3-D Optical Storage Inside Transparent Materials

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We present a novel method for 3-D optical data storage that has submicron-size resolution, provides a large contrast in index of refraction, and is applicable to a wide range of transparent materials. Bits are recorded by focusing 100-fs laser pulses inside the material using a 0.65 NA objective. The laser pulse produces a submicron-diameter structurally altered region with high contrast in index of refraction. Binary information can be recorded by writing such bits in multiple planes, and read out with a microscope objective with a short depth of field. We demonstrate data storage and retrieval with 2-μm in-plane bit spacing and 15-μm inter-plane spacing (17 Gbits/cm^3). Scanning electron microscopy and atomic force microscopy show structural changes confined to an area 200 nm in diameter.

Three-dimensional optical data storage offers the potential for very large recording capacity. In addition to extensive on-going research in volume holographic data storage, a number of recent papers report 'point-like' or 'bit-wise' binary 3-D optical storage in photopolymers[1,2] and photorefractive materials.[3,4] Using a nonlinear optical process in the medium, the optical interaction can be confined in all three dimensions to a micron-sized focal volume. Pioneering experiments demonstrated 3-D optical recording at a density of up to 1.6×10^{12} bits/cm^3 in a photopolymer using two-photon absorption and extremely tight focusing.[2] The stored information consisted of less than 1% changes in the local index of refraction, and was read out serially with a Nomarski differential interference contrast (DIC) laser microscope. Experiments in photorefractive materials[3,4] used linear absorption to record data at a density of up to 4.2×10^9 bits/cm^3. The authors suggested that the recording density could be increased using two-photon absorption. The stored data was read out with a phase-contrast microscope.[3,4]

Here we report a novel method for creating sub-micron-sized bits that have a large contrast in index of refraction and that can be read out with transmitted or scattered light under a standard microscope. The method can be used for permanent 3-D optical data storage in a wide range of materials including fused silica, fused quartz, sapphire, and various glasses and plastics, thus allowing for a storage medium that is mechanically, chemically and thermally very stable, and inexpensive. Unlike a photopolymer gel, there are no problems of distortion due to shrinkage and flow, and of isomerization due to ultraviolet light. And unlike photorefractive materials, the difficulties of fixing the recorded data are entirely avoided.

We tightly focus ultrashort laser pulses inside a transparent material to create localized structural changes, thereby altering the index of refraction. This process is used to record digital information in three dimensions by writing multiple planes as illustrated in Fig. 1. In our experiments, we translate the
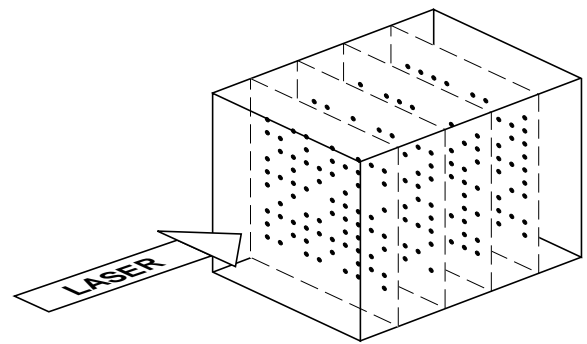

Fig. 1. Schematic diagram of 3-D optical data storage.

sample in the transverse plane, and move the focusing objective along the beam axis. Figure 2 shows an example of a random binary pattern stored inside fused silica, recorded using 0.5-μJ, 100-fs, 780-nm pulses from a regeneratively amplified Ti:Sapphire laser, focused by a 0.65 numerical aperture (NA) microscope objective. With this focusing, the threshold for observable structural change is 0.3 μJ. The pattern is read out in parallel using transmitted light in a microscope with a 0.95 NA objective. The spacing between adjacent bits is 2 μm. The written spots can be viewed as dark or bright points depending on the focusing of the read-out objective. This change in brightness can be used as a contrast enhancing feature in a read-out system. During read-out, the depth discrimination provided by the short depth-of-field of the 0.95 NA objective is sufficient if adjacent layers are spaced by about 10 μm or more. The longitudinal extent of the structurally altered regions is about 2.5 μm. We recorded 10 layers spaced by 15 μm, using a standard 0.65 NA refractive objective. Using an objective with a large working distance and an adjustment for aberrations caused by focusing into the material, it should be possible to record over 100 layers, spaced by 10 μm.

