EFFECTS OF PULSE VARIATIONS ON ULTRASHORT PULSE WIDTH MEASUREMENTS

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An alternate interpretation is presented for the exponential shapes of the second harmonic autocorrelation functions obtained from repetitively pulsed picosecond lasers such as the synchronously-pumped mode-locked dye lasers. Such pulse width measurements in the past have averaged over a large number of pulses. If the pulsewidth varies in a time short compared to the time needed to measure the autocorrelation function the observed autocorrelation function will be a weighted average of pulses of varying widths. Several pulse shapes and pulse width distributions are examined all leading to exponential shaped autocorrelation functions. This interpretation implies that the currently determined pulsewidths are shorter, possibly many times shorter than the actual average pulsewidths. A method for determining the pulsewidth distribution and of selectively descriminating the shortest pulses from the train to increase the time resolution of current laser systems is presented.

Second order autocorrelation functions are often used to measure the temporal duration of ultrashort pulses emitted by repetitively pulsed lasers such as synchronously-pumped mode-locked dye lasers. One basic assumption made in using this method is that all pulses are identical. If there exist significant pulse to pulse variations in the temporal duration, the width as determined from a second order autocorrelation method is considerably shorter than the true average pulsewidth. In addition the apparent pulse shape is altered.

Mode locked dye laser systems are providing the researcher with extremely short pulses for probes of various ultrafast phenomena [e.g. 1]. While cw argon ion pumped passively mode locked dye laser systems have provided the shortest pulses to date (less than 0.2 ps) [2] they are restricted to a narrow bandwidth around 6100 Å and a few other selected wavelengths. The use of an actively mode locked argon ion laser as a source to synchronously pump the dye laser offers the researcher the advantage of tunability. Pulses as short as 0.7 ps have been reported with such systems [3,4]. These short pulses are assumed to have a single sided exponential shape since such a shape fits the experimentally measured integral of the product of the pulse intensity with itself, $G^{(2)}$. ($G^{(2)}$ is the second order autocorrelation function). The pulse width measurements made on such systems average over many pulses (usually $>10^9$). We present here an alternate explanation of the shape of $G^{(2)}$ in terms of a weighted average of the second order autocorrelation functions of many individual pulses with a distribution of pulse widths. Such an average yields exponential shaped $G^{(2)}$'s primarily sensitive to the width of the distribution of pulsewidths and not to the shape of the individual pulses. The conclusion is that the currently used method of pulse width determination may yield widths much shorter than the average width of the dye pulses. A suggestion for experimentally determining how important these pulse width fluctuations are for a given laser system and how to selectively observe only the shortest pulses in the train is also described.

These ultrashort pulsewidths for both actively and passively mode locked systems have been determined by the indirect method of second harmonic generation (SGH) autocorrelation [3-5]. A continuous train of ultrashort pulses from the dye laser is split into two beams. One beam passes through a fixed optical delay, the other through a variable delay. These two beams are recombined in a phase matched SHG crystal, and the amplitude of the second harmonic detected. Several variations of this technique are in use [6]. The second harmonic amplitude plotted as a function of delay, τ, yields the SHG autocorrelation function for the pulse, $G^{(2)}_{\tau_o}$, where τ_o is the fhwm. Thus,

$$G^{(2)}_{\tau_o}(\tau) = \int_{-\infty}^{\infty} |E(t)|^2 |E(t+\tau)|^2 \, dt \qquad (1)$$

where E(t) is the electric field amplitude of the dye laser pulse, assumed here to be the same from one pulse to the next. It is well known that the determination of the actual pulsewidth from the SHG autocorrelation function is not unique but depends on the assumed pulse shape. This pulse shape, however, is inferred from the shape of the autocorrelation function thus yielding a pulsewidth. For example, a single sided exponential shaped pulse would yield an exponential $G^{(2)}_{\tau_o}(\tau)$ of twice the pulse fwhm ($2\tau_o$). Gaussian shaped pulses yield a Gaussian $G^{(2)}_{\tau_0}(\tau)$ of width $\sqrt{2}\tau_0$, while sech^2 shaped pulses yield a sech^2 $G^{(2)}_{\tau_0}(\tau)$ of width ~1.55 τ_0. It has been assumed in determining the pulsewidths for synchronously-pumped dye lasers that all pulses are identical.

