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ABSTRACT

We present a holographic recording technique with 150 femtosecond time resolution. This technique allows us to capture either a single hologram with fine spatial resolution (4μm), or a time-sequence of multiple holograms with reduced spatial resolution in a single-shot experiment, while preserving amplitude and phase information. The time resolution and the frame rate are limited only by the duration of the laser pulses. The holograms are recorded on a CCD camera and digitally reconstructed. We have used the technique to study the nonlinear propagation of high energy femtosecond pulses through liquids. We have observed dramatic differences in the pulse propagation characteristics depending on the strength of the nonlinear coefficient of the material and it's time response. The fine spatial resolution allows us to zoom in and visualize the spatial profile of the pulses breaking up into multiple filaments while the phase recovered from the holograms helps us identify the nonlinear index changes in the material. We have measured both positive and negative index changes. Very fast positive index changes are generally attributable to the Kerr nonlinearity. The negative index changes can be caused by electron plasma generated by multiphoton absorption.

Keywords: femtosecond, digital holography, pulsed holography, plasma imaging.

1. INTRODUCTION

The nonlinear interaction of light pulses with the medium in which they propagate gives rise to many complex and interesting phenomena[1]. The formation of long range filaments,[2] white light generation,[3] optical turbulence,[4] multiple filamentation[5] and plasma generation[2] are amongst the observed phenomena. Different techniques have been used to observe pulse propagation in different materials[6,7]. Here we present a holographic recording technique[8] which allows us to capture nonlinear pulse propagation with 150 femtosecond time resolution. This technique allows us to capture either a single hologram with fine spatial resolution (4μm), or a time-sequence of multiple holograms (holographic movie) with reduced spatial resolution in a single-shot experiment, while preserving amplitude and phase information. We use a simple on-axis holographic setup which does not require a separate reference pulse or a pulse shaper to generate a pulse train.

The single hologram technique allows us to observe the beam filamentation with good spatial resolution, while the multiple hologram technique allows us to observe the time evolution in a single experiment. The time resolution and the frame rate are limited only by the duration of the laser pulses. The on-axis holograms are recorded on a CCD camera and digitally reconstructed. On axis holograms in general suffer from the twin image problem due to the loss of phase information when the hologram is captured. In our case, the size of the objects that we observe is small compared to the size of the recording medium (the CCD chip). This allows us to achieve good reconstructions of the object using an iterative technique.

We have used our technique to study the propagation of femtosecond pulses through liquids with strong and weak nonlinearities. Carbon disulfide (CS_2) has a large third order nonlinear coefficient ($\chi^{(3)}$), while the nonlinear response in water is much weaker[9]. The nonlinearity in CS_2 has a response time of about two picoseconds. In water the response time is much shorter than the pulse duration, thus it can be considered instantaneous. We compare our results of the pulse propagation experiments in these two materials.

2. EXPERIMENTAL SETUP

2.1. Single hologram setup

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Figure 1 shows the setup used to record an on-axis hologram with a femtosecond pulse. A pulse from a Ti:sapphire laser amplifier operating at 800nm wavelength is used to generate the ultrafast event and also to record it. The laser pulses have a duration of 150 femtoseconds and a maximum energy of 2mJ. The pulse is split in two, with a major portion of the energy going into the pump beam. A delay line is used to synchronize the arrival of the pump and probe pulses. The pump beam is focused inside the sample with an achromatic lens (L3). The probe propagates in a direction perpendicular to the pump and captures the interaction of the pump with the material. The image is magnified by a factor of $M = f_2 / f_1$. A CCD camera is placed at a distance L from the image plane to capture an in-line (Gabor) hologram[10]. The digitized hologram is then numerically reconstructed to retrieve the phase and amplitude changes induced in the probe by the nonlinear changes in the material. The time resolution of the holograms is limited by the duration of the pulses (150 femtoseconds). The spatial resolution of 4μm is limited by the numerical aperture of the hologram.

