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4. TITLE AND SUBTITLE
3D Digital data-storage using Two-Photon process: A Demonstration task / Novel multifunctional chromophores for electro-optic and other photonics applications

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13. SUPPLEMENTARY NOTES

14. ABSTRACT
New opto-electronic and composite materials with enhanced photonic performance were developed and their applications in 3D optical data-storage, 3D optical-circuitry, 3D micro-fabrications and photorefractivity were studied. Two-photon based technology was used to create cross-linking at a precise volume location to produce complex 3D optical-circuitry such as waveguide splitters and grating couplers as well as for fabricating MEMS. Another area of our activity has been in preparation of photonic band gap materials for visible and near-IR range. Highly monodispersed silica and polystyrene spheres of varying diameters(100-800nm) were prepared and photonic crystals made from them showed photonic pseudogaps. We have successfully infiltrated the pores with a high refractive index conjugated polymer, PPV and etched out the silica spheres. The resulting inverted photonic crystal exhibits enhanced reflectance at the band gap. Using a new approach of photosensitization with nanocrystallites of inorganic semiconductors, we have prepared photorefractive composite for operation at 1.31μm and this approach can be extended to preparation of photorefractive materials with a tailored spectral response. Our new materials development in collaboration with Wrigley has made it possible to use two-photon approach to write information in different layers with the capacity of terabits/cm³, which can now be read by using a simple CW green laser. A three dimensional digital data-storage system based on two-photon process was demonstrated. We have written layers of digital data in a dye doped polymer media (AF-240 from Air force Polymer branch doped in PMMA) using highly localized two-photon process. We have demonstrated a new data read back system using a 532nm solid state laser and were able to read the data using confocal single photon fluorescence detection.

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Abstract:

New opto-electronic and composite materials with enhanced photonic performance were developed and their applications in 3D optical data-storage, 3D optical-circuitry, 3D micro-fabrications and photorefractivity were studied.

Two photon process based approach has allowed us to use photo-crosslinking at a precise volume location to produce complex 3D optical-circuitry such as waveguide splitters and grating couplers as well as for fabricating MEMS. Another area of our activity has been in preparation of photonic band gap materials for visible and near-IR range. Highly monodispersed silica and polystyrene spheres of varying diameters(100-800nm) were prepared and photonic crystals made from them showed photonic pseudogaps. We have successfully infiltrated the pores with a high refractive index conjugated polymer, PPV and etched out the silica spheres. The resulting inverted photonic crystal exhibits enhanced reflectance at the band gap. Using a new approach of photosensitization with nanocrystallites of inorganic semiconductors, we have prepared photorefractive composite for operation at 1.31µm and this approach can be extended for preparation of photorefractive materials with a tailored spectral response.

Our new materials development in collaboration with Wright lab has made it possible to use two-photon approach to write information in different layers with the capacity of terabits/cm³, which can now be read by using a simple CW green laser. A three dimensional digital data-storage system based on two-photon process also was demonstrated. We have written layers of digital data in a dye doped polymer media (AF-240 from Air force Polymer branch doped in PMMA) using highly localized two-photon process. We have demonstrated a new data read back system using a 532nm solid state laser and were able to read the data using confocal single photon fluorescence detection.
3D Fabrication:

A novel approach was used for micro-fabrication of three-dimensional optical circuitry using in-situ two-photon assisted polymerization in an as-formed bulk sample. The bulk media consisted of a blend of photo-curable and thermally curable epoxies. 1x2 and 1X4 splitters were fabricated inside the volume and imaged by confocal microscopy. End-fire coupling of a He-Ne laser beam into these splitters was also achieved. By controlling the...
size and shape of the waveguides, we were able to make single mode or multimode waveguides.

