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A NEW APPLICATION OF POLYMER DISPERSED LIQUID CRYSTALS: MEASUREMENTS OF ULTRASHORT LIGHT PULSES WITH FEMTOSECOND ACCURACY

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Abstract The self-diffraction technique has been applied to measure subpicosecond pulsewidth using Polymer Dispersed Liquid Crystals as nonlinear materials. The experimental data agree with the ones obtained by conventional autocorrelation technique thus pointing out the advantages of cheap and handy PDLC over commonly used high optical quality nonlinear crystals.

INTRODUCTION

Recently¹, we have demonstrated that nematic liquid crystals and polymer dispersed liquid crystals (PDLC's) can be efficiently used to measure light pulse duration in the picosecond range by inducing in these materials the formation of a transient thermal grating with consequent self-diffraction of the incoming laser beams. In this paper we show that it is possible to use PDLC's to reach at least a sensitivity of about twenty femtoseconds.

This technique² involves the interaction of two laser beams in a nonlinear medium leading to self diffraction of the beams themselves. In order to measure the light pulse duration, the laser beam is splitted (amplitude sharing) to get two coherent waves able to interfere in the nonlinear medium where they cross each other. The

time coherence between them is controlled by a variable delay which can be introduced by appropriate reflectors. In presence of a nonlinear optical response of the medium they give rise to a transient phase grating, able to produce diffraction of the two beams and the efficiency of the diffraction is measured as the ratio between the diffracted intensity vs the impinging one: $\eta = I_d/I_0$.

Usually, measurements are taken on the first order diffraction beam, which is the most intense one. By changing the optical delay between the two interacting beams one can vary the diffraction efficiency which will be maximum for zero delay (corresponding to complete overlap of the two laser pulses) and it will be zero for a delay bigger than the pulse duration since in this case no overlap of the two pulses is obtained. It has been proved that diffraction efficiency is related to the short pulses fourth-order coherence function and its measurement vs the pulses delay can provide both the pulse duration τ_p and the coherence time τ_c . This method has been successfully applied using semiconductor films and dyes as nonlinear materials; anyway, in these case the data needed to be corrected since the fast relaxation time of used materials affected the experimental observations. On the other hand this technique was shown to be very reliable when using media with nonlinearities much slower than the pulse duration³ since in this case material relaxation doesn't affect the measurements. Moreover this technique has some advantages over the conventional second order autocorrelation which exploits second harmonic generation (SHG) in nonlinear crystals with a Michelson-type interferometric geometry: it is broadband, background free and easy to align.

THEORETICAL BACKGROUND

In the following we assume to deal with a Fourier transformed pulse (pulse duration $\tau_p \approx$ coherence time τ_c). The time dependence of the pulse electric field can be expressed as⁴:

$$E(t) = E A(t) u(t) \quad (1)$$

where E is a constant amplitude,

$$A(t) = \exp\{-2\ln[t^2/\tau_p^2]\} \quad (2)$$

Being τ_p the pulse width and $u(t)$ a statistical factor which takes into account the phase of the field. When the beam is split and arranged to produce a grating, the diffraction efficiency becomes proportional to a product of four radiation fields:

$$\eta \propto \frac{1}{\tau_p^2} \int_{-\infty}^t A(t_1)A(t_2+\tau_d)A(t_2)A(t_1+\tau_d)f(u)dt_1dt_2 \quad (3)$$

where τ_d is the delay time between the two interacting pulses and

$$f(u) = \langle u(t_1)u(t_2+\tau_d)u^*(t_2)u^*(t_1+\tau_d) \rangle \quad (4)$$

is the cross correlation function of the four fields.

