pulsewidths predicted from their autocorrelations differ by only a few percentage points. Our analysis leads to pulse duration estimates as much as three times greater than that obtained by the assumption of transform limit single-sided exponentials. The explicit separation of the autocorrelation into envelope and coherence contributions is necessary for correct deconvolution of molecular response functions containing both coherent coupling and pulse envelope convolution contributions.

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