PULSE STRUCTURE STUDIES AND ABSOLUTE CAVITY LENGTH DETERMINATION FOR A SYNCHRONOUSLY PUMPED PICOSECOND DYE LASER

Daniel B. McDONALD, David WALDECK * and Graham R. FLEMING

Department of Chemistry and The James Franck Institute, The University of Chicago, Chicago, Illinois, 60637, USA

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Second harmonic cross correlation functions of a pulse with its near neighbor have been studied in a synchronously pumped cw dye laser. Measurements were made both as a function of dye laser cavity length mismatch and the number of cavity round trips separating the correlated pulses. The pulse envelope is found to have a characteristic interpulse frequency determined by the pump laser, whereas the pulse substructure has a characteristic frequency determined by the dye laser cavity length. The cross correlation measurements allow experimental determination of the dye laser length corresponding to exact synchrony. In contrast to theoretical predictions the length of exact synchrony corresponds to optimum pulse shape and duration. Our results are discussed in terms of a simple model which leads to pulse duration estimates as much as twice those obtained by conventional analysis of autocorrelation traces.

1. Introduction

Determination of the second order autocorrelation function has become a standard method for characterizing picosecond and subpicosecond pulses [1]. With the development of cw synchronously pumped dye lasers, and zero background noncollinear second harmonic generation techniques, the ensemble average second order autocorrelation function may be determined with a high degree of precision [2]. Second order autocorrelation functions are necessarily symmetric, and this cannot provide unambiguous information on pulse shape. The shapes of the measured autocorrelation functions have, however, been invaluable in determining the influence of various laser parameters (e.g. cavity length, loss, intracavity filter bandwidth) on the pulse quality [2–9]. Interpretation of these results, however, is complicated by the well understood but often neglected effects of coherence. For example, a pulse characterized as a noise burst (i.e. a burst of random noise with gaussian profile) exhibits an autocorrelation trace consisting of a sharp spike (coherence spike) sitting on a broad base [1]. The spike results from maximum overlap of the random substructure (maximum coherence) at zero time delay.

We have made a series of measurements of the cross correlation of pulses from a synchronously pump laser. Our expectation was that the substructure of the dye laser pulses would not persist for more than a few round trips, so that a cross correlation of successively removed neighboring pulses would exhibit a rapidly diminishing coherence spike. Our hope was to separate the effects of coherence and generate a correlation trace more directly related to the ensemble average pulse shape. This loss of coherence was not observed experimentally, but the results we obtained provide new information on pulse structure in synchronously pumped dye lasers and on the effect of cavity mismatch on pulse characteristics.

Our results show that the substructure of the mode locked pulse is reproduced with very little distortion, while the shape of the pulse envelope depends strongly on the time of arrival in the gain medium. This can lead to asymmetric cross correlations where the coherence spike is displaced from the center of the correlation of the pulse envelope. Our results strongly support the noise

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burst model for mode locked laser pulses. They also provide further insight into the gain buildup and amplification process in synchronously pumped systems.

A number of authors have considered the influence of cavity length detuning on synchronously pumped dye lasers [7–9]. A vital piece of information in this work is the dye laser length, corresponding to perfect synchronization with the pumping pulses, but to date this has not been experimentally accessible [7]. The change in symmetry of the cross correlation function as a function of cavity length tuning allows us to pinpoint the length of exact synchronization to within 10 μm.

2. Experimental

A CR12 argon laser was acousto-optically mode locked with a Rockland 5600A frequency synthesizer and ENI 300L rf amplifier driving a standard mode locking prism. The cavity of a CR590 dye laser with 3-plate birefringent filter was extended to achieve synchronous pumping. Auto and cross correlations were obtained by the zero background second harmonic technique [11] using a 2 mm thick LiIO₃ 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 was detected by a photomultiplier and, after processing by a Brookdeal 9503 lock in amplifier and voltage to frequency converter, stored in the memory of a multichannel analyzer (Tracor TN 1706). Cross correlations were obtained by increasing the length of the fixed arm in multiples of the dye laser length. In this manner correlations of the form n, n–m where m = 1–6 were obtained. For the m values greater than 1 a lens of 2000 mm fl was placed at its focal length from the final mirror in the delay to maintain a well collimated beam. Correlation traces for each value of m were recorded for a range of dye laser cavity lengths. The length changes were calibrated using a dial gauge and were reproducible to within 10 μm. More reproducible changes over small ranges were made with the fine adjust screw of the Aerotech AT5300 translation stage upon which the dye laser output coupler was mounted. In order to avoid uncertainties in the precise position of the sometimes broad maximum of the autocorrelation trace, care was taken to start each scan at the same position of the variable delay translation stage. Zero delay could then be related to a specific channel in our multichannel analyzer.