For very long relaxation time $t \rightarrow \infty$, $f(u)$ becomes the fourth order coherence function of the radiation field $\Gamma^{(4)}$ and the diffraction efficiency becomes:

$$\eta \propto \exp\{-2\ln[t^2/\tau_p^2]\} \quad (5)$$

Therefore as a function of the delay time τ_d , η has a Gaussian shape with full width at half maximum given by

$$\Delta = \sqrt{2}\tau_p \quad (6)$$

As a consequence, a pulsewidth measurement can be obtained by detecting the relative intensity of the first diffracted beam vs the delay time between the two pulses. We have already demonstrated that it can be done with light pulses in the picosecond range by using a liquid crystalline sample as a nonlinear medium which is really inexpensive if compared with high quality polished nonlinear crystals. Moreover in the latter case the extension to the measurement of ultrashort light pulses would introduce further limitations because of pulse dispersion occurring into the crystals, thus requiring very thin expensive and fragile samples to obtain femtosecond accuracy. For this reason the usual small thickness of PDLC samples becomes an advantage to carry out measurements in the femtoseconds scale.

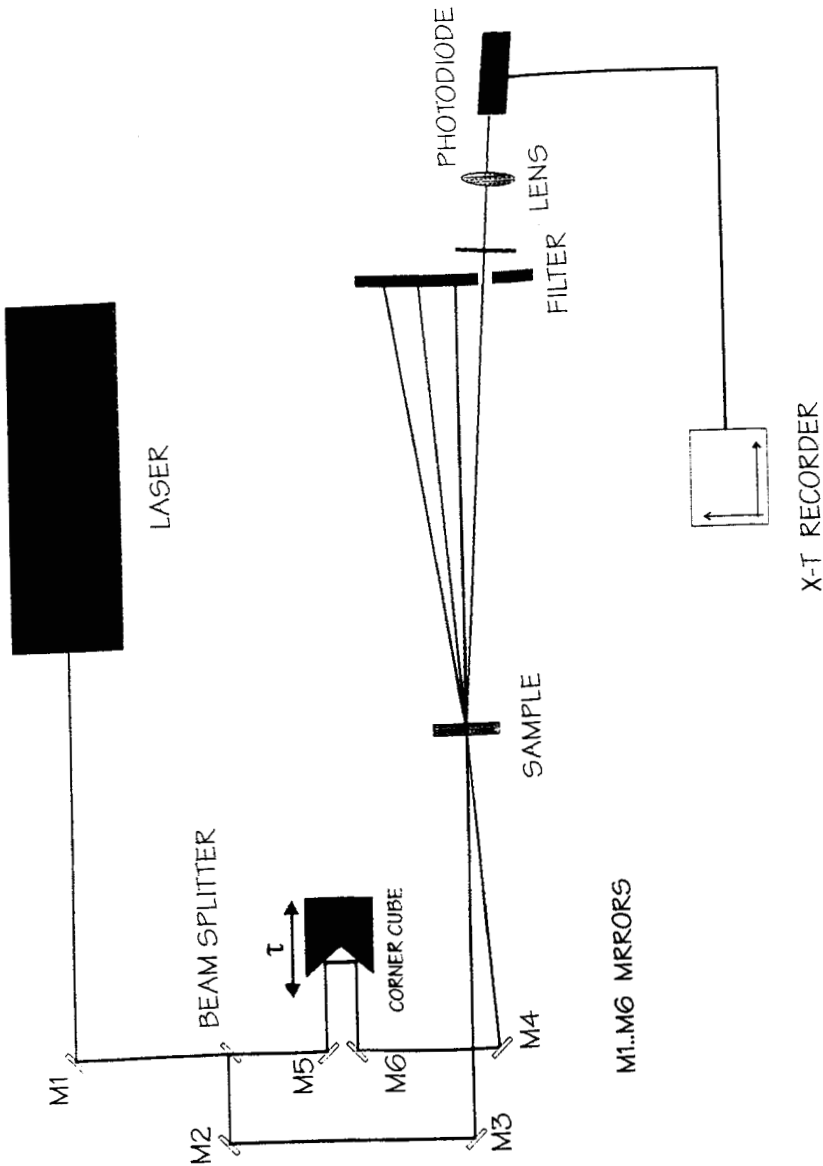


FIGURE 1. Sketch of the experimental setup

EXPERIMENT AND RESULTS

The experimental setup is sketched in Fig.1. The source of the ultrashort light pulses is a dye laser synchronously pumped by a mode-locked Nd-YAG laser at a repetition frequency $\Omega = 76$ MHz. After a first mirror we have a "Mach-Zender" type interferometer where one of the arms has a variable length controlled by an actuator. This moves the retroreflector corner cube CC thus introducing a variable delay