Towards ultra high density multi-layer disk recording by two-photon absorption

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ABSTRACT
The influence of the numerical aperture on a 3-D multi-layer optical data storage system is analyzed by simulation and validated experimentally. By increasing NA it becomes feasible to store more than a terabyte of data in a two-photon recorded 5 ¼” disk media.

Keywords: volumetric data storage, multi-layer data storage, two-photon recording, photochromic optical memory, fluorescent readout, high NA
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1. Introduction

Advances in healthcare imaging products and multimedia entertainment systems are demanding higher media capacity (TB/disk) and faster data transfer speeds (1Gb/s) from future removable disk based data storage systems. One promising direction to satisfy these requirements with low cost systems is the use of optical volumetric multi-layer disk media. At Call/Recall we have demonstrated the scalability of two-photon recordable photochromic doped polymeric Write Once Read Many (WORM) disk media to more than 100 layers with negligible interlayer crosstalk and excellent stability of the written bits1,2,3,4,5,6. The media employed in our WORM system, consist of photochromic organic molecules designed and synthesized so that they change their structure, upon excitation at the absorption band of the molecule5,7,8,9,10. A spot is written in the volume of the medium only at points of temporal and spatial intersection of two photons with sufficient energy to record by altering the structure of the photochromic molecule. A high power short pulse laser beam is tightly focused for recording. Recording occurs only within a small volume around the focus of the laser beam due to two-photon absorption. The recording response of the material follows the square of the optical system point spread function (PSF) resulting in a recorded bit size that is 30% less than the Rayleigh criterion PSF. The recorded bits are read by fluorescence when excited by suitable optical radiation absorbed within the written spot volume.

The doped polymer media is low cost, flexibly shaped and molded, and its properties may be customized (by changing the dopant molecules) to match evolving application and technology requirements. We had previously reported bit dimensions of 0.5 x 0.5 x 4.5µm² with NA =0.75 and 460nm wavelength exhibiting raw bit-error rates (BER) of 10⁻⁵. Here we show a further decrease in the bit size to 0.4 x 0.4 x 2µm with a higher NA=1.4 objective lens at 532nm wavelength, resulting in ultra high density volumetric disk recording using up to 100 layers and providing a potential for 400bits/µm² “effective areal density”. This paper discusses the theory and approach as well as experimental results we used and obtained to demonstrate the feasibility of high NA recording of volumetric multilayer disks by two photon absorption.

2. Bit size of ultra high density recording

The basic concept of the 2-photon multi-layer disk system is shown in Figure 1.

Figure 1. Two-photon multi-layer disk concept and experimental fluorescent confocal microscope images of data bits
For two-photon absorption recording, the excited molecular distribution is considered to be proportional to the square of the irradiance distribution of the recording laser. The recorded bit shape is modeled as:

$$P(x, y, z) = \alpha \times I^2(x, y, z)$$

(1)

with $\alpha$, a constant for a given media type that contains material parameters, and $I(x,y,z)$ the recording laser beam irradiance. At focus, the laser is Gaussian-shaped, giving:

$$I(x, y, z) = \frac{I_0}{\omega_0^2[1 + \left(\frac{\lambda z}{n \pi \omega_0^2}\right)^2]} \exp\left\{-\frac{2(x^2 + y^2)}{\omega_0^2[1 + \left(\frac{\lambda z}{n \pi \omega_0^2}\right)^2]}\right\}$$

(2)