The extreme sensitivity of synchronously pumped dye laser systems to the mode-locker frequency, and cavity length detuning, suggest that the pulses may indeed not be identical [7]. Fluctuations in the pump or cavity length may lead to pulses of varying widths (or shapes). The observed autocorrelation function, $G^{(2)}_{obs}(\tau)$, is then a weighted average of the second order autocorrelation functions of the individual pulses given by:

$$G^{(2)}_{obs}(\tau) = \int_0^\infty G^{(2)}_{\tau_0}(\tau)P(\tau_0)d\tau_0 \quad , \tag{2}$$

where $G^{(2)}_{\tau_0}(\tau)$ is the autocorrelation function of a pulse of width (fwhm) τ_0 and $P(\tau_0)d\tau_0$ is the probability that a pulse has a width between $\tau_0 + d\tau_0$. This probability is normalized to 1 (i.e. $\int P(\tau_0)d\tau_0 = 1$). Since $G^{(2)}_{\tau_0}(\tau)$ depends on the square of the optical intensity the shortest pulses are most heavily weighted in such an average. Thus, $G^{(2)}_{obs}(\tau)$ tends to peak up more near $\tau = 0$ than $G^{(2)}_{\tau_0}(\tau)$ for an individual pulse. This is consistent with observations of synchronously pumped dye laser outputs. The smoothly varying $G^{(2)}_{obs}(\tau)$ observed for some passively mode-locked systems is replaced by a steeply peaked function, the exponential [2-4].

A reasonable assumption for these dye laser pulses is that the total energy per pulse is constant (ε_0). That is

$$\int_{-\infty}^{\infty} |E(t)|^2 dt = \varepsilon_0 \quad . \tag{3}$$

The constant energy assumption is experimentally justified by noting that the laser output (linear detector) does not fluctuate from pulse to pulse. (Since the detectors integrate the intensity, this does not imply the pulsewidth is constant).

We can now evaluate $G^{(2)}_{obs}(\tau)$ for various pulse shapes and pulse width distribution functions. In order to make comparisons between the various $G^{(2)}_{obs}(\tau)$'s the average pulsewidths are held constant at $\tau_{ave} = \int_0^\infty \tau_0 P(\tau_0)d\tau_0$. Fig. 1(a) shows $G^{(2)}_{obs}(\tau)$ (solid lines) for Gaussian shaped pulses with a Gaussian pulse width distribution $P(\tau_0)$ centered about $\tau_{ave} = 10$ having a fwhm of 10 units. The dashed curve is the autocorrelation function for a single pulse of fwhm $\tau_{ave} = 10$ units. The distribution, $P(\tau_0)$, is cutoff at some lower limit since the probability of obtaining pulses shorter than the inverse bandwidth goes to zero. The upper cutoff is picked to keep τ_{ave} at the center of the distribution. The integral is relatively insensitive to the upper cutoff because the shorter pulses are more heavily weighted. The dotted line in Fig. 1(a) is a simple exponential of a width closely matching the width of $G^{(2)}_{obs}(\tau)$. It would be exceedingly difficult to experimentally distinguish the two curves.

Similar results are obtained for Gaussian shaped pulses with a rectangular $P(\tau_0)$ as shown in Fig. 1(b). Here the pulse width is assumed to vary from 2 to 18 units or by a factor of 9. Doubling this pulse width variation (i.e. a factor of 18 variation) still gives a good fit to an exponential but halving the variation (i.e. a factor of 4 variation) results in a distribution that is between an exponential and a Gaussian. A pulse width variation of approximately 6 or greater is needed to obtain a curve that is well fit by an exponential for Gaussian or sech^2 shaped pulses and a rectangular $P(\tau_0)$. The exponential shape of $G^{(2)}_{obs}(\tau)$ is primarily determined by the width of the distribution $P(\tau_0)$. Not even the initial pulse shape affects the exponential shape of $G^{(2)}_{obs}(\tau)$ substantially as can be seen from Fig. 1(c). Rectangular pulses with triangular autocorrelation functions were used to obtain this figure. Again an exponential fits $G^{(2)}_{obs}(\tau)$ well. It should also be noted that the model presented here does not exclude having a distribution of single sided exponential shaped pulses. Averaging the autocorrelation functions of one-sided exponentials over a distribution of temporal widths also yields a function barely distinguishable from an exponential except that for wide distributions a small background is present. Such a background is normally attributed to optical noise or to slight optical misalignment in the case of autocorrelation functions with background.

