In-situ measurement and the reconstruction in 3D of femtosecond inscription induced complex permittivity modification in glass.

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We demonstrate a new approach to in-situ measurement of femtosecond laser pulse induced changes in glass enabling the reconstruction in 3D of the induced complex permittivity modification. The technique can be used to provide single shot and time resolved quantitative measurements with a micron scale spatial resolution.

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1. Introduction

Femtosecond laser pulse induced changes in dielectric materials have become a topic of much interest recently for the direct writing of photonics microstructures. The physical processes which take place on time scales comparable to and longer than the duration of the laser pulse are not well understood. There is a need for improved online diagnostics to measure the induced changes in time in order to enhance the understanding of the processes involved and to enable improved control during device fabrication.

There are a number of high spatial resolution imaging techniques in use which provide only a qualitative indication of induced refractive index distribution including pump-probe transmission microscopic imaging,[1] time-resolved photography and ultrafast transient absorption spectroscopy,[2] DIC-microscopy[3] and time-resolved phase contrast microscopy.[4,5] Techniques which can provide quantitative time-resolved plasma diagnostics on small volumes include a classical (Michelson) interferometric approach,[6] shadowgraphy,[7] quantitative phase microscopy (QPm)[8] and digital holographic microscopy (DHM).[9,10] The current state of the art technique for measuring the profile of refractive index (or electron density) use a classical interferometer followed by Fourier filtration of fringes or the inverse Abel transform (e.g.[6]). It can provide only qualitative measurements in real time from the interferometric fringes. Shadowgraphy[7] requires low-aperture imaging optics with consequent insufficient spatial resolution for the typical micro-fabrication applications. Quantitative phase microscopy (QPm)[8] demands a complex system with multiple cameras is suitable for ultrafast probing but only for the case of a thin non-absorbing object. Determination of accumulated phase is relatively straightforward but reconstruction of the refractive index from a single map of retarded phase requires additional computations and involves a priori information on symmetry of the refractive index distribution. The quantitative techniques mentioned above rely on paraxial and geometrical optics approximations that may not be applicable when a high numerical aperture (NA) is used to achieve a high spatial resolution, and on the assumption that the object is non absorbing at the wavelength of the probing laser which may not be true if the plasma density approaches an appreciable fraction of the critical density.

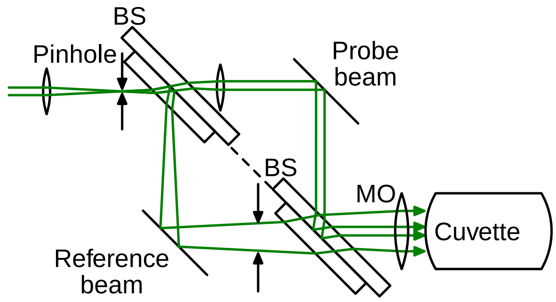

Fig. 1. (Colour online) Experimental arrangement for in-situ holographic measurement

2. Technique

Particularly relevant to the work carried out here is Digital Holographic Microscopy (DHM) which can be used at resolutions up to the diffraction resolution of the optics.[10] This method involves using a Mach-Zehnder geometry to record an off-axis interferogram (hologram) between a reference beam (which typically has a plane wavefront) and the object scattered beam. The interferogram may be digitally captured at high speed. Computer reconstruction based upon the Fresnel diffraction integrals (in scalar approximation) enables reconstruction of the wave front just after transmission of the probe beam through the object and this can be related to the object permittivity.[11] The approximation requires that the structures be thin and be placed near the focal plane of a high NA objective - effectively the object is treated as having negligible optical depth and the accumulated complex-valued transmittance is measured. Additionally it is necessary to install the same optics into both reference and scattered paths to minimise the effects of optical aberrations so that diffraction limited resolution can be approached.

The technique discussed here overcomes the limitations of previous DHM measurements. Firstly a simplified optical layout (figure 1) is used where the reference and probe beams traverse the same optical path, and the reference and scattered beams match both wavefront and polarisation at the detector plane. On the basis of this the theoretical analysis[12] was re-examined in the first order (Born) approximation, taking into account both the wavefront and polarisation of the fields which form the interference pattern. The analysis is beyond the scope of this paper and will be published elsewhere however it can be shown that, assuming axial symmetry along the inscription beam direction and a probe beam perpendicular to this axis of symmetry, the complex permittivity change in 3D can be determined from the measured interferogram. In practice the full reconstruction process requires appropriate filtering, noise subtraction and normalisation followed by further calculations to determine the plane wave amplitudes and subsequent convolution with the Bessel functions corresponding to the axial symmetry.