where, $I_0/\omega_0^2$ is the peak irradiance, $\omega_0 = 0.61 \lambda/NA$ is the radius of beam waist, $\lambda$ is the wavelength and NA is the numerical aperture. An OPTISCAN\textsuperscript{12} simulation shows the irradiance squared, $I^2$, distribution of a $\lambda=460$nm, $NA=0.5$ system to have bit dimensions of $0.6*0.6*6\mu m^3$ as shown in Figure 2(a). Bit size is valued at the dimensions of $1/e^2$ of the peak fluorescence. Figure 2(b) is the image of a real experimental recorded bit obtained with an Olympus fluorescence confocal microscope having dimensions of $0.6*0.6*6\mu m^3$. This particular bit was recorded with a frequency doubled Ti:Saph mode-locked laser (76MHz repetition rate, 200fs pulsewidth) with wavelength of $\lambda=460$nm and 0.5NA recording optics. Bit dimensions observed agree very well with the simulated value of the irradiance squared axial point spread function of the 0.5NA optical system. As additional data points, Figure 2(c) shows the experimental recorded bit with the 532nm laser and 0.5NA objective lens and the bit size is about $0.7*0.7*7\mu m^3$ as expected. Figure 2(d) is the recorded bit with the 532nm laser and 0.75NA objective lens and the bit size is about $0.5*0.5*4.5\mu m^3$ as expected. The bit density for this recording is $3.3$Tb/in$^3$. The recorded bit using the 532nm laser also agrees well with simulation. This shows that the bit shape and size are modeled accurately when the NA and wavelength are specified.

Data bit areal size, longitudinal size, and data bit volume are simulated as a function of NA and compared with experimentally achieved data size and volume indicated in the plots. Figure 3(a) shows the data bit areal size $A = \pi r^2$, where $r = 0.61 \lambda/NA$ is the radius of the Airy disk. Figure 3(b) shows the longitudinal extent of the axial point spread function modeled in Matlab using physical optics tools. Figure 3(c) shows the data bit volume as a function of NA obtained by multiplying the values of the areal and longitudinal size. The simulations are indicated in Figure 3 by the solid curves and experimental values at NA=0.5, 0.75, and 1.4 are indicated by the dots along the curves. The experimental values and trends agree very well with the simulation where increasing NA reduces the data bit volume.

Figure 2. (a) Simulation bit: 460nm@ 0.5NA, bit size: 0.6*0.6*6\mu m$^3$; (b) Recorded bit: 460nm@ 0.5NA, bit size: 0.6*0.6*6.5\mu m$^3$; (c) Recorded bit: 532nm@0.5NA, bit size: 0.7*0.7*7\mu m$^3$; (d) Recorded bit: 532nm@0.75NA, bit size: 0.5*0.5*4.5\mu m$^3$
3. Disk capacity of ultra high density multi-layer recording

For a 2-photon 3-D optical data storage disk, the total areal raw capacity is expressed as:

\[ C_{\text{areal}} = \frac{\pi(r_{\text{max}}^2 - r_{\text{min}}^2)}{l_{\text{bit}} \times w_{\text{pitch}}} \]  

(3)

\( r_{\text{max}}, r_{\text{min}} \) are maximum and minimum recording radius of the disk, \( l_{\text{bit}} \times w_{\text{pitch}} \) is the bit size in the X-Y plane.

The volume raw capacity of a 2-photon 3-D optical data storage disk is:

\[ C_{\text{total}} = C_{\text{areal}} \times N_{\text{layer}} \]  

(4)

\( N_{\text{layer}} = T_{\text{disk}} / S_{\text{layer}} \) is the number of layers. \( T_{\text{disk}} \) is the thickness of the disc; \( S_{\text{layer}} \) is the layer separation. Generally, \( T_{\text{disk}} \) should be smaller than the working distance of the objective lens.

The working distance is defined as the longitudinal separation between the rear lens surface and the focal plane. When NA<1, a geometric first order plot of objective lens working distance vs. NA is straightforward to visualize as shown in Figure 4. For NA>1 there is no simple geometric description of the working distance. The objective lenses with NA>1 have traditionally been considered for single-layer data storage systems. However, upon experimental investigation of commercially available Olympus®
microscope objective lenses it is observed that there is significant working distance for the NA range between NA = 1.2 → 1.65 as shown in Table 1.

