Modification of Glass by FS Laser for Optical/Memory Applications

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- Precipitation and growth of silicon in the glass
When a transparent material like glass is irradiated by a tightly focused femtosecond laser, photo-induced reactions should occur only near the focused part of the laser beam.

Femtosecond laser can be used to modify transparent materials like glass microscopically and three-dimensionally.
Typical structural changes

alkali silicate glass

(A) Coloring line due to defect formation
(B) Refractive index changes due to local densification by a single pulse
(C) Melting due to heat accumulation

The structural changes induced by a femtosecond laser take various forms depending on the condition of laser irradiation.
We found that a shock wave forms at the focal point inside the glass of the beam created by a femtosecond laser and propagates to the surrounding area.

The change in the spatial pattern of the probe beam was monitored to obtain the refractive-index distribution.

Optical setup used to observe the shock wave

The change in the spatial pattern of the probe beam was monitored to obtain the refractive-index distribution.

We found that a shock wave forms at the focal point inside the glass of the beam created by a femtosecond laser and propagates to the surrounding area.
The density at the center decreases and the high-density area due to pressure-wave generation rapidly expands into the surrounding area.

The positive peak due to the pressure wave propagates outward with a constant velocity.

\[ \Delta \phi(r) \text{ phase change} \]

\[ r/\mu \text{m} \]

~6.2 \( \mu \text{m/ns} \)

The temperature and pressure of this region rises very rapidly and dramatically.

>3000 K, \( \sim 1 \text{ GPs} \)
The irradiation of a single pulse

Temperature increase and thermo-elastic stress should be induced in a very limited volume.

The relaxation of the thermo-elastic stress produces the force driving the shock wave.

The shock wave propagates outward.

The structure of the glass is extended outside.

After pulse irradiation

Compressive stress moves back toward the center.

A graduated high-density region is formed in the laser-focusing area.

After the thermal diffusion from the irradiated region, the structural change becomes a permanent refractive-index change.

If laser pulses are repeatedly irradiated, the high-temperature area rapidly expands, and melting regions can be formed at arbitrary sites within the glass.
Array of refractive index changes inside silica glass

250 kHz Laser Beam

Microscope Objective

Waveguide

Refractive index change

By using a femtosecond laser with a high repetition rate, refractive index changes are continuously induced along a path traversed by the focal point.

We can write waveguides in glass.

Fig. Photo-written waveguides were written by translating the sample (a) parallel or (b) perpendicular to the axis of the laser beam

Similar waveguides can be written inside various types of glass, such as silica, borosilicate, fluoride and chalcogenide glass.
The characteristics of a writing waveguide can be controlled by adjusting the writing conditions.

**Table**  Guide mode and mode field diameter calculated from the refractive-index profile

<table>
<thead>
<tr>
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<th>Refractive index reference[%]</th>
<th>Change diameter[μm]</th>
<th>MFD@1.3μm</th>
<th>MFD@1.55μm</th>
<th>Calculated guide mode@1.3μm</th>
<th>Calculated guide mode@1.55μm</th>
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<tr>
<td>I</td>
<td>1.32</td>
<td>20</td>
<td>6.0</td>
<td>6.7</td>
<td>LP01, LP02, LP11</td>
<td>LP01, LP11</td>
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<td>LP01, LP11</td>
<td>LP01, LP11</td>
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<td>12.0</td>
<td>13.5</td>
<td>LP01</td>
<td>LP01</td>
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</tbody>
</table>

**Objective** (NA) **power** (mW) **Common Writing Condition**

- Wavelength : 800 nm
- Repetition rate : 250 kHz
- Pulse width : 270 fs
- Scanning rate : 50 μm/s

**Fig.** The refractive index profiles of the core of waveguides written in silica glass at various numerical apertures and average powers.
Characterization

- This device has matrix-arrayed multichannel optical I/Os.
- The optical path are redirected by 90 degrees.
- This structure and size enable optical interconnects to be integrated in a board-to-board or board-to-backplane communication structure more densely.

It's not easy to fabricate an optical-path redirected waveguide using the conventional method, so this is an example where the laser writing technique has a clear advantage.
Diffractive optical elements

Grating & Binary lens (microscopic view)

- Δ = 46.5 μm
- f = 9 mm
- Beam diameter = 12-13 μm
- Beam profile at the focal plane
- 254 μm
- Beam diameter = 12-13 μm
- Split beam power ~ 27% of input beam
Precipitation of gold nano-particles

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Laser intensity: (a) < (b) < (c)

550 °C for 1 hr

Blue shift

Peak position [nm]

Absorbance [a.u.]

