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Demonstration of high-density three-dimensional storage in fused silica by femtosecond laser pulses

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Three-dimensional optical recording of high contrast spherical bits (diameter<300 nm) at a density of 500 G/cm^3 in fused silica using a Ti:sapphire femtosecond laser is demonstrated. Bits are optically read out using both a confocal and a phase-contrast scheme. The recording density for different materials and recording mechanisms are discussed. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1589596]

I. INTRODUCTION

The need to store large quantities of data has led to considerable research into high-density memory. For optical memory, the density is ultimately limited by the diffraction of electromagnetic waves. The present two-dimensional techniques, such as compact disks and magneto-optical storage, have almost reached this limit. Further increases in storage density require the use of the third dimension. Femtosecond laser induced plasma ablation in transparent materials, which produces a submicrometer microexplosion, provides a powerful way to realize three-dimensional (3D) optical storage.[1–6] Since the recording process results in structural damage to the storage medium, this technique offers the advantages of long storage life and temperature stability (to ~1100 °C)—key considerations for data archiving.[7]

In this article, we report on an experimental investigation into the recording and readout of 3D high-density optical data storage in fused silica and other optical glasses using a femtosecond laser.

II. EXPERIMENT

Light from a home-built chirped pulse amplified Ti:sapphire laser system is first passed through a spatial light filter (10 μm pinhole) to improve laser beam quality and enlarge its diameter. A microscope objective [40× magnification, numerical aperture (NA)=0.65 or NA=0.85] focuses the pulse into fused silica that has been prepared in a cubic shape with four optical surfaces to allow the laser–matter interaction zone to be observed from different orthogonal directions. Bits are recorded successively, layer by layer, each by a single pulse. A computer-controlled three-axis translation stage (100 nm resolution) is used to move the storage medium between pulses.

III. RESULTS

A. Recording in fused silica

Figure 1 is an optical image of recorded bits. The shape of the bit is clearly conical rather than reflecting the ellipsoidal distribution of laser intensity at the focal point.

In recording layers of bits, either record layers closest to the excitation source or layers farthest from the excitation source may be recorded first. When the layer separation is >14 μm, there is no difference—the bit size in all layers is the same. If, however, the layer separation is reduced to <7 μm, the shape of the bits is dependent on the order of recording. If one records layers closest to the excitation source first, bits on layers farther away are smaller (as previously observed by Li *et al.*[8]). If, however, one records the farthest layers first, bit size is independent of position.

With this NA=0.65 objective, over 60 layers could be recorded with separation between layers of 7 μm. The area of the refractive index change is ~500 nm in diameter and ~3 μm deep. Each bit occupies a volume of 1×1×7 μm, corresponding to a memory density of 143 Gb/cm^3, comparable to the 17 Gb/cm^3 storage density obtained by Glezer *et al.*[1] using a similar recording mechanism.

As is seen in Fig. 1, while the diameter (ϕ) of a bit is ~400 nm, the depth is of the order of 3 μm. By employing a higher NA objective lens (shorter focal depth) and reducing the excitation intensity, the depth of the bits can be decreased. Replacing the NA=0.65 objective lens with a NA =0.85 objective lens resulted in the damage threshold decreasing from 700 to 100 nJ (same beam quality and pulse width). Using 500 nJ pulses, a bit depth of <2 μm was achieved.

For data storage applications, a spherical bit rather than a conical bit is clearly preferable. Figure 2 shows that by further reducing the pulse energy to 400 nJ, the volume of the refractive index change can be made nearly spherical with individual bits having a diameter of <300 nm. At this pulse energy, we recorded a matrix of bits with an in-plane separation of 1 μm and a layer separation of 2 μm. This corresponds to a density of 500 G/cm^3. This is a high storage density obtained using a femtosecond laser in a transparent material. By improving the focusing system, we believe that the bit diameter and depth can be further decreased, resulting in storage densities in the order of dozens of T/cm^3.

B. Recording in ZBaF15 optical glass

An alternative transparent material is ZBaF15 optical glass (Ba atoms optically active). 50 nJ pulses with the 0.85

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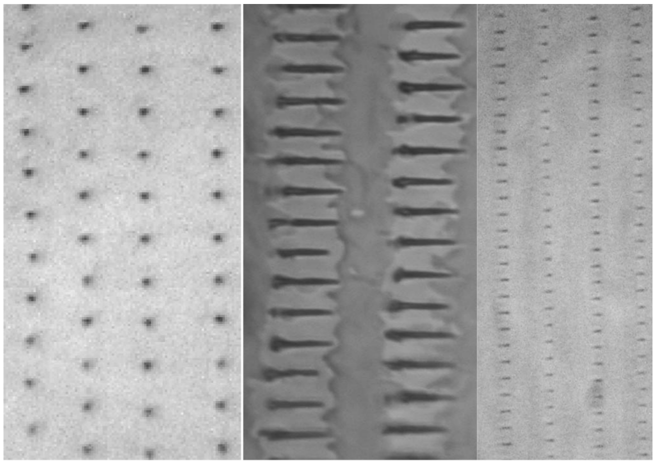