3. Results

Conventional autocorrelation traces as a function of cavity length were essentially identical with those described by Ausschnitt et al. [7]. For long cavity lengths they exhibit definite shoulders. Shortening the cavity eliminates the shoulders and yields optimal pulse shape and duration. Progressively shorter cavities introduce small, broad satellite peaks which sharpen, strengthen and move closer to the (diminishing) central peak. If the dye laser was operated with insufficient loss the satellite peaks developed even at cavity lengths longer than those yielding the optimum central peak shape.

Autocorrelations with a full width at half maximum (FWHM) of 5.7 ps were obtained for 1.2 W pump, 55% T output coupler with 120 mW average power. Fig. 1e shows a typical trace. When the cavity length was mismatched autocorrelations of the type shown in fig. 1c were obtained, with a broad base and a sharp central coherence spike with height equal to that of the broad base.

Fig. 1 also shows the results of cross correlation (n, n–m) measurements for m = 6. For optimal cavity length the auto (fig. 1e) and cross (fig. 1d) correlations are essentially identical. For the mismatched cavities in the cross correlations the coherence spike marches to one side or the other of the broad base, depending on whether the dye laser cavity was too long or too short. The peak of the broad base (pulse envelope autocorrelation) remains in the same position in all cases.

The shape of the coherence spike remains remarkably constant under most conditions. Its FWHM is consistently 3.0–4.5 ps and it retains a regular gaussian shape as the cavity is detuned. High values of m with very short cavity lengths were occasionally observed to broaden the coherence correlation maximum, but not in any reproducible fashion. This temporal broadening should be interpreted as a spectral narrowing which would result from the combined tuning effects of the 3-plate birefringent filter and cavity group velocity dispersion. The height of the spike decreases relative to that
of the broader maximum but is always within experimental error of being equal to the height of the pulse envelope correlation trace on which it sits. The pulse envelope correlation trace itself depends only on the cavity length, the value of $m$ simply determines where the coherence spike sits on the envelope.

4. Discussion

4.1. Noise burst model of mode locked pulses

Our observations can be readily explained in terms of the model of a partially mode locked laser pulse discussed by Pike and Hercher [12] and Bradley and New [13]. They treat the pulse as a burst of bandwidth limit noise as illustrated in fig. 2. All three pulses were generated from the same squared sampling of simulated noise by multiplying by three gaussian functions of the same widths, but different origin displacements. The simulations in fig. 2 illustrate pulses that one might expect from a dye laser having mismatched cavity length. In the case shown here the cavity is too long, and the

![Fig. 1. Auto $(n,n)$ and cross $(n, n-m)$ correlation function measurements. a–c cavity length mismatched; d,e cavity length optimised. a) $n, n-6$ cavity length 500 $\mu$m too short; b) $n, n-6$ cavity length 500 $\mu$m too long; c) $n, n$ cavity length 500 $\mu$m too long; d) $n, n-6$ cavity length optimum; e) $n, n$ cavity length optimum.](image1)

![Fig. 2. Origin of the "coherence spike" for auto and cross correlations. Each trace represents the same sample of random noise shaped with a gaussian envelope. The envelopes are of equal width but are progressively displaced in the series $n, n-m, n-2m$. The cases shown correspond to maximum envelope overlap and maximum coherence of the noise (maximum substructure overlap).](image2)
pulse circulating in the dye laser arrives at the gain medium significantly later than the argon laser pulse. The leading portion of the pulse is amplified more than the trailing portion because of gain saturation, and the position of the peak of the pulse envelope is advanced [14]. The result is that dye pulses circulate with the same period as the pumping pulses, even if the cavity length is mismatched. The substructure on the pulse, however, is reproduced during the amplification process, but becomes shifted relative to the pulse envelope. In other words the substructure remembers the length of the dye laser cavity and accumulates a phase shift on each round trip.

The reason for the displacement of the coherence spike in the cross correlation measurements is now apparent. The spacing of the dye laser pulses is locked to the spacing of the argon pumping pulses (which are in turn locked to the rf driver) and is independent of the cavity length or number of cavity round trips separating correlated pulses. On the other hand, for a dye cavity too long (short) the delay of the \( n \)th pulse must be decreased (increased) in order for the noise to add in phase with \((n-m)\)th pulse. Only when the cavity periods of the pump and pumped lasers are equal will the coherence spike be centered on the envelope portion of the correlation trace. Since the pulse becomes shorter as the mismatch is decreased, the symmetry of the correlation trace becomes a sensitive indication of length detuning and allows determination of the resonant length to better than 10 \( \mu \)m for \( m = 6 \).

The above explanation clearly predicts that the coherence spike displacement should depend only on the cavity mismatch and the number of round trips separating the correlated pulses. Fig. 3 demonstrates that this is observed experimentally. The measured points for \( m = 1, 4, 6 \) are shown; the solid lines are the displacements calculated from the product of \( m \) and the round trip cavity detuning in picoseconds.