The 1-μm apparent size of the spots in Fig. 2 is at the resolution limit of the optical microscope. To obtain information about the lateral extent of the
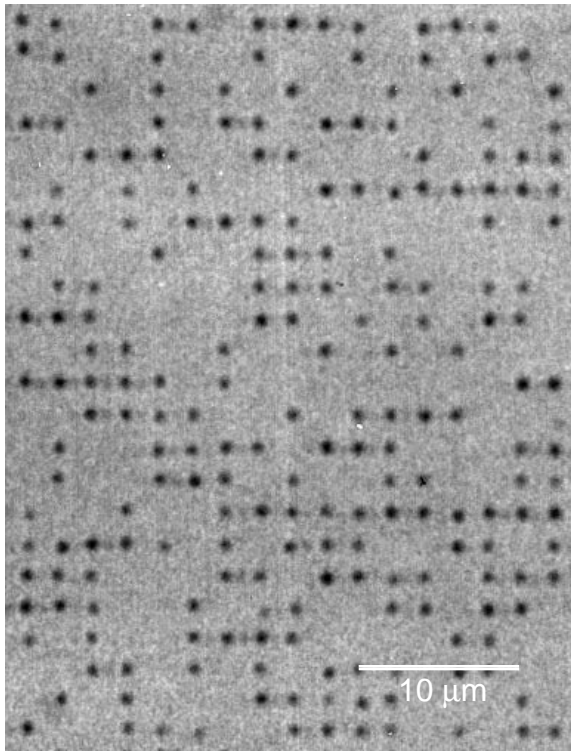
Fig. 2. Binary data pattern stored inside fused silica with 2-μm bit spacing, photographed with an optical microscope using transmitted light.

Fig. 3. A tilted SEM view of a polished cross-section through a regular array of bits with 5-μm spacing. Inset shows a single bit at higher magnification.

structural changes in the material, we polished away the fused silica sample until the surface level reached the internally recorded structures, applied a 30-nm gold coat, and viewed the sample under a scanning electron microscope (SEM). Figure 3 shows such a SEM image of a 5×5-μm regular array of spots recorded under conditions identical to those used in Fig. 2. The sample is tilted in the SEM to better show the morphology. The bright spots correspond to protrusions on the surface, while the dark spot (top row second from the right) corresponds to a cavity in the surface, as verified with an atomic force microscope (AFM). Whether a structurally altered region appears as a protrusion or a cavity depends on the level to which the material is polished. The inset in Fig. 3 shows a cavity at higher magnification. Both types of features have diameters of roughly 200 nm, implying a recording density limit of about 10^{13} bits/cm^3 with a 0.65 NA objective.

It may be possible to write even smaller diameter and shorter length features using an objective with NA > 0.65. However, to read out data stored at densities significantly higher than what we have demonstrated requires using (serial) scanning techniques such as DIC laser microscopy.[2]

A likely mechanism for the creation of these structures is a 'micro-explosion' that occurs inside the material. Energy from the ultrashort laser pulse is coupled into the transparent material through a combination of multiphoton absorption and ava-
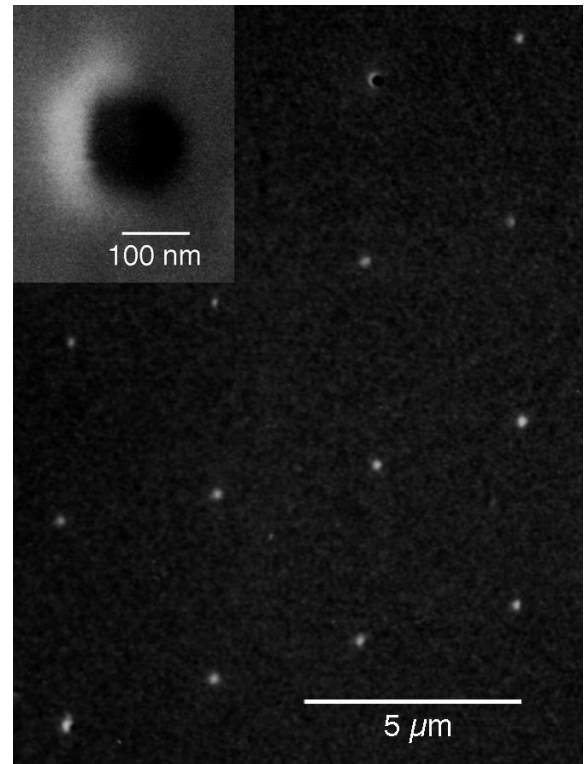
lanche ionization.[5-8] Unlike ultrafast surface damage experiments,[5-8] however, ablation is not possible since the excited region is internal to the material. The ultrafast energy deposition creates very high temperatures and pressures inside the region; material is ejected from the center and forced into the surrounding volume, leading to the formation of a structure consisting of a void (or at least less dense material) surrounded by densified material. This mechanism is consistent with the SEM and AFM observations: the protrusions suggest the creation of denser, harder material, more resistant to the mechanical polishing; deeper polishing reveals a pit corresponding to a void which is created at the center of the micro-explosion.

The submicron bit size is not due to a simple thresholding effect which would occur with an excitation that is only a few percent above the threshold. The 0.5-μJ pulse energy used to make the patterns shown in Figs. 2 and 3 is well above the observed threshold of 0.3 μJ. Instead, the small size is likely caused by the nonlinearity of the absorption, which creates an excited region significantly smaller than the linear intensity distribution. Furthermore, self-focusing may be reducing the size of the beam waist. Finally, the dynamics of the micro-explosion may further confine the extent of the structurally altered region.