**Table 1.** Commercially available high NA oil-immersion Olympus® objective lenses.

<table>
<thead>
<tr>
<th>Objective</th>
<th>Item</th>
<th>NA</th>
<th>W.D. (µm)</th>
<th>Cover Glass Thickness (mm)</th>
<th>Immersion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plan Apochromat</td>
<td>PLAPO 60XO3</td>
<td>1.40</td>
<td>150</td>
<td>0.17</td>
<td>Oil</td>
</tr>
<tr>
<td></td>
<td>60XO3TIRFM</td>
<td>1.45</td>
<td>150</td>
<td>0.17</td>
<td>Oil</td>
</tr>
<tr>
<td></td>
<td>100XO3</td>
<td>1.40</td>
<td>100</td>
<td>0.17</td>
<td>Oil</td>
</tr>
<tr>
<td>UPLAPO</td>
<td>60XW3/IR</td>
<td>1.2</td>
<td>280</td>
<td>0.13-0.21</td>
<td>Water</td>
</tr>
<tr>
<td>Others</td>
<td>APO 100XOHR</td>
<td>1.65</td>
<td>100</td>
<td>0.15</td>
<td>Oil</td>
</tr>
<tr>
<td></td>
<td>MPlanAPO 100XO</td>
<td>1.40</td>
<td>80</td>
<td>0</td>
<td>Oil</td>
</tr>
</tbody>
</table>

In our experiments an Olympus® MplanApo 100X/1.4NA oil immersion objective lens having 80µm of working distance is used to record data bits inside the volumetric photochromic media using two-photon absorption techniques. Within this 80µm, multiple layers may be recorded if the depth of the bit is sufficiently small. Since the bit depth has been observed to be 2µm it is possible to record as many as 20-30 layers within 80µm of material. Figure 5 shows experimental results of fluorescent confocal microscope images of recorded data bits across multiple layers that are recorded within the 80µm working distance of this particular lens at different layer spacing. The 4-5µm layer separation shows good layer separation, and 2-3µm layer separation shows poor layer separation to minimize layer crosstalk. This indicates that for the bit size of 0.4x0.4x2µm that 4-5µm layer separation appears practical to achieve 20 layers with this objective lens achieving ~ 80bits/µm at a given radial location. If a longer working distance high NA objective lens is used, more layers can be recorded and the recording density of 400bits/µm can be realized.

**Figure 5.** Experimental results showing fluorescent confocal microscope images of recorded data bits using an Olympus® 1.4NA oil immersion microscope objective lens across multiple layers that are recorded within the 80µm working distance of this particular lens at different layer spacings. Data bits are scanned in depth y-z, across multiple-layers.
From the analysis of the recorded bit size the raw capacity of a 5.25” disk is calculated as indicated in Figure 6. Figure 6(a) shows the raw areal capacity as a function of NA at two wavelengths. Figure 6(b) shows the volumetric raw capacity with different recording wavelength and NA. Here, the $T_{disk}$ is equal to working distance of the objective lens. In order to increase the volumetric capacity (aerial density, and volumetric density) a tradeoff between high NA and lens working distance is considered, as well as recording laser power requirements that are now examined.

$$C_{\text{areal}} = \frac{\pi(r_{\text{max}}^2 - r_{\text{min}}^2)}{l_{\text{bit}}w_{\text{pitch}}}$$

5.25” diameter disk

$$C_{\text{total}} = C_{\text{areal}}S_{\text{layer}} = C_{\text{areal}}N_{\text{layer}}$$

4. Ultra high density recording laser power and speed

Two-photon absorption recording requires high peak power due to the fundamental low two-photon absorption cross-section. The numerical aperture influences not only the recorded bit size, but also the recording power required. For 2-photon recording, the required peak power, $P_{\text{peak}}$, of the laser can be expressed as:

$$P_{\text{peak}}^2 = \frac{\pi^2 2h\nu M_{\text{unit}} (0.61\lambda)^4}{D_M \sigma T \cdot t_p f_{\text{rep}}} \cdot \frac{1}{\text{NA}^4}$$