The observation that the height of the coherence spike is always twice the height of the part of the envelope on which it sits is consistent with the description presented above. When the pulse peak is advanced or retarded in the gain medium a portion of the input pulse is suppressed and a new portion of the pulse produced (e.g. at the front of the pulse if the pulse arrives late). The new portion of the pulse arises from spontaneous emission and will have substructure unrelated to the structure on the same portion of the envelope of a pulse from a previous round trip. Thus the amount of coherence (for a mismatched cavity) also decreases with mismatch and number of round trips \( m \) separating correlated pulses. However, the structure on the “old” part of the envelope is replicated (but shifted on the envelope). One exception to this simple situation has been observed when the cavity length is slightly short. The substructure of the amplified pulse can then, through the memory of the gain medium, impose some periodicity on the substructure of the new portion of the pulse envelope. Such regularity in the substructure could account for the triplet and quintuplet structure observed in some autocorrelation traces.

4.2. Pulse duration determination

The representation of the pulse as a noise burst leads to a measured autocorrelation function of the form [12]

\[
G(\tau) = G_p(\tau)[1 + G_N(\tau)],
\]

where \( G_p(\tau) \) represents the autocorrelation of the pulse envelope and \( G_N \) is a gaussian function resulting from the noise bandwidth. For pulse envelopes significantly longer than the noise bandwidth limit eq. (1) can be approximated by
\[ G(\tau) = G_p(\tau) + G_N(\tau). \] (2)

Even a factor of 2 in the time scales is sufficient to make the difference in half width of \( G_N \) and \( G_N G_p \) less than 12%. The contributions from \( G_N \) and \( G_p \) can be easily observed for large cavity length mismatch. However, in cases where there is little or zero mismatch the contributions are much more difficult to distinguish. It is possible to simulate autocorrelation traces with nearly exponential decays over two decades by summing two error functions of equal peak magnitude with standard deviations in the ratio 2.0–2.5 to 1. Such a trace would also result from a single or double sided exponential pulse having no substructure, but we feel a more reasonable picture is that of a gaussian (or skewed gaussian) pulse envelope of width 2.0–2.5 times the substructure bandwidth. The assumption of gaussian envelopes is consistent with the cavity mismatch case where the two components of \( G(\tau) \) clearly resemble gaussian functions much more closely than single sided exponentials, and does not require gradual pulse shape change with cavity length.

Applying eq. (2) to the pulse in fig. 1e we find \( \Delta \tau_p = 5 \) ps (for FWHM \( G(\tau) = \Delta \tau = 5.7 \) ps). This duration is significantly longer than would be obtained by the almost unanimous convention of dividing \( \Delta \tau \) by between 1.41 and 2 (\( \Delta \tau_p \approx 4 \) – 2.9 ps).

4.3. Cavity length determination

The position where the \( n, n-6 \) cross correlation trace becomes symmetric (or equivalently the zero intercept of the fit to the data in fig. 3) provides the first experimental determination of the dye laser length for perfect synchrony. Previously, cavity lengths were related, for example, to the length giving maximum second harmonic power [8,9]. The length of perfect synchrony is significantly shorter than the length for maximum SHG. In our experiments it does correspond to the optimum pulse duration, as might be expected on intuitive grounds, but in contrast to theoretical predictions requiring slightly shorter [9] or slightly longer [8] cavities than resonant length for optimal pulse duration.

When operating with excess gain the dye gain may exceed threshold a second time after the circulating dye laser pulse has passed through the gain medium. This leads to a second pulse circulating in the dye laser. This second pulse can be suppressed by (1) increasing the cavity loss or (2) lengthening the laser cavity. Both methods retard the phase of the dye laser pulse relative to the argon pump pulse and thus leave less remaining pump to reestablish an above threshold gain, however only the former methods will lead to the optimum pulse duration. The occurrence of multiple pulses even at the cavity length of exact synchronization is in good accord with the calculations of Scavennec [15] who also predicted our observations and that the maximum pulse intensity occurs at slightly larger cavity lengths.

5. Summary

The length of perfect synchrony for a synchronously pumped dye laser has been determined. It is found that optimal pulses are produced when the dye laser cavity is at this length, in contrast to theoretical predictions. The pulse shape is described in terms of two functions: one describing the pulse envelope and the other, the pulse substructure. The pulse envelope has a characteristic interpulse frequency determined by the repetition frequency of the pumping pulses. By contrast, the substructure has a characteristic interpulse frequency determined by the dye laser length. Thus, for mismatched cavities the substructure suffers a “phase shift” on each round trip, and in cross correlation measurements the position of maximum coherence is shifted from the position of maximum envelope overlap. The above picture of the pulse shape leads to estimates of the pulse duration of up to twice the currently accepted values. The pulse to pulse reproducibility of both pulse envelope and substructure is extremely high. It is this reproducibility over a small number of pulses that gives at least part of the large increase in signal to noise obtained in the high frequency chopping/lock in amplifier technique of Heritage et al. [16], and makes synchronously pumped lasers attractive sources for ultrafast spectroscopy.

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