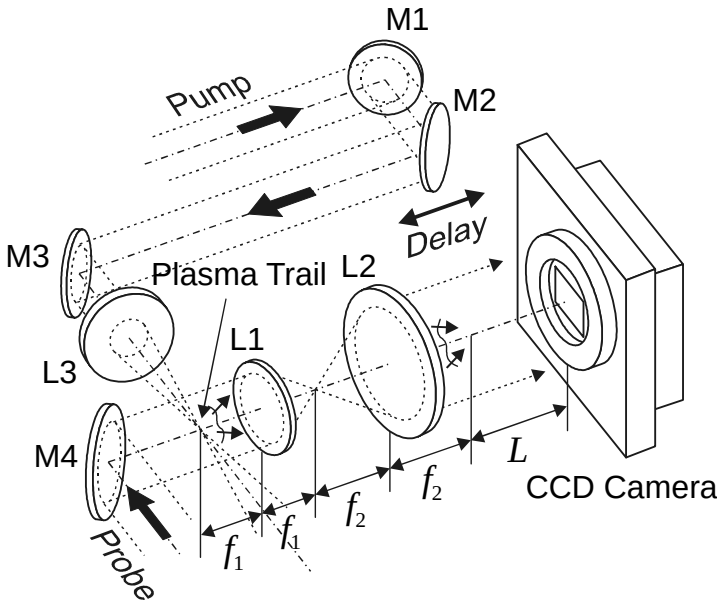

Figure 1. Experimental setup for on-axis femtosecond holography. A laser pulse is split into a pump (high energy) and a probe (low energy). The probe is used to capture a hologram of the pump pulse traversing the material. M = mirror, L = Lens.

2.2 Movie Setup

The setup in Figure 1 was modified to record a time-sequence of four holograms with a single laser pulse. Mirror M4 is replaced with a mirror array consisting of four mirror segments (Fig. 2), each of which has independent controls for angular and axial displacements. Four probe pulses are generated by reflecting a single pulse off the mirror array. The position of the mirrors controls the relative time delay between them, while the angle controls the propagation direction. The four probes are made to spatially overlap in the interaction region (and then spatially separate on the recording plane). The relative time delay can be adjusted to match the time window of interest. Each probe pulse samples the event at a time set by the displacement of the mirror. The angle between the pump and probe pulses is reduced to 30 degrees in order to increase the region of interaction of the pump and probe pulses. The four spatially multiplexed holograms are captured with a CCD camera. The variable delay line synchronizes the probe pulses to the arrival of the pump pulse. We set the separation angle between the probe pulses sufficiently small (<2º) so that the events are captured at approximately the same angle. In order to spatially separate the four holograms in the CCD sensor, the effective angular aperture of each individual hologram is limited to the separation angle between the probe pulses.

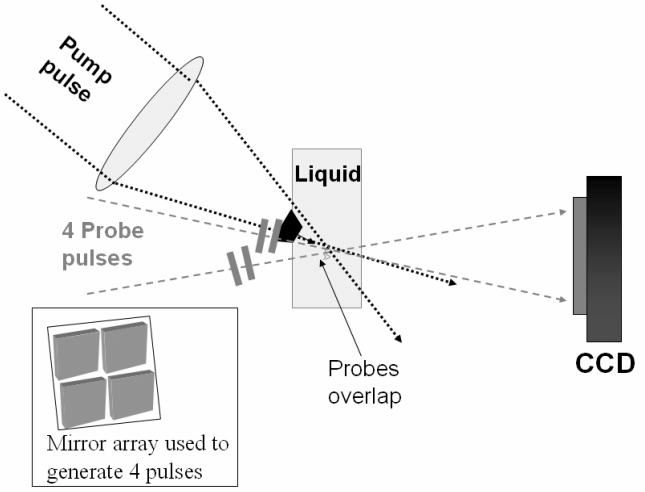

Figure 2. Single shot capture of 4 time-delayed holograms. The probe pulse is split into four time-delayed probes using a mirror array.