(5)

here, $h\nu$ is photon energy, $M_{\text{unit}}$ is recorded molecules per unit volume, $\lambda$ is recording wavelength, $D_M$ is density of the original unrecorded molecules, $\sigma$ is 2-photon cross section, $t_p$ is the pulse width of recording laser, $f_{\text{rep}}$ is the repetition rate of recording laser, and $T$ is recording time. Rearranging this equation, including the NA, energy, and photon arrival rate through,

$$\frac{1}{A^2} = \frac{\text{NA}^4}{\pi^2 (0.61\lambda)^4}, \quad E_T = E_{p}p^# = N_{p}h\nu p^#$$

(6)

the recording efficiency is obtained as

$$\eta = \left(\frac{\sigma}{2h\nu}\right) E_{p}P_{\text{peak}}p^# \left(\frac{\text{NA}^4}{\pi^2 (0.61\lambda)^4}\right)$$

(7)

Figure 6. Capacity vs NA for 5.25 inch diameter format (a) single layer, and (b) multi-layer that includes, for NA>1, representative working distances of commercially available high NA lenses, as shown in Table 1.
\[\sigma: 2\text{-photon cross section}\]
\[\lambda: \text{recording laser wavelength}\]
\[D_{M}: \text{density of unrecorded molecules}\]
\[M_{\text{unit}}: \text{density of recorded molecules}\]
\[\eta: \% \text{ of recorded molecules}\]
\[P_{\text{peak}}: \text{peak laser power}\]
\[t_{p}: \text{pulse width of recording laser}\]
\[f_{\text{rep}}: \text{repetition rate of recording laser}\]
\[f_{\text{data}}: \text{recording data rate}\]
\[h\nu: \text{photon energy}\]
\[P_{\lambda}: \text{recording laser wavelength}\]
\[p#: \# \text{of pulses}, p# = f_{\text{rep}}/f_{\text{data}}\]
\[N_{p}: \# \text{of photons/pulse}\]
\[N_{p}p#: \text{total \# of photons}\]
\[E_{p} = P_{\text{peak}}t_{p}: \text{pulse energy}, \quad E_{T} = N_{p}h\nu: \text{total energy required to record a bit}\]
\[d = 1.22\lambda/NA: \text{d, focused spot diameter}\]
\[A = (\pi/4)d^{2}: \text{A, area of focused spot}\]

**Figure 7.** Legend of material, recording laser parameters, and optical system properties that influence the 2-photon recording efficiency, \(\eta\).

Figure 7 describes the system, laser, and material properties used in the above equations. Figure 8(a) plots the recording laser parameters of total energy to record a data bit multiplied by the photon arrival rate, or peak power, vs. NA for a constant recording efficiency, \(\eta\). From Figure 8(a) it is observed that the inverse 4th order, NA\(^{-4}\), dependence can dramatically reduce the recording laser requirements to achieve 2-photon recording. Figure 8(b) plots the recording efficiency, \(\eta\), as a function of NA for constant energy and laser properties and also shows a NA\(^{4}\) dependence that increases the localized recording efficiency.

**Figure 8.** (a) NA dependency of recording laser properties with constant recording efficiency. (b) NA dependency of the recording efficiency, \(\eta\), with constant recording laser properties.

To illustrate this trend a 532nm Nd:Vanadate laser having a pulse width of 6.5pSec and repetition rate of 76MHz was used in experiments. Figure 9 shows the static recording setup where the recording laser is modulated by an AO modulator, collimated, and then launched into the high NA objective lens. With a 0.5NA objective lens a 1Mb/s recording speed is achieved with average power of 1.5W and peak power of 3kW. When an Olympus® MplanApo 100X/1.4NA oil immersion objective lens is used, data bits inside the volumetric photochromic media can be recorded with lower laser power, or with a higher recording speed. Figure 10 shows fluorescent confocal microscope images of recorded data bits using an Olympus® 1.4NA oil immersion microscope objective lens. Two different recording conditions were investigated. The first recording condition uses 200mW of average power (400W peak power), and an exposure time of 1us corresponding to a 1Mbit/s data rate (To maintain same recording data rate as used with the 0.5NA optical system). These conditions lead to a data bit size of 0.4x0.4x2\(\mu\)m\(^3\) that agrees well with the 0.35x0.35x1\(\mu\)m\(^3\) bit size that was predicted by our simulations. This experiment also demonstrates that using 1.4NA we can reduce the average recording power to 200mW by 5 times relative to 0.5NA for the same 1Mbit/s recording rate.