Determining the pulsewidth from $G^{(2)}_{obs}(\tau)$ under the assumption that all pulses are identical single sided exponentials in shape results in a value smaller than the actual average

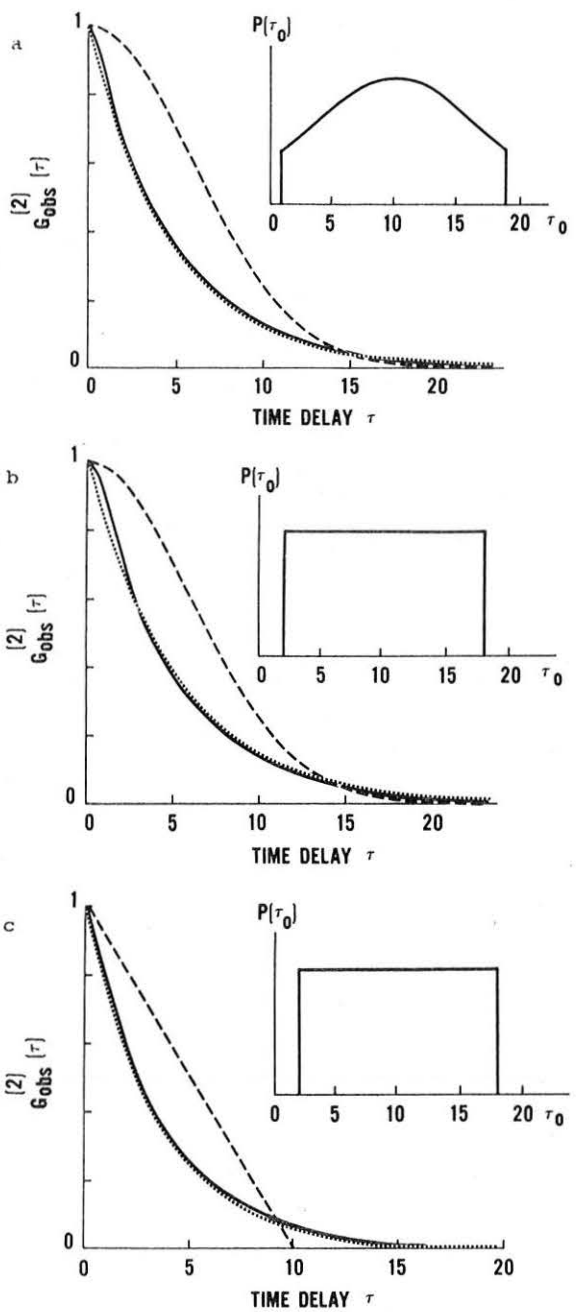

Fig. 1. The solid lines in a, b, and c show $G^{(2)}_{obs}(\tau)$ using the appropriate $P(\tau_0)$ shown in the inset for each figure. Pulses of Gaussian temporal profile were used to determine a and b while rectangular pulses were used for c. The dashed lines in a, b, and c represent the SHG autocorrelation function of individual pulses of fwhm equal to τ_{ave} ($G^{(2)}_{\tau_{ave}}(\tau)$) using a Gaussian pulse for a and b, and a rectangular pulse for c. The dotted line in all figures is a simple exponential fit to each $G^{(2)}_{obs}(\tau)$.

pulsewidth, τ_{ave}. For example, from Fig. 1a the measured autocorrelation width of $G^{(2)}_{obs}(\tau)$ is approximately 0.5 τ_{ave}. Not only is this width shorter than τ_{ave}, but rather than dividing by $\sqrt{2}$ as one should do for Gaussian shaped pulses the autocorrelation width is divided by 2 under the assumption that the pulses are exponential in shape. This gives a calculated pulsewidth of $\tau_{ave}/3$, or a factor of three too short. Similar results are obtained from figs. 1b and 1c. There is no unique way to determine the average pulsewidth or pulse shape from $G^{(2)}_{obs}(\tau)$ if there is an unknown spread in pulsewidths.

It should also be noted that pulsewidth fluctuations can also affect lifetime measurements if the lifetime being measured is of the same order of magnitude as the pulsewidth. The magnitude of the effect depends on the particular experimental situation and most importantly on the nonlineaity of the interaction.

In order to experimentally determine the pulsewidth fluctuation, the ratio of the square of the fundamental intensity to the second harmonic intensity can be monitored as a function of time. This ratio is directly proportional to the pulsewidth if the spatial distribution is constant and the temporal pulse shape doesn't change (e.g. remains Gaussian) [8]. The variation in the ratio immediately determines the variation in pulsewidth. Monitoring this ratio from pulse to pulse also allows one to select the shortest pulses in the train (i.e. those with the smallest ratio). This type of pulse selection has been used successfully in the past on low repetition rate laser systems [9]. Such pulse descrimination would lower the effective repetition rate but could substantially increase the time resolution if the pulsewidth variations are substantial [10].

The author greatly appreciates fruitful discussion with J.-C. Diels and A. L. Smirl and wishes to thank P. Perryman for his assistance with computer programming. The support of the Faculty Research Fund of North Texas State University is gratefully acknowledged.

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