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Recieved 23 October 1984

A new time domain single shot conjugation autocorrelator is described. Three beams derived from the same initial laser pulse interact in a nonlinear medium to produce a backward signal wave in a 90° phase conjugate geometry. The pulse duration is determined from the spatial width of the phase conjugate beam emerging from the interaction region. Pulse duration measurements of pulses selected from a YAG laser train show that shorter pulses are generated toward the tail of the pulse train.

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## 1. Introduction

The time duration of a picosecond or subpicosecond laser pulse can be measured by a number of different techniques such as two photon fluorescence (TPF) [1], second harmonic generation (SHG) [2–4], and streak cameras [5]. These methods are limited by poor resolution and small S/N ratio (TPF), by phase-matching requirements (SHG), and by insufficient time resolution. The duration of a single laser pulse can be determined by measuring the spatial width of the phase conjugate beam generated from a nonlinear medium in a 90° geometry background free direction [6–9]. This time duration measurement of a single laser pulse is based on degenerate four wave mixing and therefore, wavelength independent. The range of the pulse duration that can be measured can be easily changed from subpicosecond to hundreds of picoseconds by adding an adjustable optical imaging system and by increasing the length of the nonlinear medium. In this paper, we describe a simple, compact, single shot, wavelength independent, time autocorrelator, which converts time information into spatial information with temporal resolution of a few tenths of a picosecond and has a temporal range extending over several hundreds of picoseconds. An important characteristic of this new technique is that, depending on the nonlinear medium, one can obtain information about the intensity or about the coherence correlation function of the laser pulse [10, 11].

## 2. Theoretical background

Fig. 1 shows a schematic diagram illustrating the operation of the phase conjugate autocorrelator. Two counterpropagating pulses, $E_1$, and $E_2$, overlap

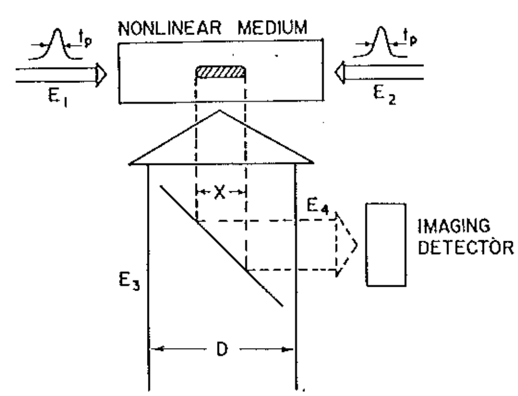


Fig. 1. Schematic diagram of 90° phase conjugate time domain process.

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exactly at the center of the nonlinear medium. A third probe beam, $E_3$, irradiates the overlap region of $E_1$ and $E_2$. All three beams, $E_1$, $E_2$, and $E_3$ have the same frequency and are derived from the same initial laser pulse. The interaction of the three beams produces a backward signal wave, $E_4$, at the same frequency. This latter beam is the phase conjugate beam. The nonlinear wave equation for the phase conjugate field, $E_4$, in the slowly varying envelope approximation form (SVEA) [6–8], is given by

$$[\partial/\partial z + (k/\omega)\partial/\partial t] E_4(r, t)$$
$$= \mathrm{i}(2\pi\chi^3/cn) E_1(r, t) E_2(r, t) E_3(r, t), \qquad (1)$$

where we assumed no depletion of $E_1$, and $E_2$, and $\chi^3(-\omega, \omega, \omega, -\omega)$ is the third order nonlinear susceptibility. The conditions for the buildup of a phase conjugate beam are:

$$k_1 + k_2 = k_3 + k_4 \qquad (2a)$$

for the wave vectors, and

$$\omega_1 + \omega_2 = \omega_3 + \omega_4 \qquad (2b)$$

for the frequencies. Both conditions are satisfied in the geometric arrangement shown in fig. 1. In this experiment, the input frequencies are degenerate, i.e.

$$\omega_1 = \omega_2 = \omega_3 = \omega_4 = \omega. \qquad (3)$$

Beams $E_1$ and $E_2$ propagate in opposite directions, such that $k_1 = -k_2$, resulting from eq. (2a) $k_3 = -k_4$. The energy distribution detected on a slow detector far from the interaction region is proportional to

$$I_4 \to \int_{-\infty}^{+\infty} |P^3(t)|^2 \, \mathrm{d}t, \qquad (4)$$

where

$$P^3(t) = \iiint \phi(t', t'', t''')$$
$$\times E(t') E(t'') E(t''') \, \mathrm{d}t' \, \mathrm{d}t'' \, \mathrm{d}t'''$$

is the third order polarization of the medium and $\phi(t', t'', t''')$ is the nonlinear response function.