Wavelength [nm]

Laser intensity [W/cm²]

Particle size: Small

TEM

100 nm
Applications may also include developments in techniques such as surface plasmon resonance and near-field scanning optical microscopy.
LASER BEAM
Wavelength: 800 nm
Pulse width: 50 fs
Repetition rate: 250 kHz
Average power: 100 mW
Objective: 50x, NA=0.6

Photo-reduction
Sm$^{3+}$ → Sm$^{2+}$

GLASS
SmF$_3$: AlF$_3$·YF$_3$·BaF$_2$·SrF$_2$·CaF$_2$·MgF$_2$

Laser-irradiated areas (photo-reduced areas) recorded inside glass can be detected only by emissions at 680, 700 or 725 nm.

Fig. Photoluminescence spectra for glass excited at 488 nm: red line shows spectrum for laser-irradiated areas; yellow line shows spectrum for non-irradiated area.
Alphabetical character comprised 300 to 500 photo-reduction bits. The bits had a diameter of 200 nm. The spacing between each character was 1 μm.

Recording: $\text{Sm}^{3+} \rightarrow \text{Sm}^{2+}$
Femtosecond laser
100 nJ/pulse at 800nm

Readout:
$\text{Ar}^+ \text{ laser}$
1 mW at 488 nm

Erasure: $\text{Sm}^{2+} \rightarrow \text{Sm}^{3+}$
$\text{Ar}^+ \text{ laser}$
10 mW at 488 nm

Three-dimensional optical memory with rewrite capability is achievable.
The refractive index of nano lines is lower than that of the circumference, and periodic nano lines function as a grating.

\[ \Delta n \approx -0.1 \]

SiO\(_2\)-x (x \(\approx\) 0.4)

Oxygen defect
Elemental distributions of Ba and Al at around the focal point of the laser.

The concentration of Al increased at the center of the irradiated area, and Ba increased outside the center with the passage of time.

Measurement of the x-ray diffraction patterns confirmed that only beta-BBO crystals grew in the focused area.
Metallic clusters or particles can be precipitated.
Specific ion can be reduced or oxidized.
Oxygen-deficiency centers (≡Si-Si ≡, etc.) can be formed.
Specific ions can be diffused at the micrometer order.

Silicon cluster or particle can be extracted from silica or silicate glass.

General oxide glasses can not trap superfluous oxygen ions generated by cutting the Si-O bond and growth of the silicon in a closed space like the glass inside it.

We tried to deposit silicon from silicate glass prepared using metallic aluminum as the starting material.
Silicon as a photonic medium has unique advantages for photonic integrated circuits.

In order to achieve compact and integrated devices, high refractive index difference is required. The high refractive index contrast between the silicon waveguide core and a surrounding glass cladding enables the fabrication of small optical structures.

If silicon can be deposited in the glass at three dimensions, this leads to further compactness and higher integration of optical devices.
Silicate glass prepared using metallic aluminum

Metallic aluminum: ~20 mol% in air
Melting Temp.: 1400 - 1500 °C
Crucible: Al₂O₃

**Si - Na – Al - O**
**Si - Ca – Al - O**
**Si - Ca - Na – Al - O**
**Si - Ca - B – Al - O**
Silicate glass prepared using metallic aluminum

Oxygen deficiency defect: $O_3 \equiv Si-Si \equiv O_3$

- PL (ex: 256 nm)
- PLE (em: 310 nm)
- PLE (em: 480 nm)

SiO$_2$–CaCO$_3$ (CaO)–Al (20 mol%)
Element distribution formed by pressure wave

**LASER**
- Pulse width: 130 fs
- Pulse energy: 3 \( \mu \)J
- Repetition rate: 200kHz
- Objective: 50x, NA=0.85
- Irradiation Time: 5 sec.

**Si - Ca – Al – O glass**

Microscope

Backscattered electron

Sample glass

Irradiation of fs laser

Polish

EPMA
Example of silicon deposition
Glass prepared using metallic aluminum as the source material has oxygen deficiency centers. Si-O bonds are continuously broken and re-formed with numerous unbound atoms. Si and Al-rich structures are formed at the center of the irradiated area. Si rich structure grows into a particle because of the thermite-like reaction promoted by the heat treatment.