between the two beams which cross on the PDLC sample with an angle θ . This angle must be made small (0.5°) in order to ensure a good superposition of the beams, thus getting the optimal condition for the onset of the dynamic grating. The first diffracted beam is then detected after an iris diaphragm (which blocks all the other beams) by using an additional filter to increase the signal to noise ratio. The pulse duration has been previously tested by a conventional interferometric SHG measurement, by placing a mirror to pick up the beam just after the laser cavity - dumper. This measurement, which is shown in Fig.2, gives a pulse duration $\tau_p = 425 \pm 33$ fsec. It has been proved that also in this configuration the measured Gaussian curve has a width which is $\sqrt{2}$ times the actual width⁵.

Measurements were taken using a $36 \mu\text{m}$ thick PDLC sample obtained by PIPS method starting from the nematic liquid crystal K15 (by BDH) and the epoxy fluid prepolymer EPON 815. A small quantity of dye (0.1%) was added to the liquid crystal to enhance the sample absorption. No low frequency voltage was applied to the sample and therefore the self - diffraction effect took place above the threshold intensity for self - transparency; anyway the application of a suitable bias voltage to switch the sample to a partially transparent state could be suitable to lower the minimum pulse energy required for the onset of self - diffraction⁶. The nonlinear grating was originated by thermal effects like in other experiments reported by some of us.

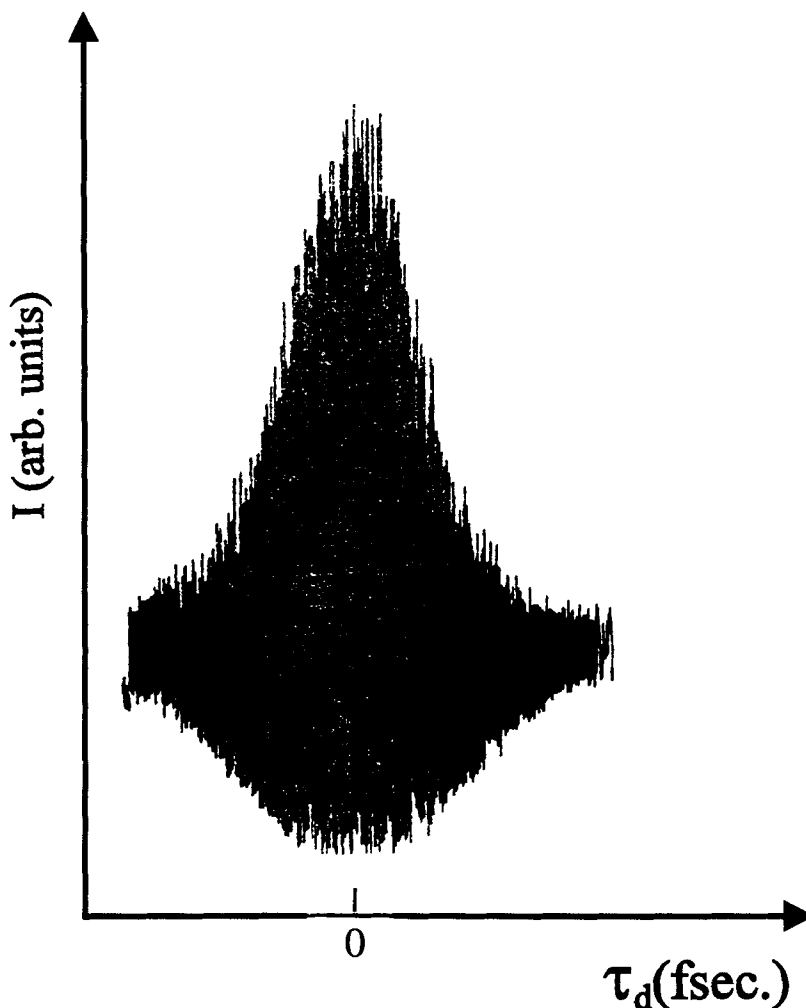


FIGURE 2. Shape of the signal obtained by conventional interferometric second harmonic generation technique

