# Three-dimensional multilayered fluorescent optical memory using two-photon reduction of Au(III)-ions

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## 1. Introduction

Recent progress in the increase of the capacity of optical memory devices has extended the application of the optical memory not only for storing computer data but also for recording the Hi-Fi music, photographs, video clips, and so on. However, due to the nature of light, the recording density of the optical memory is fundamentally limited by the minimum of the spot size, which is almost as large as half the size of the wavelength. To overcome this limitation and to realize the next generation large-capacity optical memory system, we have studied three-dimensional multi-layered optical memory recording [1-4]. In this method, the longitudinal (z) direction is utilized in addition to the lateral (x-y) dimension, and the data are stored not on the surface of the recording medium but three-dimensionally inside a thick recording medium.

## 2. Developed recording material for 3D optical memory

To develop three-dimensional multi-layered optical memory, an optical pickup must be able to write and read data on and from a particular layer without crosstalk from the adjacent layers. For this purpose, we focused on the three-dimensional imaging properties of the two-photon and confocal laser scanning fluorescent microscopy. Because both two-photon and confocal laser scanning fluorescent microscopy have a transmission band on the kz axis, which corresponds to the longitudinal direction; as well as the high lateral resolutions.[5, 6, 7]. This means that two-photon and confocal fluorescent microscopy can be utilized as the pickup in three-dimensional multi-layered optical storage. In order to use these methods for optical system, the recording medium should store the bit-data as a fluorescent pattern inside it. Hence, we developed a recording medium that can record bit data as a fluorescent pattern three-dimensionally inside a thick medium.

The recording medium we developed is a rhodamine-B (Rh-B) and Au(III)-ions doped poly-methyl-methacrylate (PMMA) [8]. Figure 1 illustrates the mechanism of recording 3D fluorescent pattern inside the medium. When Au(III)-ions exist in the vicinity of a Rh-B molecule, the energy of the optically excited Rh-B molecule is transferred to the Au(III)-ions and the Rh-B molecule is quenched; as a result, no fluorescent light is emitted (Fig. 1(a)). By irradiating femto-second near-infrared laser (fsec NIR laser) on this medium, Au(III)-ions are photochemically reduced by the two-photon absorption and changed to gold nano-particles whose diameter are around 10 nm (Fig. 1(b)). Since the photo-reduced gold particles do not quench the Rh-B molecules, the Rh-B molecules are re-activated and become fluorescence-emissive (Fig. 1(c)). Using this mechanism, we recorded bit data as a 3D fluorescent pattern inside the developed medium. The fluorescent signals from the recorded data were read by using a pick-up system that incorporates confocal fluorescent microscopy.

## 3. Experimental results

Figure 2 shows an experimental result of the fluorescent pattern recording. The distances between dots were $1 \times 1\mu m$. Orange area in this photograph is corresponding to the fsec-laser-irradiated area. From this result, the Rh-B molecules were activated and emitted fluorescence only in the area irradiated of fsec laser, while the fluorescence of Rh-B was eliminated by Au(III)-ions in the non-irradiated area, and the high contrast fluorescent recording was done by using interaction between Au(III)-ions and Rh-B molecules, and two-photon induced photoreduction of Au(III)-ions to the gold nano-particles. Figure 3 shows the result of three-dimensional multi-layered fluorescent recording. Five layers were recorded inside the developed recording medium at 10 μm distances. In each layer, the bit data were written every $5\times5$ μm. Figure 3(a) shows the top layer, which is nearest to the surface of the medium, and (b), (c), (d), and (e) are second, third, fourth, and fifth layers, respectively. These results exhibit that the developed medium can record the fluorescent bit-pattern three-dimensionally, and that the recorded data on each layer are read with sufficient lateral and longitudinal separation by the use of a confocal fluorescent microscope

pick-up.

    Figure 4 shows the multi-layer structure inserting the transparent buffer layer between the recording layers. The introduction of the buffer layer realizes high longitudinal resolution because the size of recorded bit is restricted to a value smaller than that of the focused spot [9]. We used above mentioned Rh-B and Au(III)-ions doped PMMA medium for recording layers and polyvinyl alcohol for buffer layers. The recording layer of 1.2μm in thickness and the buffer layer of 4.2μm in thickness were spin-coated alternately on the glass substrate without any interferences or detachment of the films. Inside this recording medium, 3D bit pattern was recorded, and it was read out by using the confocal pick-up. Figure 4(b) shows experimental result of the longitudinal cross-section of the recorded bits. This figure demonstrates good axial separation of the recorded bits along the z-axis.

    Figure 5 shows the multi-layered optical memory cube.

## 4. Conclusion and Discussion

    We have developed a novel recording medium for three-dimensional multi-layered optical memory. The material can store the bit data three-dimensionally as a fluorescent pattern using the energy transfer between Rh-B and Au(III)-ions doped PMMA. In addition, we have already developed the structured multi-layer optical disk in which recording layers and buffer layers were alternatively fabricated. Incorporation of the buffer layers realizes high axial resolution and low cross talk. In my presentation, I will talk also about this three-dimensional memory disk with an experimental result of three-dimensional focusing and tracking servo system.
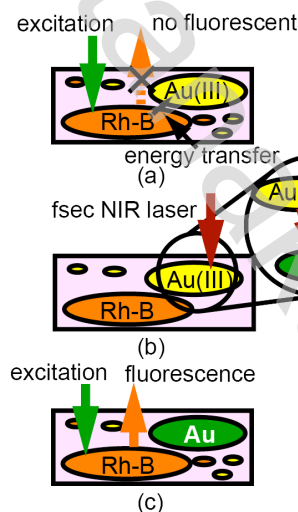
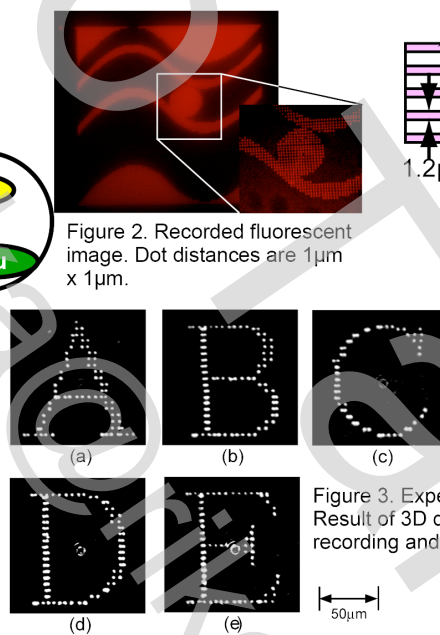Figure 1. The recording mechanism of the fluoscent bit data.

Figure 2. Recorded fluorescent image. Dot distances are 1μm x 1μm.

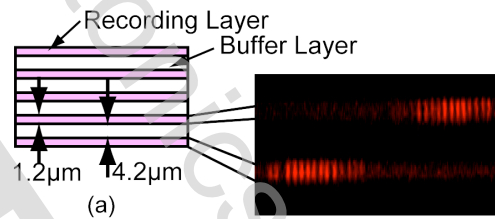Figure 3. Experimental Result of 3D data recording and readout.

Figure 4. The recording mechanism of the fluoscent bit data.

Figure 5. Multi-layered optical memory cube.

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