FIG. 1. Optical image of bits written inside fused silica by a single pulse focused by a NA=0.65 objective lens. (Left) bits as viewed parallel to the 1 μJ 200 fs excitation pulse. Each bit is separated by 2.5 μm from its nearest neighbors. Nonlinearity in the step motor is the reason the bits are not perfectly aligned. (Middle, right) bits as viewed orthogonal to the excitation pulse. In the middle image, excitation energy is 2 μJ, the in-plane bit separation is 5 μm, and each layer is separated by 14 μm. In the right image the excitation energy is 1 μJ, the in-plane bit separation is 2.5 μm, and each layer is separated by 7 μm.

NA objective, are sufficient to observe white-light emission with a charge-coupled-device (CCD) camera, along with changes in both the color and refractive index of the excited region. Figure 3 shows the refractive index change induced by a 200 nJ femtosecond pulse. Long filaments are due to the generation of color centers. The three high contrast bits (relative to both background and long filaments) were generated by microexplosions in the middle of the filament (color center).

While the modification due to the color center is reversible by increasing the crystal temperature, the microexplosion is irreversible. In addition, the refractive index change due to the color center is less than that resulting from a

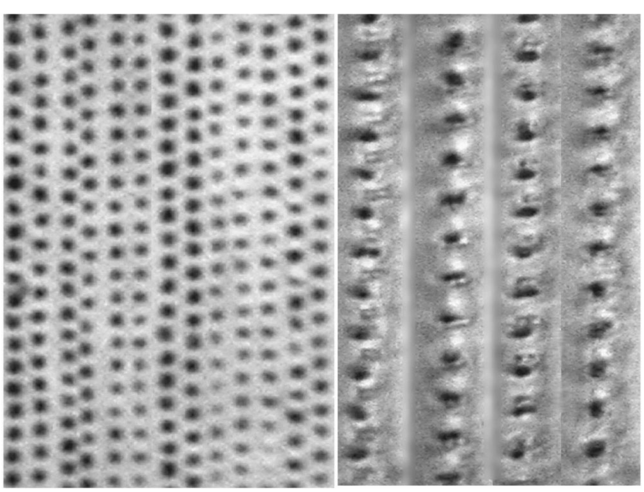

FIG. 2. Optical image of bits written inside fused silica as viewed parallel to the 400 nJ 200 fs excitation pulse. Light is focused by a NA=0.85 objective. In-plane bit separation is 1 μm and the layer separation is 2 μm. (Left) bits as viewed parallel to the excitation. (Right) bits as viewed orthogonal to the excitation pulse.

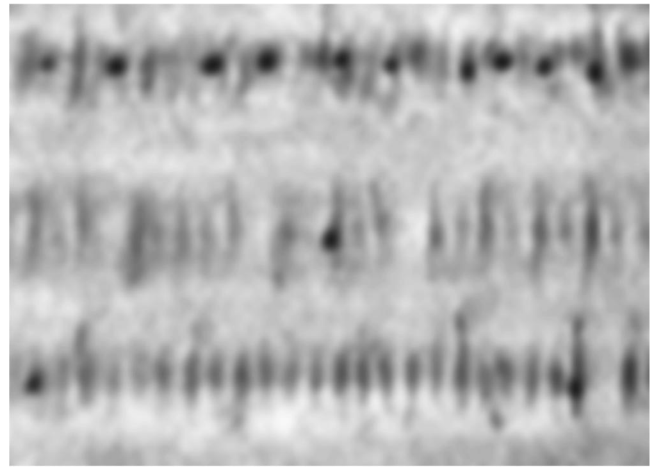

FIG. 3. Optical image of bits inside ZBaF15 optical glass as viewed parallel to the 200 nJ, 200 fs excitation pulse. Light is focused by a NA=0.85 objective. In-plane bit separation is 2.5 μm and the layer separation is 7 μm.

microexplosion. The threshold of the color-center generation is lower than that of the microexplosion. These suggest that refractive index modification induced by femtosecond laser pulses in optical glass at low intensities is due to color-center generation, while that at high intensities is due to microexplosions with structural damage only occurring as a result of the second mechanism.