A typical measurement obtained using this technique is shown in Fig.3, where the diffracted signal is recorded vs the time delay τ_d . According to the presented analysis, it gives a pulse duration $\tau_p = 450 \pm 17$ fsec which is in good agreement with the value obtained by means of the conventional autocorrelation method. This result increases the interest of the self - diffraction method with the use

of nonlinear materials like PDLC's. Indeed, this technique becomes particularly reliable and easy to use in the subpicosecond range where the maximum arm travel necessary to achieve the required delay is of the order of few hundreds of micrometers: as a consequence it is easy to maintain a good alignment of the Mach - Zender interferometer along this length variations and the complete scanning doesn't take too long. Moreover the actual maximum sensitivity which can be obtained by using PDLC's may be higher than the one we got in the presented experiment since the experimental error is here given by the step

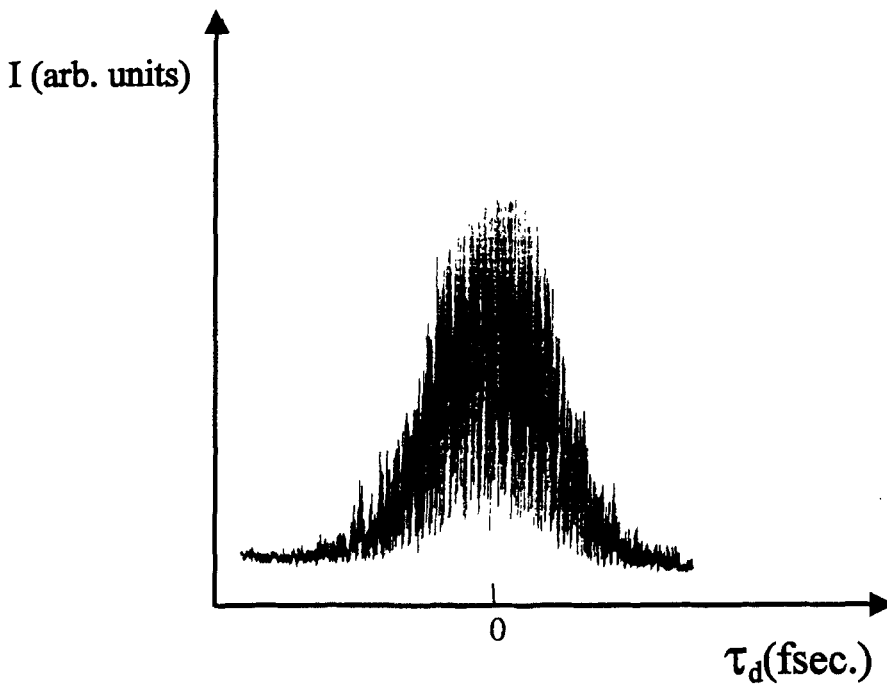


FIGURE 3. Intensity of the diffracted signal recorded vs the time delay τ_d

inducing the optical path variation in the interferometer arm, which can certainly be improved. On the other hand a shorter laser pulse would be needed to investigate the intrinsic limitation of PDLC's as nonlinear materials for this application.

As we have remarked above, an increasing sensitivity is reached in nonlinear crystals by making them very thin with, at the same time, a rise in cost and in the brittleness of the sample. On the contrary a PDLC sample is usually made thin (10 to 30 μm), cheap and can be sustained by low index small glasses thus getting a sample which is difficult to break and keeping low the intrinsic frequency dispersion.

A further step in the study of this method will be the investigation of frequency chirped ultrashort light pulses which will give a complete temporal and phase information on the laser beam in a way that is alternative to the interferometric SHG autocorrelation technique.

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