3. RESULTS

3.1 Single hologram

We have captured holograms of the interaction of high energy pulses with water and CS_2. A probe pulse is synchronized to spatially and temporally overlap with the propagation of a strong pump pulse in the material. The on-axis holograms are reconstructed digitally to recover the amplitude and phase of the probe pulses. Figure 3 shows the amplitude and phase reconstructions. Figure 3 shows the amplitude(a) and phase(b) reconstruction for a 700μJ pulse propagating in water, while c) and d) are the amplitude and phase reconstructions for a 50μJ pulse propagating in CS_2. Different pulse energies were used because the nonlinearities are much stronger in CS_2.

In water, the beam envelope follows the linear focusing of the lens, while inside the beam breaks up into multiple filaments. The filamentation of the beam is caused by self phase modulation through the Kerr effect. As the pulse goes through it leaves a trace in the material. In figure 3-a) the pulse is at the leading edge, while the very dark region to the left is the trace left over in the material. The trace is characterized by a decrease in transmission of about thirty percent with very small phase changes. In contrast, the filaments at the leading edge have large negative phase changes and small amplitude changes. The size of the filaments is approximately 5μm with a maximum index change on the order of $\Delta n = -10^{-2}$. We attribute this negative index change to the formation of a plasma in the regions of highest intensity of the beam. The dark trace seems to be the result of many more filaments forming and scattering or absorbing light from the probe pulses. If the leading edge is observed just two picoseconds after the pulse has gone through it also becomes a dark trace with many filaments. It is not clear to us at this time what the mechanism is that drives this very fast change from a few to many filaments. From further pump and probe experiments we were able to estimate the lifetime of the dark trace to be 0.5 nanoseconds, much longer than the time it takes to generate it.

In CS_2 we see a trace that is much shorter than in water. Also, we see a positive phase change and an increase in intensity near the center of the pulse position. The phase change corresponds to a positive index change generated by the pump pulse through the Kerr effect. The Kerr coefficient is much stronger in CS_2 than in water, thus no positive index change was observed in water. As opposed to water, no plasma was observed in CS_2, possibly because the intensity is not as high as in water. For highly nonlinear materials (like CS_2), if the pulse energy is increased the beam very quickly breaks up into multiple filaments before reaching the focus. In general, higher intensities can be achieved in materials with weaker Kerr nonlinearity. In the reconstructed phase (Fig 3-d) we can also see multiple filaments, with

a diameter of 8-15μm. The maximum index change in the filaments is $7x10^{-4}$. The duration of the trace is determined by the lifetime of the Kerr effect in CS2, which is about 2 picoseconds[9]. The increase in intensity near the center of the pulse (Fig 3-c) is due to a focusing effect caused by the positive index change. The region of nonlinear index change acts as a focusing lens for the probe light. We now present the results on the dynamics of the pulse propagation.

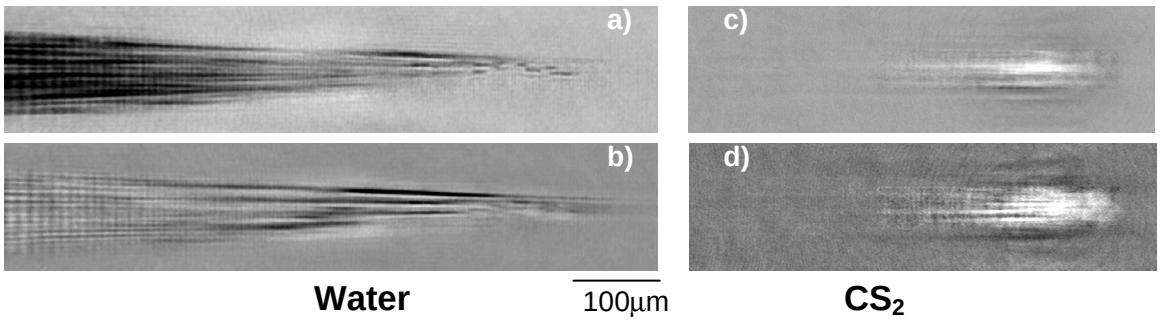

Figure 3. Digital reconstructions of amplitude and phase. a-b) are the amplitude and phase reconstructions for pulse propagation in water. c-d) are the amplitude and phase reconstructions for pulse propagation in CS_2. The maximum phase change is -0.5 radians in b) and 0.05 radians in d).

