Available online at www.sciencedirect.com
ScienceDirect
Current Applied Physics 8 (2008) 412–415
Current Applied Physics
An official journal of the KPS
www.elsevier.com/locate/cap
www.kps.or.kr

Formation of nano-voids in transparent dielectrics by femtosecond lasers

Eugene G. Gamaly [a,*], Saulius Juodkazis [b], Hiroaki Misawa [b], Barry Luther-Davies [a], Andrei V. Rode [a], Ludovic Hallo [c], Philippe Nicolai [c], Vladimir T. Tikhonchuk [c]

[a] *Laser Physics Centre, Research School of Physical Sciences and Engineering, The Australian National University, Canberra ACT 0200, Australia*
[b] *Research Institute for Electronic Science, Hokkaido University, CRIS Building, Sapporo 001-0021, Japan*
[c] *Centre Lasers Intenses et Applications, UMR 5107 CEA, CNRS, Université Bordeaux 1, 33405 Talence Cedex, France*

Available online 1 November 2007

Abstract

Formation of three-dimensional (3D) patterns of 0.2–0.5 μm diameter voids inside a transparent solid has been demonstrated. The nano-voids could be created with a spacing ~2 μm thus suggesting the possibility of permanent 3D optical memory with density ~100 Gbit/cm^3. Extremely high pressures of ~10 TPa and temperatures up to 10^5 K can be produced by a single sub-picosecond laser pulse tightly focused inside transparent dielectrics. Analysis of the size of the void and the shock affected zone as a function of the deposited energy shows that the experimental results can be explicitly understood on the basis of conservation laws and be modeled by plasma hydrodynamics. Therefore, the size of the void can be controlled and predicted by proper focusing conditions and laser parameters.

PACS: 81.07.−b; 96.50.Fm; 62.50.+p; 81.40.−z; 47.40.Nm

Keywords: Femtosecond laser microfabrication; Shock waves; Dielectric breakdown; Sapphire; Nano-structuring

1. Introduction

The pressures in excess of 0.1 TPa have been obtained using a diamond anvil in stationary conditions, whilst transient pressures behind shock waves generated by chemical or nuclear explosions or generated using powerful lasers up to 50 TPa have been reported [1]. Recent studies have demonstrated [2–5] that sub-picosecond laser pulses tightly focused inside transparent dielectrics can produce detectable sub-micrometer-sized structural modifications, including voids. This requires intensities in excess of $I = 0.1$ PW/cm^2, which results in a highly non-linear light–matter interaction with most dielectrics being ionised early in the laser pulse. So far the formation of voids in silica was associated with self-focusing of the laser beam [2].

Here we present, experimental evidence that one can create TPa pressures, many times exceeding the strength of any material, with low energy pulses from a conventional tabletop sub-picosecond laser without self-focusing. We demonstrate that nano-voids are formed by the extreme temperatures and pressures created by optical breakdown and these drive shock and rarefaction waves. The nano-void can be used in three-dimensional (3D) memory applications.

2. Experimental

The interaction between an intense laser pulse and the material is fundamentally different when the laser beam is tightly focused inside a transparent solid rather than on its surface. Inside the solid the interaction zone containing

* Corresponding author.
E-mail address: gam111@rsphyse.anu.edu.au (E.G. Gamaly).

1567-1739/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.cap.2007.10.071

high energy density is confined in a cold and dense material. For this reason hydrodynamic expansion is highly restricted and proceeds as a micro-explosion when a high pressure and temperature volume is created inside bulk (Fig. 1). In our experiments 100 nJ, 150 fs, laser pulses at 800 nm were tightly focused using high numerical aperture objective lens ($NA = n \times \sin(\theta) = 1.35$, where θ is half-angle of focusing cone) inside a dielectric.

To avoid self-focussing, and thus deliver the laser energy to a specific location inside the transparent solid, requires the laser power to be lower than the critical value for self-focusing. This critical power P_{cr} depends on the nonlinear part of the refractive index $n_2 = n_0 + n_2 I$ as $P_{cr} = \lambda^2/(2\pi n_0 n_2)$ [6]. The pulse energy was varied in a range 20–150 nJ. This irradiance exceeded the optical breakdown threshold but the pulse power was below P_{cr}. We define the focal volume as that confined inside a cylinder with the diameter equals to FWHM of laser intensity. The radius of this cylinder for a Gaussian beam is $r \cong \sqrt{\ln 2/2}\, r_0$, while $r_0 = 0.61\lambda/\mathrm{NA}$. The axial length of the focal volume is twice the Rayleigh length $2z_0 = 2\pi r_0^2 n/\lambda$. Hence, the focal volume is $V \simeq 0.95\lambda^3 n/\ \mathrm{NA}^4$ and the focal spot area is $S \simeq 0.41\lambda^2/(\mathrm{NA})^2$. For our experiments: $V = 0.29\ \mu\mathrm{m}^3$ and $S = 0.15\ \mu\mathrm{m}^2$ leading to an intensity of $0.35\ \mathrm{PW/cm}^2$ for a 100 nJ pulse.