In the framework of the SVEA approximation, one can consider the first pair of interactions as creating a real polarization change at time $t'$. This change, contributes to the polarization at a later time $t$, which is the source for the emergent radiation. Contributions to the polarization change must occur at all times $t'$, prior to $t$, and the response of the system, $A(t - t')$, determines the importance of earlier events. From this model, one has

$$P^3(t) \sim E_3(t) \int_{-\infty}^{t} E_1(t' + \tau) E_2(t') A(t - t') \, \mathrm{d}t'. \qquad (5)$$

Usually, the response of the medium, $A$, can be approximated by an exponentially decaying function with a characteristic relaxation time $\tau$.

Two experimental cases are possible. In the first case, the duration of the pulse is much greater than the relaxation time of the medium ($t_p \gg \tau_D$). In this regime, the response instantaneously follows the driving field. This corresponds to the following nonlinear response function;

$$\phi(t', t'', t''') = \chi^3 \delta(t - t') \, \delta(t' - t'') \, \delta(t'' - t'''), \qquad (6)$$

expressed in terms of the Dirac $\delta$ function. The energy distribution at the detector plane is given by

$$I_4 \sim E_3^2(r = \text{const}, t = \text{const})$$
$$\times \int_{-\infty}^{+\infty} E_1^2(t + \tau) E_2^2(t) \, \mathrm{d}t$$
$$\sim E_3^2 \int_{-\infty}^{+\infty} I_1(t + \tau) I_2(t) \, \mathrm{d}t. \qquad (7)$$

The detected signal corresponds to the *intensity correlation* of the pulse, assuming that $E_3$ is constant and uniform in time and space perpendicularly to the overlap direction.

In the second case, the duration of the pulse is much smaller than the relaxation time of the medium ($t_p \ll \tau_D$). In this case, the response cannot follow the excitation pulse and the medium reaction can be approximated by a constant. The energy distribution on the detector is given by

$$I_4 \sim E_3^2(r = \text{const}, t = \text{const})$$
$$\times \left| \int_{-\infty}^{+\infty} E(t' + \tau) E(t') \, \mathrm{d}t' \right|^2 \mathrm{d}t. \qquad (8)$$

This term is responsible for creating a temporal grating. In this case, the coherence of the excitation pulse is measured.

As pointed out above, the time distribution of a pulse in a 90° phase conjugate geometry (fig. 1) is transformed into a spatial distribution. The measurements of the time duration of the pulse will be accurate only if the geometrical length of the pump pulses, $L_p = ct_p$, is smaller than the probe beam cross section diameter, $D$, such as ($L_p < D/3$). For $L_p = D$ a more complicated analysis involving the temporal and spatial shapes of the different light pulses has to be used. A small cross section of the pump pulses (relative to the probe pulse) is also required to satisfy the assumption that the probe pulse $E_3$ is uniform in time and space in the direction perpendicular to the overlap plane. This will allow for the extraction of $E_3$ ($t$ = const, $r$ = const) from the integrals in eqs. (7) and (8).

Some of the factors which limit the resolution of the SGH method also affect the four wave mixing technique. These include temporal spreading of the pulse wave packet due to dispersion in the nonlinear medium, diffraction effects due to the finite size of the interaction region resulting in the broadening of the image on the detector, and finally the divergence of the beams and alignment problems. An experimentally achievable time resolution should be better than one tenth of a picosecond, by properly choosing the nonlinear medium and by imaging the signal with a telescope on a photo-diode array detector, i.e. reticon array.

### 3. Design of the phase conjugation time domain autocorrelator

The three beams must overlap both spatially and temporally. Certain geometrical conditions have to be fulfilled, as described in the previous section. The experimental arrangement shown in fig. 2 corresponds to the principle of the DFWM described earlier (see fig. 1). A beam splitter, BS, reflects a few percent of the laser beam to be used as the probe beam. The mirror, M, directs the probe, through a telescope, toward the nonlinear medium. The front surface of the liquid cell or the nonlinear material should be slightly misaligned so that direct reflections will not be detected. For a phase conjugate reflected beam, the telescope, or the subtractive prism expander, works as an optical inverter.