Figure 1 shows the optical arrangement used to enable inscription and in-situ holographic measurement. The inscription laser used for these experiments was a single-box chirped-pulse oscillator (CPO) (Scientific-XL, Femtolasers) operating at a wavelength of 800 nm, and capable of delivering of up to 1 W of average power, 11 MHz repetition rate, and with a pulse duration of about 50 fs ($\approx$ 90 nJ per pulse). In this experiment the beam was pre-focused using a Mitutoyo $\times$100 objective with an NA of 0.55. An optically polished BK7 glass sample of $1.5 \times 1.5 \times 15$ mm was used as the target and could be positioned using a 3D-translation stage (Aerotech) inside a specially designed cuvette (figure 2). For first demonstration a single frequency CW diode pumped solid state laser operating at 532 nm was used for online diagnostics. This laser was divided to provide probe and reference beams using a Mach-Zehnder interferometer which used broad-band beam-splitters (BS), the reflecting layers are being optically contacted between two thick (9.5 mm) glass slabs. Both beams were focused through a long working distance objective in front of the cuvette ($\times$5, NA=0.14).

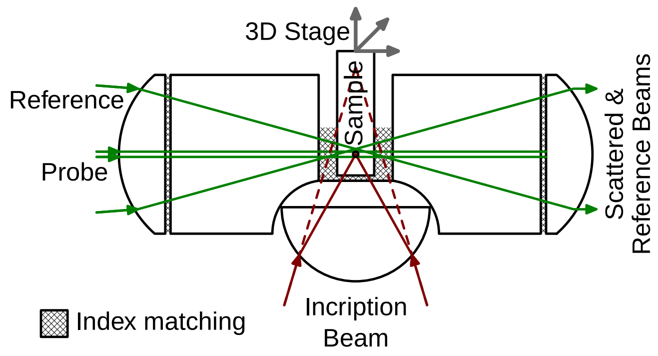

Fig. 2. (Colour online) Cuvette and sample arrangement.

In the cuvette the two beams do not overlap. The reference beam is transmitted over unmodified glass and is tightly focused to enable even illumination and parallel interference fringes on the detector plane. The probe beam is centred on the inscription. It has a much smaller diameter at the microscope objective than the reference beam with the result that it has a much confocal parameter and larger beam waste over the inscribed region and may be treated as collimated.

The optical paths for the two interfering beams are kept as close as possible to minimise differences in optical path and the aberrations experienced such that when the two beams interfere on the detector their electric fields can be considered parallel, and the scalar approximation can be used.

The cuvette (figure 2) was built from the same material as the sample, used index matching glue internally and had the sample immersed into index matching oil in order to minimise parasitic reflections. Additionally the optics used for all beams were designed to satisfy the aplanatic conditions in order to minimise aberrations. This has the additional advantages for the inscription process in that it enables aberration free focusing at depths of a few mm into the sample - much deeper than is possible using the microscope objective alone. The effective numerical aperture is also increased as can be seen in figure 2 by comparing the dashed line corresponding to the focus with microscope objective alone and the solid line when focusing through the aplanatic optics. In our configuration the effective numerical aperture of the inscribing objective is increased from 0.55 to approximately 1.26.

In order to demonstrate the technique the interferograms were monitored at 18 frames per second during exposure from the laser source at 11 MHz. Figure 3 shows the measured absolute amplitude of the scattered wave calculated from the interferograms at average laser powers of (a) 475 mW and (b) 700 mW after exposure. This scattered amplitude is calculated from the filtered single interference term in the Fourier transform of the change in the interferogram induced by the inscription and can be monitored in real time.

The concentric ring patterns observed are indicative of a spherical shape with a sharp edge and the differences between the two different inscription powers can clearly be seen. The inscription beam direction is vertical in these figures. The mirror symmetry in horizontal direction confirms the validity of assuming axial symmetry along the beam direction while the asymmetry in the vertical direction implies that the change in epsilon must have both real and imaginary components.

Where the collimated probe beam is incident on the camera the amplitude saturates the sensor with the result that a separate measurement or experiment may be required to measure $|S|$ at that point. In figure 4 the value of $\Delta\epsilon$ is calculated by interpolation from neighbouring unsaturated regions.