An array of laser-affected spots was created aligned along the c-plane inside the sapphire crystal using a sequence of single laser pulses. The sample was then cleaved along the c-direction and examined using a scanning electron microscope (SEM). The observed pattern is shown in Fig. 2. The central void was surrounded by a shell extending to about twice the void diameter. This shell was identified as amorphous material by chemical etching, since amorphous sapphire has a much higher solubility in HF acid compared with crystalline sapphire [7]. The teardrop shape along the direction of pulse propagation is due to spherical aberration caused by the refractive index mismatch between the immersion oil and sapphire [8].

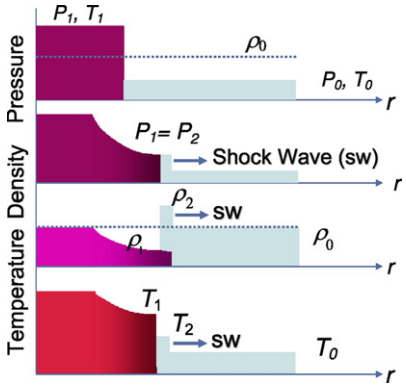

Fig. 1. Schematics of pressure, P, temperature, T, and density, ρ, transients in the case of strong explosion and shock wave (SW) formation (the index 0 marks the unperturbed values). The initial high pressure P_1 and temperature T_1 sphere is centered at $r = 0$; r denotes a radial coordinate.

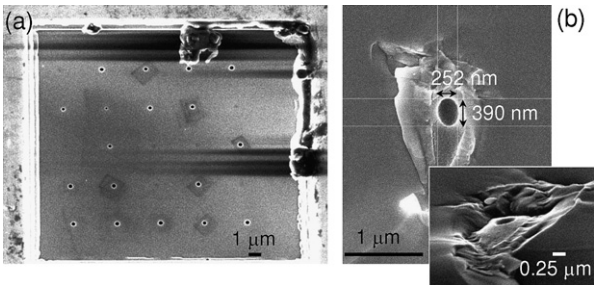

Fig. 2. (a) The top-view (c-plane) SEM image of array of voids recorded at 5 μm depth in sapphire. The voids were revealed by FIB etching with Ga^+ ions. Pulse energy was 120 nJ, pulse duration 150 fs at the wavelength of 800 nm. (b) The typical side-view (along c-axis) SEM image of the void surrounded by amorphous shell inside sapphire. The inset shows a tilted view of the void.

3. Results and discussion

3.1. Void formation

The formation of a void and shock-affected zone can be understood from simple reasoning based on the laws of mass and energy conservation. For simplicity, a spherically symmetric motion is considered. The shock wave propagating in a cold material loses its energy due to dissipation, that is, due to the work done against the internal pressure, the Young or bulk modulus, that resists material compression. The distance r_s at which the shock front effectively stops defines the shock-affected volume. At this point the shock wave converts into a sound wave, which propagates further into the material without inducing any permanent changes. The distance where the shock wave stops can be estimated from the condition that the internal energy at the shock front is comparable to the absorbed energy: $4\pi r_s^3 \rho_0/3 = E_{abs}$. At this position the pressure behind the shock front equals the internal pressure of the cold material [9]. One can reasonably suggest that the sharp boundary observed between the amorphous (laser-affected) and crystalline (unaffected) sapphire corresponds to the distance where the shock wave effectively stopped.

Computer simulations were performed in spherical geometry using the hydrodynamic code Chivas [11], which considered the ionisation equilibrium, separate temperatures for electron and ion components, and electron heat conduction. The cylindrical region where the energy was absorbed was approximated by a sphere of the same volume. The absorbed energy $E_{abs} = 50$ nJ was deposited homogeneously in this spherical volume with radius $r_d = 0.13$ μm in a material with initial density ρ_0. The equation of state implemented in the code [11] describes solid-melt-plasma and reverse transitions. The simulations started after the end of the laser pulse with the initial electron temperature $T_e = 26.2$ eV and the average ion charge (number of electrons that are stripped off an atom) $\langle Z \rangle = 4.3$ (see Section 3.2). The hydrodynamic motion commences after the electrons transfer the absorbed energy to ions in excess of that necessary to break the inter-atomic