Surprisingly, the 0.5-μJ pulse energy used in these micro-explosions is *much less* than the energy

required to write bits in photosensitive materials: 20 μJ per bit (2 mW for 10 ms, in 100-fs pulses at 100 MHz) was used for two-photon recording in a photopolymer with extremely tight focusing by a 1.4 NA objective[2]; linear absorption in a photorefractive[4] required 60 μJ per bit (20 mW for 3 ms), also very tightly focused by a 1.0 NA oil-immersion objective lens. The threshold is higher in the experiments on photopolymers and photorefractives because the fraction of the incident laser energy that is absorbed in the focal volume is orders of magnitude smaller than in our experiments. Furthermore, while the change in index of refraction, Δn, in the photopolymer is 8×10^{-3},[2] and in the photorefractive material (LiNbO$_3$ crystal) only about 10^{-6},[4] in our experiments the refractive index changes are large enough to be clearly visible under a standard microscope. Using diffraction from periodic arrays of bits (to be discussed in an upcoming publication) we estimate that Δn is in the range of 0.05–0.45, with the upper limit set by the silica-vacuum interface.

The structures produced by 100-fs pulses differ drastically from those produced by 200-ps and 10-ns pulses. With the longer pulses, the resulting structures are irregularly shaped, and cracks appear in the material even at energies only slightly above the threshold for structural change. Fig. 4 shows an example of the features produced by 200-ps pulses from the same laser system. The focusing conditions and beam profile are identical to those used in Fig. 2; the pulse energy is 9 μJ, which is three times the observed threshold, and the point spacing is 10 μm. Note the large size of the spots, and clearly visible cracking produced with these longer pulses. With 10-ns pulses from a Nd:YAG laser, even larger, irregularly shaped spots are produced, and the cracking is more extensive.

With 100-fs pulses, the small, regular features described above are produced by pulses in a wide energy range of up to three times the 0.3-μJ threshold. Femtosecond pulses of even higher energy produce a 'head and filament' structure of 20–40-μm length that is visible to the unaided eye, and that can be used for internal marking and engraving 3-D patterns in a wide variety of transparent materials. We observed no cracking with the 100-fs pulses even up to pulse energies 100 times the threshold.

We have written information in a wide variety of transparent materials including fused silica, fused quartz, sapphire, BK7 optical glass, and plastic (acrylic). Surprisingly, the threshold for structural change is very similar in all of these materials — within a factor of 2 of the threshold of fused silica. A notable exception is diamond, where the threshold is at least 100 times greater. In all materials, and especially in diamond, the threshold for internal structural change is significantly higher than for surface ablation.

In conclusion, we have demonstrated a novel method for high density 3-D optical data storage using ultrashort laser pulses. The recording is done with ultrafast-laser-induced micro-explosions, pro-
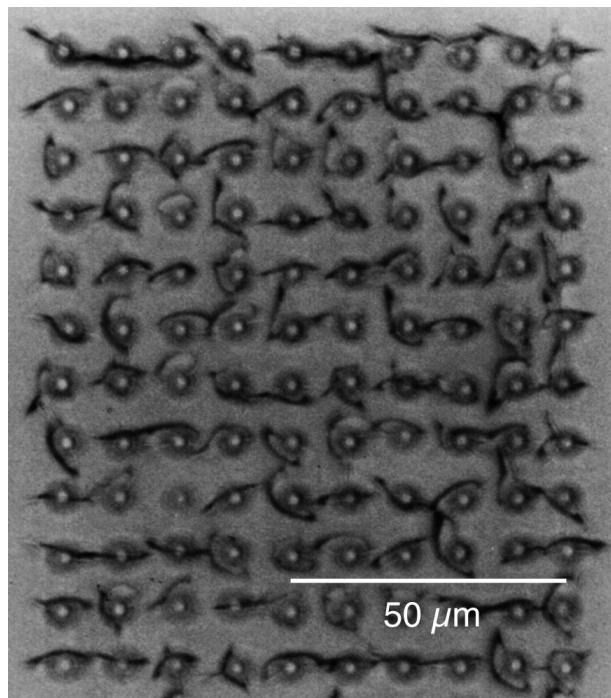

Fig. 4. Damage produced inside fused silica by 200-ps pulses. Optical microscope photograph using transmitted light. Note the difference in scale relative to Fig. 2; bits are spaced by 10 μm.

ducing localized submicron-diameter structures with high contrast in index of refraction. Parallel readout and depth discrimination are demonstrated with a standard microscope. Because a photosensitive medium is not required, a wide range of transparent materials can serve as the storage medium.

The method can also be used for engraving very fine-scale patterns inside transparent materials, without damaging or altering the surface. Other applications could include producing 3-D diffractive optical elements and periodic structures, patterning gratings in optical fibers, and creating materials with increased density and hardness.

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Three-dimensional optical storage inside transparent materials: errata

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In our recent Letter[1] a line of text was dropped in printing.   On p. 2024 the beginning of the paragraph above Fig. 3 should read as follows: "Surprisingly, the 0.5-μJ pulse energy used in these microexplosions. . .".

Reference

1. E. N. Glezer, M. Milosavljevic, L. Huang, R. J. Finlay, T.-H. Her, J. P. Callan, and E. Mazur, Opt. Lett. **21,** 2023 (1996).