Figure 4 shows the real 4(a) and imaginary 4(b) parts of the reconstructed distribution of ($\Delta\epsilon_\tau$) - the modified permittivity in real space corresponding to figure 3(a). The shape of the real component agrees with that previous studies.[13] The positive imaginary component of this indicates that there is an induced

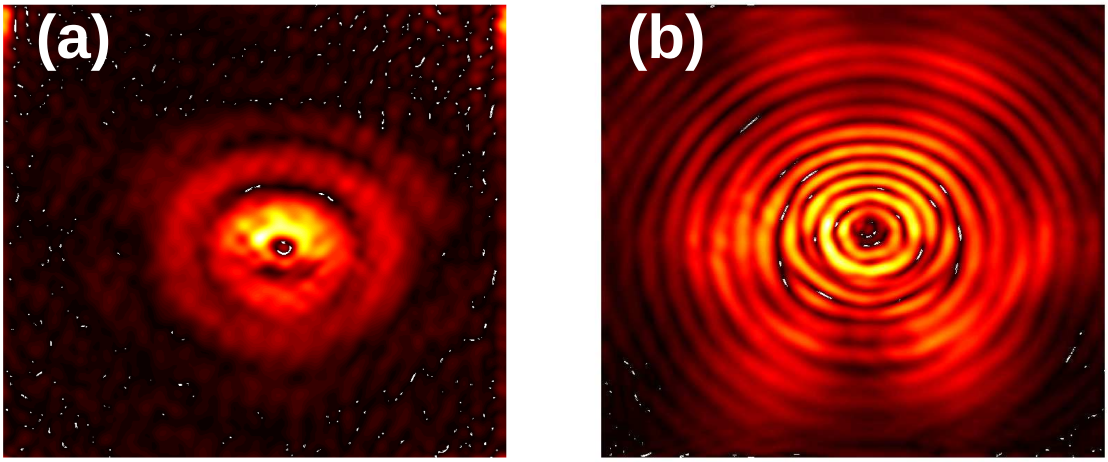

Fig. 3. (Colour online) Plot of absolute value of the scattered wave $|S|$ for inscription at (a) 475 mW after 5 seconds exposure and (b) 700 mW after 2.5 seconds exposure.

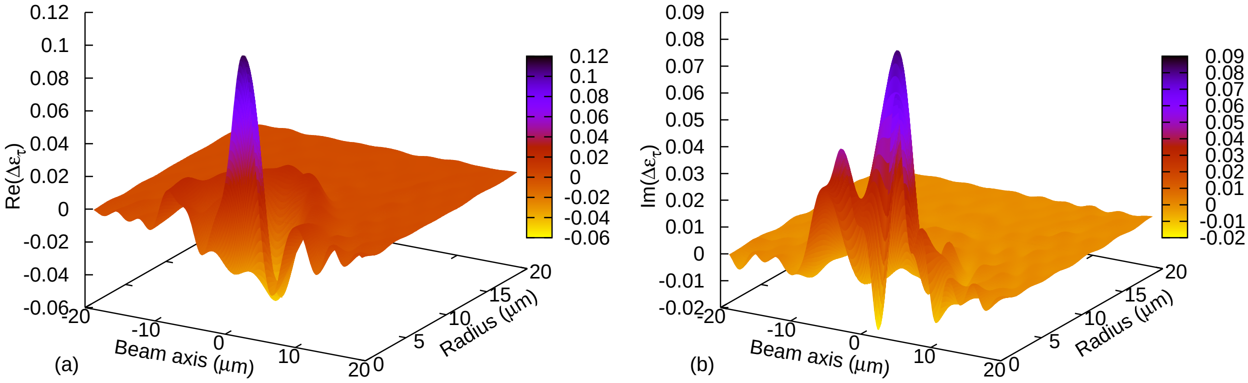

Fig. 4. (Colour online) Plot of axially-symmetric reconstructed $\Re(\Delta\epsilon_\tau)$ (a) and $\Im(\Delta\epsilon_\tau)$ (b) in cylindrical coordinates at an inscription laser power of 475 mW

absorption in the exposure process as well as the induced refractive index. Negative values of $\Im(\Delta\epsilon_\tau)$ which would be unphysical give an estimate for errors in both experimental and reconstruction procedures.

3. Conclusions

We have demonstrated experimentally a practical technique capable of measuring evolution of change in complex permittivity in dielectric materials with resolutions on the scale of microns when under direct femtosecond irradiation. Analytic solution of the EM scattering problem beyond the paraxial approximation has enabled development of a numerical technique for the inverse scattering problem for the case of transverse imaging of the modified volume under the assumption of axial symmetry along the inscription beam axis. We have used this to observe the development of induced permittivity during and after high repetition rate femtosecond laser irradiation. The observed maximum induced changes of $\Re(\Delta\epsilon_\tau)$ and $\Im(\Delta\epsilon_\tau)$ were 0.1 and 0.06 respectively,at a laser pulse energy of 20 nJ and repetition rate of 11 MHz. This technique paves the way for improving our understanding of the processes and the control during femtosecond inscription of dielectric materials. The technique could be applied in a pump-probe regime which would enable measurement of the evolution of index on ultrafast timescales.

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