**Photonic Band Gap Materials:**

We have achieved successful fabrication of highly monodispersed silica and polystyrene spheres. The size of these spheres can be varied between 100 and 800 nm to allow for proper tuning of the photonic crystal properties. Optical absorption of the silica photonic crystal shows the presence of a photonic pseudogap. The sphere size and thermal treatment can easily control this gap. Strong angular dependent Bragg reflections were also measured. This supports the SEM result that the crystal is packed throughout the bulk. We have successfully infiltrated the pores with PPV and etched the silica spheres. The resulting inverted PPV photonic crystal exhibits enhanced reflectance at the band gap. We have also successfully fabricated inverted titania and polystyrene photonic crystals.

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**SiO₂ Photonic Crystal**

**Inverted TiO₂ Photonic Crystal**

**Inverted polystyrene Photonic Crystal**
Optical Data Storage:

We also developed novel photorefractive nano-composites for applications at communication wavelengths. A new approach of using inorganic semiconductor quantum dots of well-defined excitonic resonance for photosensitization was used. These quantum dots were dispersed in a hole trapping polymer containing electro-optic chromophore to prepare the photorefractive nano-composites. Using PbS quantum dots as photosensitizers we demonstrated, to the best of our knowledge first time, photorefractive operation in a polymer composite at 1.31μm.

We have achieved a high density data-storage with gray-scale control in multiple planes as stacked Compact Disks (CDs) at a separation of 10 μm, using a polymer block doped with a highly efficient two-photon dye. The absorption and fluorescence of the dye at the written spot shift to a longer wavelength, permitting easy fluorescence mode readout with a linear excitation using an inexpensive laser source.

The storage capacity in this case is estimated to be $10^{12}$ bits/cm³. To demonstrate this technique we used Poly-methyl methacrylate (PMMA) doped with dye AF240 (7-benzothiazol-2-yl-9, 9-diethylfluoren-2-yl) diphenylamine developed at Airforce
Research Laboratory at Dayton, as the storage medium. A tightly focussed pulsed IR beam (A Ti:Sapphire laser operating at 800nm with pulse width 80fs and a repetition rate of 90MHz as light source and a high NA objective for focussing) was used to write barcodes in to the dye doped polymer medium mounted on a computer controlled scanning stage. This way we were able to write multiple layers of barcodes/images in a single polymer block at a vertical separation of 10 microns, and up to a depth of couple of hundred microns. This can be used to make security identification labels with secret identification codes well concealed deep inside the label.on in a polymer composite at 1.31μm.

Digital Datastorage:

Extending the above mentioned technique, a digital datastorage, system based on two-photon process also was demonstrated. We have written layers of digital data in a dye doped polymer media (AF-240 from Air force Polymer branch doped in PMMA) using highly localized two-photon process and were able to read the data using confocal single photon fluorescence detection. The data is written into the medium using femto second pulses from a Ti:Sapphire laser tuned to 800nm, and the fluorescence change induced in the medium is detected by single photon confocal detection.

We have demonstrated a new data read back system using a 532nm solid state laser, which shows promise towards the miniaturization of the read back system. These solid state lasers can be compact and inexpensive compared to the bulky gas lasers. We have designed a new fiber coupled read back system and developed new software for this purpose. In the new reader, the single photon fluorescence signal is delivered to a photo multiplier tube using a fiber delivery system. This signal is digitized and analyzed to retrieve the stored digital data. To make the system compatible with the existing CD system, we have adopted an edge detection algorithm, so that any change in fluorescence (edge of the written bit) will be taken as 1. With this system we are projecting a storage capacity of ~0.3Tb in a CD sized disk with 5mm thickness. The current system works with a writing speed of 1Kb/s and reading speed of ~10Kb/s.

The schematic diagram of the current digital data writer and reader are shown in the figure given below (figure 1 & 2). A confocal microscope image of the data bits written using this system and a diagram of the signal pattern obtained from the data
reader (figure 3) with the digital conversion using the edge detection algorithm are shown in the figure 4.

**Approach: Digital Writing**

![Diagram of Approach: Digital Writing](image)

**Approach: Digital Data Reading**

![Diagram of Approach: Digital Data Reading](image)
Confocal Microscope image of the bit pattern
Written in polymeric media

Fig. 3

Fig. 4

Original Signal

Thresholded signal

10111000011110000010101001010