A second recording was carried out with 900mW of average power (2kW peak power) and exposure time of 100ns or 10Mbit/s data rate, demonstrating a 10 X increase in recording data rate when compared to the performance of a 0.5NA recording with similar recording power. The 900mW average power recording at
10Mbit/s results in a similar data bit size of 0.5x0.5x2\(\mu\)m\(^3\) that agrees well with theory. Also, the number of pulses used to record a data bit at 10Mbit/s is reduced to 7 pulses where 76 pulses were used at 1Mbit/s.

Figure 9. Static recording setup to experimentally evaluate 1.4NA objective lens recordings. Olympus® confocal fluorescent microscope used to evaluate the recorded data bits.

![Recording Setup](image)

Figure 10. Experimental results showing fluorescent confocal microscope images of recorded data bits using an Olympus® 1.4NA oil immersion microscope objective lens for recording. Data bits are scanned radially in x-y as well as in depth y-z.

5. Ultra high density multi-layer disk readout

During readout, fluorescence is emitted in 4\(\pi\) steradians. The objective lens collects only a small portion of the fluorescence, as shown in Figure 11(a). The substrate of the disk is PMMA, \(n_1=1.492\), Figure 11(b) shows the relationship of the collection angles in the interface of the disk and air. The critical angle affects the maximum solid angle that can be collected. Total collection efficiency is the ratio of the collected solid angle to 4\(\pi\):

\[
\eta_{\text{Collection}} = \left(1 - \sqrt{1 - \left(\frac{\text{NA}}{n_1}\right)^2}\right)/2
\]

(9)

here, \(\text{NA}=n_2\sin\theta_\text{lim}\) is the numerical aperture of the objective lens. Figure 11(c) shows the collection efficiency for different NA. Total collected fluorescence is defined as:
where, $V_{bit}$ is the recorded bit size. Figure 11(d) shows the collected fluorescence vs. NA. The data is normalized with the data at 0.5NA. After normalization, the relationship is approximately the same for 532nm and 460nm recording wavelengths. This analysis assumes the fluorescence emitted per unit volume is the same. The collected fluorescence is affected by the increase in collection efficiency and decrease of the bit size. Figure 11(d) shows a static readout experimental result. In the experiment, the bits are recorded with 0.5NA objective lens and readout by 0.7NA objective lens and 1.4NA objective lens separately. The intensity of the readout fluorescence is compared. The experimental result shows similar performance with the simulation result. Although the recorded bit size is smaller with the higher NA optical system leading to less readout fluorescence; the higher collection efficiency of the high NA optical system compensates the loss of the readout fluorescence caused by the smaller bit size.

\[ F = \eta_{collection} \cdot V_{bit} \]  

6. Summary

A high NA objective lens (NA>1) with a long working distance (>100um) used together with a multilayer volumetric optical media provides an important direction towards the realization of ultra high density
multi-layer recording by two-photon absorption. A high NA objective lens decreases bit size, increases the volumetric density, and reduces the recording laser power requirements for a given recording speed. Static recording experiments have verified the recording potential performance of a high NA lens utilizing a commercially available Olympus® MpPlanApo 100X/1.4NA oil immersion objective lens with recording lasers. Based upon these results, 1.2TB capacity can be achieved in a 5.25” format disk of 80µm of material thickness. Recording results at 1.4NA indicate ~10x reduction in peak recording power at 1Mb/s or recording data rate increase of 10x to 10Mb/s with similar peak powers used at 0.5NA recordings. In addition our simulation and static readout experiments show that a high NA objective lens collects more fluorescence from a smaller volume and compensates the loss of the readout fluorescence caused by the smaller recorded bit size.

7. References

13 Olympus objective lens data sheet.