3.2 Movies

We record a sequence of four holograms which allows us to observe the time evolution of the pulse propagation in a single shot. The angle between the pump and probe pulses is reduced to 30 degrees. The longer interaction length increases the signal strength and allows us to observe the index change due to the Kerr effect in both water and CS_2. A 150 femtosecond pump pulse with 50μJ energy is focused inside a glass cuvette filled with CS_2. In Figure 2-a) we see the reconstructed phase change for four different time snapshots, t = 0, 1, 2 and 3 picoseconds. Water has a weaker nonlinear response so the pulse energy was increased to 300μJ. Figure 2-b) shows the reconstructed phase change for time delays of 0, 0.7, 1.3 and 2.3 picoseconds. The propagation speed is different in the two cases because CS_2 has a higher index of refraction (n = 1.63). In the first image the probe pulse interacts only with the leading edge of the pulse, so a weak signal is recorded. The second and third holograms capture the pulse before the focal point, while the fourth one captures the pulse after the focal point. The Kerr effect in CS_2 has both instantaneous and non-instantaneous contributions, so as the pulse traverses the liquid it leaves a trace of index change. Using longer time delays between the pump and the probes we measured a decay time constant of the non-instantaneous index change of 1.7 picoseconds, in agreement with the values reported in the literature.[9] The nonlinear index change in water is much faster than the duration of the pulse, so instead of a trace we get a direct snapshot of the pulse. The maximum phase change was 2 radians for CS_2 and 0.6 radian for water. These correspond to index changes of $3x10^{-4}$ in CS_2 and $1x10^{-4}$ in water. The index change measured in CS_2 is lower than the previous value due to the lower resolution. In the previous experiment we were able to measure the local maximum at a filament, while here it is averaged over a larger region. Note that even though the pulse energy was 5 times higher in the water, the index change in CS_2 greater. The positive index change measured in water is due to the Kerr effect, in contrast to the negative index change due to plasma generation. The phase reconstruction in water also shows a weak trail, but in this case it is a negative index change which corresponds to the long lasting plasma trail observed with the high resolution holograms. With this setup the positive index change due to the instantaneous Kerr effect dominates over the negative index change due to plasma generation. The long interaction region causes a higher phase change for the Kerr effect, which occurs everywhere the pump and probe pulses overlap. In contrast, the strong negative index change is located in very small filaments which cannot be resolved with the movie setup.