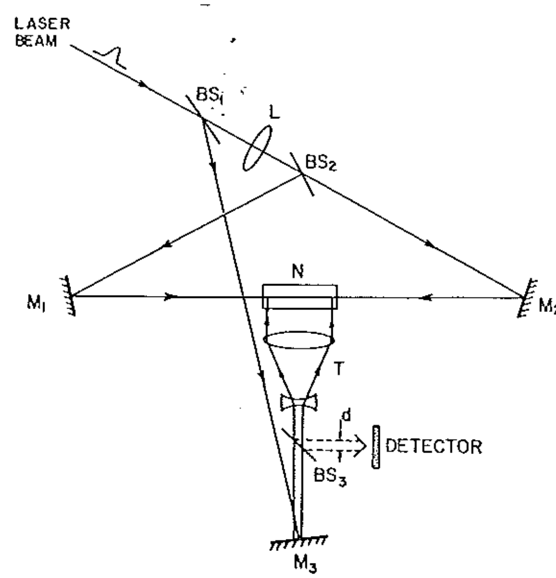


Fig. 2. Design of phase conjugate time domain autocorrelator. $M_1$, $M_2$, $M_3$, broad band mirrors, $R$ = 100%. $BS_1$ 4% beamsplitter. $BS_2$ 50/50 beamsplitter. L focusing lens, $f$ = 80 cm. T telescope. N nonlinear medium.

The diameter of the phase conjugated reflected beam is measured by a one dimensional imaging detector (photographic plate, video system) so that the duration of the pulse may be expressed by:

$$t_p = \delta d / c_{01},$$

where $d$ is the length of the interaction region calculated from the diameter of the phase conjugate beam, $c_{01}$ is the group velocity of the pulse in the nonlinear medium, and $\delta$ is a coefficient which value depends on the shape of the pulse (for a gaussian pulse, $\delta$ = 1.44).

The two pump beams are focused into the nonlinear medium to decrease the transverse dimension of the interaction region and increase the nonlinear response. The optical path of the probe beam must be carefully adjusted and set equal to the optical path of the pump beams to assure temporal and spatial overlap. The autocorrelator was calibrated by introducing known optical delays on the path of one of the pumping beams and by observing simultaneously, the movement of the peak of the trace on the imaging detector. For example, if the thickness of a glass plate, introduced in the path, is $s$ with a group index of refraction $n_g$, the shift of the peak of the autocorrelation trace will be

$$t = n_g s/2c,$$

where $c$ is the velocity of light in vacuum.

## 4. Experimental results

A time domain phase conjugation autocorrelator has been built and tested according to the design shown in fig. 1. The technique is background free, wavelength independent, and there is no need for phase matching as in the SHG method. The geometry is independent of the laser wavelength and of the nonlinear medium used in the experiment. A temporal resolution of 100 fs and a range of up to a few hundred picoseconds can be achieved. Fig. 3 shows an oscilloscope picture of the laser pulse correlation of a single pulse from a train of pulses emitted by a Quantel YAG laser. The four wave mixing signal was obtained using the experimental arrangement of fig. 2, with liquid $CS_2$ as the nonlinear medium. The relaxation time of $CS_2$ is about 2 ps and is much shorter than the duration of the YAG laser pulse. Therefore, the intensity correlation function was measured and pulse duration was calculated according to eq. (7).

The measurements were made with the second harmonic of the laser beam at 530 nm. The second harmonic laser pulse was focused by an 80 cm focal length lens into the 4 cm $CS_2$ cell. The energy of the laser pulse, before entering the autocorrelator was 0.5 mJ. About 4% of the pulse was then reflected by a beam splitter, expanded by a 4X telescope, and used as the probe beam. The diameter of the probe beam (~4 cm) was much later than the geometrical length of the overlapping pump pulses; therefore, no complicated analysis was needed to determine the pulse duration $t_p$. This was accomplished by using eq. (3). The diameter of the phase conjugated beam is decreased by the same factor 4X by the telescope. The reflected beam is then analyzed with a 1 cm long reticon diode array with a 25 μm diode to diode spacing, allowing a time range of 100 ps with a 300 fs resolution.

Fig. 3 shows the time duration of pulses selected at different positions from a train of pulses generated by a Quantel YAG laser. The pulses extracted from the beginning had a duration of about 20 ps, near the maximum they were about 30% longer, while the 6th to 8th pulse was consistently in the 12–15 ps range

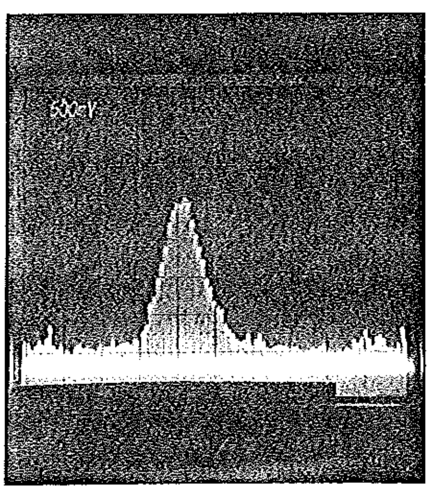

Fig. 3. Laser pulse correlation of a 530 nm single pulse emitted by a YAG laser. Oscilloscope photograph of a spatial profile from the diode array imaging system of the time domain autocorrelator.