C. Data readout

Figure 4 shows a single row of bits read out with high signal to noise (S/N) using a CCD camera attached to a conventional phase-contrast optical microscope. The full width at half maximum is the size of the bits and the minimum contrast is 67% of the maximum. The line error of the step motor is responsible for the variations in peak contrast. This contrast is good enough for reliable reading out of the stored information.

In Fig. 5, the influence of the blemish in the sample on the recording bits is shown. A 2-μm-diam blemish in the sample is at the position of a bit that should be recorded (left). Along with leading to a bit error on this level (left), the blemish also leads to bit error at the layer below (middle) as

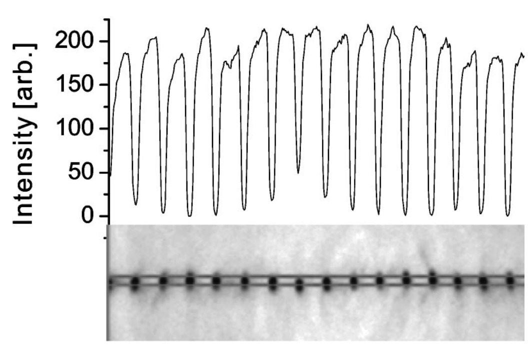

FIG. 4. Read out of data through a phase-contrast microscope. The upper part of the figure is the signal recorded by a single row of the CCD camera. The pits being imaged are shown in the bottom portion, with the two lines denoting the edges of the row. High contrast is evident even for those bits not perfectly centered.

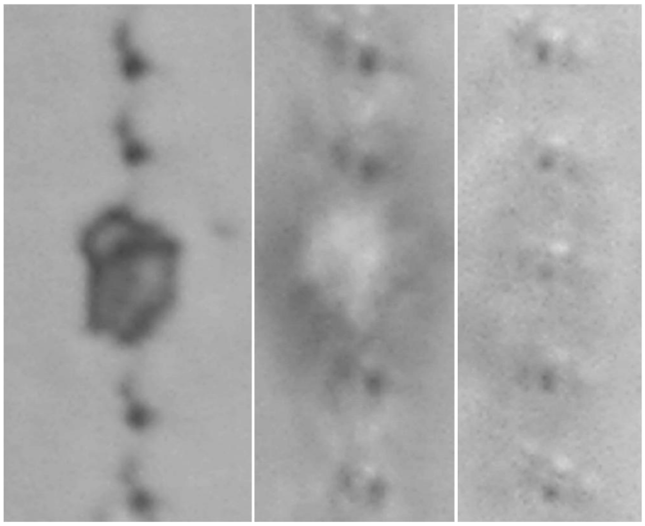

FIG. 5. Optical images illustrating the influence of a blemish in fused silica on the recording of bits. (Left) layer in which blemish occurs. (Middle) layer below blemish. (Right) two layers below the blemish.

well (layer separation ~7 μm). Data can be successfully recorded two layers below the blemish (right). In practical applications, this should not be a problem as fused silica is generally blemish free.

While the above system provides accurate read out, it is hard to build a compact digital reading system based on a conventional phase-contrast optical microscope or CCD camera. In addition, the shutter speed of the CCD (10-4s) camera limits the speed of the reading. Figure 6 (top) shows a transmission confocal-type readout system that overcomes these problems. The mechanism of this readout system is based on optical scattering from the data bit. One 0.65 NA objective (Fig. 6, OBJ-1) was used to focus the He–Ne laser (632.8 nm) beam on the sample, and another 0.45 NA objective (Fig. 6, OBJ-2) used to collect the transmitted light. The intensity of the transmitted light decreases dramatically at points where the He–Ne laser beam is focused at the data bit. A *PIN* optical-electronic detector is used to detect the optical signal. A capacitor is used to discriminate against direct current electronic signal. (Without the capacitor, diffracted light is a part of the background signal and results in a decrease of the S/N.) Figure 6 (bottom) presents a sample result of bit data read out using this system. The reading speed is 5 mm/s, which is same the as the writing speed. Increasing the reading and writing speed by a factor of 3 was observed to cause no deterioration in the S/N. The limit to the reading speed is the response time of the optical-electronic detector.