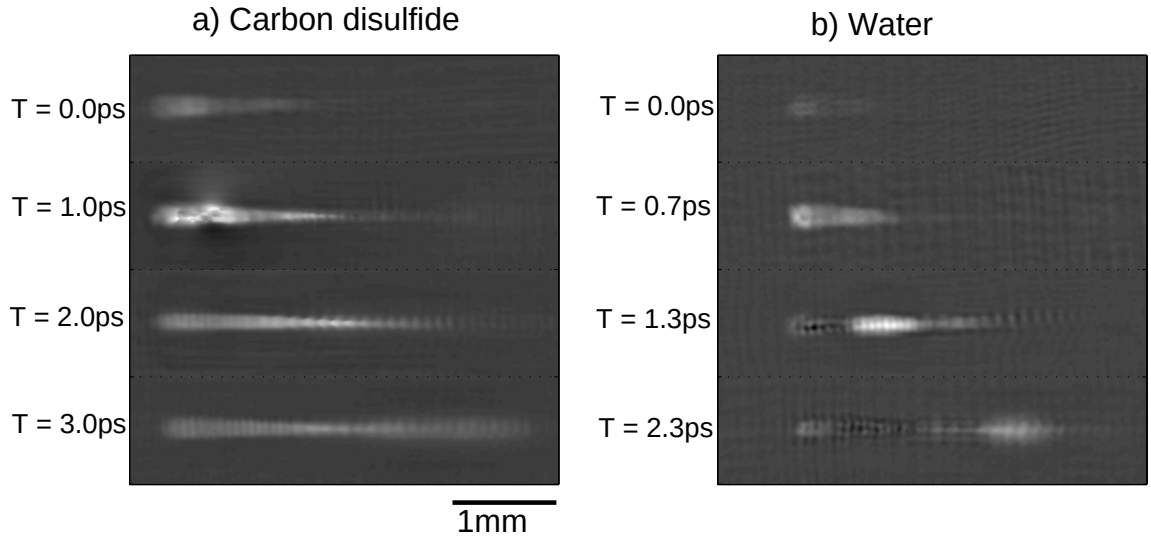

Figure 4. Holographic phase reconstructions of femtosecond pulse propagation in a) CS_2 and b) water. The maximum phase change in a) is 2 radians and 0.6 radians in b). The time delay between the holograms is indicated in the figure. Each sub-frame is 4mm(horizontal) by 1mm(vertical).

4. CONCLUSION

We presented a simple holographic recording technique which allows us to capture ultrafast nonlinear index changes with 150 femtosecond time resolution and 4μm spatial resolution or a time-sequence of four holograms (holographic movie) with reduced spatial resolution. Femtosecond pulses are used to record on axis holograms on a CCD camera. We have observed the propagation of high energy femtosecond pulses in liquids and compared the results for pulse propagation in water and carbon disulfide. In water we have observed a positive index change due to the Kerr effect and a negative index change due to the formation of plasma filaments. In CS_2 we observed a positive index change due to the Kerr effect and no plasma generation.

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REFERENCES

1. A. J. Campillo, S. L. Shapiro, and B. R. Suydam, "Periodic breakup of optical beams due to self-focusing", Appl. Phys. Lett. 23, 628 (1973).
2. A. Couairon, S. Tzortzakis, L. Berge, M. Franco, B. Prade, A. Mysyrowicz, “Infrared femtosecond light filaments in air: simulations and experiments”, J. Opt. Soc. Am. B, 19, 1117 (2002).
3. R. Fork, C. Shank, C. Hirlimann, R. Yen, W Tomlinson, “Femtosecond white-light continuum pulses”, Opt. Lett, 8 (1983).
4. M. Mlejnek, M. Kolesik, J. V. Moloney, and E. M. Wright, "Optically Turbulent Femtosecond Light Guide in Air", Phys. Rev. Lett. 83, 2938 (1999).
5. L. Berge, S. Skupin, F. Lederer, G. Mejean, J. Yu, J. Kasparian, E. Salmon, J. P. Wolf, M. Rodriguez, L. Woste, R. Bourayou, R. Sauerbrey, "Multiple filamentation of Terrawatt laser pulses in air", Phys. Rev. Lett. 92, 225002 (2004).
6. M. Fujimoto, S. Aoshima, M. Hosoda, and Y. Tsuchiya, “Femtosecond time-resolved optical polarigraphy: imaging of the propagation dynamics of intense light in a medium”, Opt. Lett. 24, 850 (1999).

7. H. Schroeder and S. L. Chin, "Visualization of the evolution of multiple filaments in methanol", Opt. Comm. 234, 399 (2004).
8. M. Centurion, Y. Pu, Z. Liu, D. Psaltis, T. W. Hänsch, “Holographic recording of laser-induced plasma”, Opt. Lett 29, p772 (2004).
9. R Boyd, Nonlinear Optics, Academic Press, 2003.
10. D. Gabor, “A new microscopic principle”, Nature 161, 777 (1948).