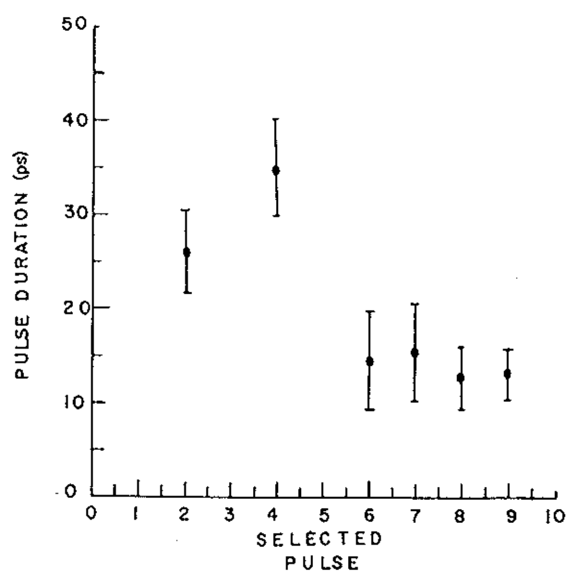


Fig. 4. Pulse duration versus position of a selected pulse from a YAG laser pulse train at 530 nm.

with an energy per pulse about half of the energy of a pulse selected near the maximum of the train. The pulse to pulse variation in duration in the pulse train most likely arises from competition between mode locking and self phase modulation in the YAG crystal, dye, and cavity elements. One can speculate that the spectral width of the pulses at the beginning of the mode-locked train is narrower compared to the width of later pulses which are broadened by self phase modulation. It is also possible that at the beginning of the train only a fraction of the possible modes of the pulse are locked. Both effects will result in longer pulses. For pulses selected after the peak of the train the spectral content can reach the 6 $cm^{-1}$ maximum spectral width allowed by the gain profile and more modes can be coupled. This leads to the generation of shorter pulses toward the end of the train. The small 6 $cm^{-1}$ gain profile limits the spectral width and pulse duration to 2 ps. Recently, pulses as short as the 2 ps theoretical limit for a YAG laser were reported by Clark et al. [12] for pulses at the end of the pulse train.

## 5. Conclusion

We have described a time domain phase conjugate autocorrelator for the measurement of ultrafast laser pulses. The autocorrelator is wavelength independent and has a range of hundreds of picoseconds with a subpicosecond resolution. It was used to determine the duration of pulses selected at different positions from a train of pulses generated by a YAG laser. These measurements confirm that shorter pulses are generated toward the tail of the train in YAG lasers.

This research was supported in part by Air Force grant AFOSR #84-0144.

## References

[1] J.A. Giordmaine, P.M. Rentzepis, S.L. Shapiro and K.W. Wecht, Appl. Phys. Lett. 11 (1967) 216.
[2] M. Maier, W. Kaiser and J.A. Giordmaine, Phys. Rev. Lett. 17 (1966) 1275.
[3] J. Janszky, G. Corradi and R.N. Gyuzafdian, Optics Comm. 23 (1977) 293.
[4] C. Kolmender, W. Zinth and W. Kaiser, Optics Comm. 30 (1979) 453.
[5] E.K. Zavoiskii and S.D. Fanchenko, Sovietski Physitcheskii Doklady 1 (1956) 285;
N.S. Schiller, Y. Tsuchiya, E. Inukuka, Y. Suzuki, K. Kinoshita, K. Kamiya, H. Iida and R.R. Alfano, Optical Spectra 15 (1982) 56.
[6] R.A. Fisher, B.R. Suydam and B.J. Feldman, Phys. Rev. A23 (1981) 3071.
[7] T.R. O'Meara and A. Yariv, Opt. Eng. 21 (1982) 237.
[8] Optical phase conjugation, ed. R.A. Fisher (Academic Press, New York, 1983).
[9] J. Janszky, G. Corradi and D.S. Hamilton, Appl. Optics 23 (1984) 8.
[10] B.S. Wherrett, A.L. Smirl and T.F. Boggess, IEEE J. Quantum Electron. QE-19 (1983) 680.
[11] H.J. Eichler, U. Klein and O. Langhaus, Appl. Phys. 2 (1980) 215.
[12] H. Nathel, D.M. Guthals and J.H. Clark, Conf. on Lasers and electro-optics, Th144, 182 (1984) and private communication.