In order to maximize the signal to noise, it is important to match the focal spot size of the reading beam to the diameter of the data bit. In the case that the spot size is greater than that of the data bit the ratio between scattered and not scattered light will drop. In the above system replacing the NA0.65 focusing objective by one with a NA of 0.25 reduced the S/N by a factor of 6. Conversely, a spot size less than the data bit also results in a reduced S/N due to the fact that the data bit is not a uniform scatterer. Thus, the high-density limit for optical storage may not be the bit size itself. Rather, one needs to take into account the wavelength of the

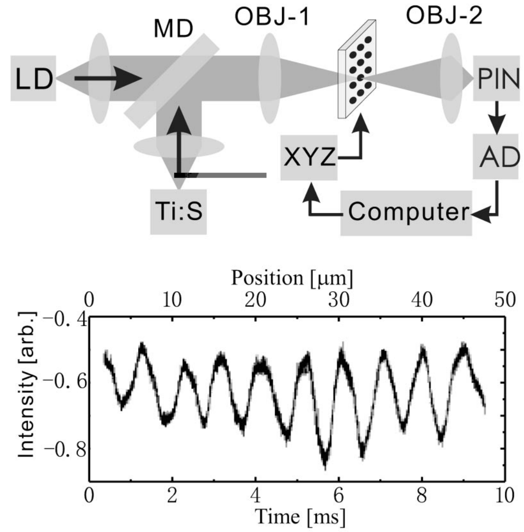

FIG. 6. (Top) schematic of a transmission confocal system for recorded data out of the 3D optical memory. Data are first recorded by a femtosecond (Ti:S) laser pulse, which is focused on the sample by a high-power objective lens (OBJ-1). Light from a laser diode (LD) is collimated and after passing through a dichroic mirror (DM) is focused by the same objective lens (OBJ-1) onto the sample surface. A second objective lens (OBJ-2) is used to focus the transmitted light onto a photodiode (*PIN*), whose output is converted to digital form (A/D), and stored on a computer. (Bottom) sample read out of data through the above system performed by scanning (*XYZ*) the sample at 5 mm/s.

reading laser and the focusing lens to be used reading out the data—there is no use writing ϕ=100 nm bits if the readout beam cannot be focused to ϕ=100 nm. For maximum storage density a short wavelength reading laser and a high NA focusing lens with long working distance is required.

IV. DISCUSSION

The mechanism of the optically induced refractive index change is not very clear, but some key principles are understandable. While a transparent material of such fused silica absorbs only a little energy from a nonresonant laser pulse, an extremely intense laser pulse can ionize atoms through multiphoton ionization. These free electrons induce avalanche ionization generating a high-density free-electron plasma and an intense electronic field. This plasma absorbs further photons from the femtosecond pulse, resulting in additional free electrons from favorable collisions with the bound electrons and the lattice. A plasma is thus formed in which the free-electron density increases exponentially.[9]

The shape and volume of the region of the index of refraction change is another issue that we would like to discuss briefly. In the lateral plane, at the highest recording densities (Fig. 2), the minimum bit diameter is ~300 nm (as observed by near-field optical microscopy)—a factor of 2 smaller than the diffraction-limited laser beam waist [~570 nm (l/e^2) for an 800 nm Gaussian laser pulse]. This is due to the fact that only in the central portion of the beam is the laser intensity above the threshold for avalanche ionization or the generation of fifth-harmonic photons. This is supported by the fact that at higher laser intensities, the spot size is considerably larger. While spherical bits are burned when the laser pulse is near threshold (Fig. 2), increasing the laser power well above threshold, results in a conical rather than the ellipsoidal shape that might be expected from the laser intensity around the focal region (as seen in Fig. 1). This conical shape is the result of a dynamic balance between the competitive processes of self-focusing induced by the optical Kerr effect and defocusing induced by plasma formation.[10]

There are two possible approaches to reduce the cost of 3D optical data storage based on the above mechanisms. In the first approach, one looks for materials such as ZBaF15 optical glass in which the threshold for microexplosion is low.[11–14] Unfortunately, color-center generation requires lower incident intensity than microexplosion and is unstable (the information recorded by the color center self-erases). The second approach is to seek to develop cheaper femtosecond laser sources (i.e., femtosecond fiber-based lasers and amplifiers) capable of the intensities required to record data in fused silica. It is our opinion that the second approach will be most fruitful.

In conclusion, the generation of a high-density plasma in a small region allows a high optical storage density to be obtained. In this article, we have demonstrated the writing of spherical bits (diameter$<$300 nm) in three dimensions in fused silica at a density of 500 Gb/cm^3 using a chirped amplified femtosecond laser. As laser intensity is increased above threshold, bits assume a conical rather than ellipsoidal shape. Data can be read out at high contrast using either a conventional phase-contrast microscope and CCD camera (low cost) or a transmission confocal microscope (compact, high speed). Blemishes are shown not only to affect the layer in which the blemish occurs but also readout from the layer below. By increasing the NA of the focusing objective so as to decrease the depth of focus (steeper intensity gradient) and reducing the interaction time between the laser and plasma (by ensuring the beam intensity remains close to but a little above the threshold of optical breakdown), it should be possible to obtain data storage densities in the tens of Tb/cm^3.[15,16]

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