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Optically Stimulated Luminescence Based Optical Data Storage

May 2016

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Nigel Browning
Eric Jensen
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Nathan Canfield

U.S. DEPARTMENT OF
ENERGY

Prepared for the U.S. Department of Energy
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Executive Summary

We design, built, and demonstrated a photocathode based pulsed electron system to produce optically stimulated luminescence (OSL) for next generation data storage. We successfully demonstrated 10ps pulses of up to 10^6 electrons and used those pulses to form OSL patterns in LiF without causing mechanical damage. By adjusting the accelerating voltage, we were able to vary the ratio of red to green optical emission, demonstrating a potential path for increasing storage density using orthogonal encoding. We also modeled a new approach to reading data that exploits the unique nature of OSL based data storage where the writing and reading processes have different size scales. Based on the technical demonstrations, we developed a roadmap to increase the storage density beyond 50TB using a standard 5.25" optical storage disc.

Introduction

The goal of this program was to demonstrate the fundamental feasibility of creating optically stimulated luminescence (OSL) patterns in LiF at a size and speed compatible with high density data storage. Based on earlier design work, we targeted a BluRay™ compatible pattern, with 150nm target spot size and <1ns write time. To demonstrate this writing, we required a tunable pulsed electron source capable of generating variable intensity highly-focused electron pulses at high repetition rates. Specifically, we needed a high-speed variable electron gun capable of generating a variable intensity electron pulse containing between 10^2 and 10^5 electrons with an accelerating voltage of 1-10kV focused into a spot size <1μm in diameter at a pulse rate >10MHz (with path to 300MHz). This combined performance cannot be achieved by commercially available electron sources. The primary technology path selected for this work is a photocathode, where an easily modulated UV laser is used to stimulate electron emission.

Key challenges identified and addressed by this work:

- Demonstrate ability to form OSL centers with ultra-short e-beam pulses
- Determine if LiF will be ablated by high e^- flux
- Identify market and determine suitability of the technology.

Table 1: Gantt chart for CRADA project

	Month										
Tasks	1	2	3	4	5	6	7	8	9	10	11
Phase 1 - Design											
1.1 Go/No Go review of design		X									
1.2 Build variable e-beam				X							
Phase 2 - System Build											
2.1 Complete laser build					X						
2.2 Integrate photocathode						X					
Phase 3 - Demonstration											
3.1 Substrate holder for LiF					X						
3.2 Expose OSL with variable pulse								X			
Phase 4 - Data patterning											
4.1 OSL media disc								X			
4.2 Rotating substrate holder									X		
4.3 Demonstrate data writing											X

Background

To meet the ever-increasing data storage need for cloud computing, scientists and engineers are constantly pushing the limit of data storage density. Hard disk drive (HDD) has been the dominant computing data storage device for the past 50 years, with over 800 million units sold in 2012. Until recently, HDD density increased by 100% every year, but advancing technology has decreased this rate to 25%. Solid-state hard drive (flash) is the storage method based on ferroelectricity, where direct wiring enables immediate access to the bit. However, flash suffers memory wear and thus can be used only as short-term storage. Although CD, DVD, and Blu-ray are popular optical data storage methods, they have a significantly lower density (under 10% of HDD). Still another recent advanced storage method is holography, a promising technology because of its large density and fast data access rate, reading a page each time instead of a bit (like flash). However, sensitivity to vibration and extremely low speed have prevented this technology from succeeding.

We are developing a new data approach that is capable of making a paradigm change in storage technology by using optically stimulated luminescence (OSL) to enable storage of more than a single bit of data in a spot on a disc. This will enable very high density rapid access data storage that builds on the extensive existing hardware technology base in BluRay™ optical disc drives. OSL has been in use for many years as a radiation dosimetry technology. OSL media has an extraordinary linear optical response (over eight orders of magnitude for LiF) that will enable reliable encoding of multiple bits of data in a single point, thus storing more data in each defined location.

Photocathode pulsed e-beam system

In order to simplify the generation and focusing of the electron beam, a JEOL 5900 scanning electron microscope (SEM) was modified to enable integration of a photocathode. The SEM was a very versatile tool which allowed for multiple configurations of the beam in terms of acceleration voltage, apertures, magnification, etc. ***Figure 1*** shows an image of the SEM used in this project.

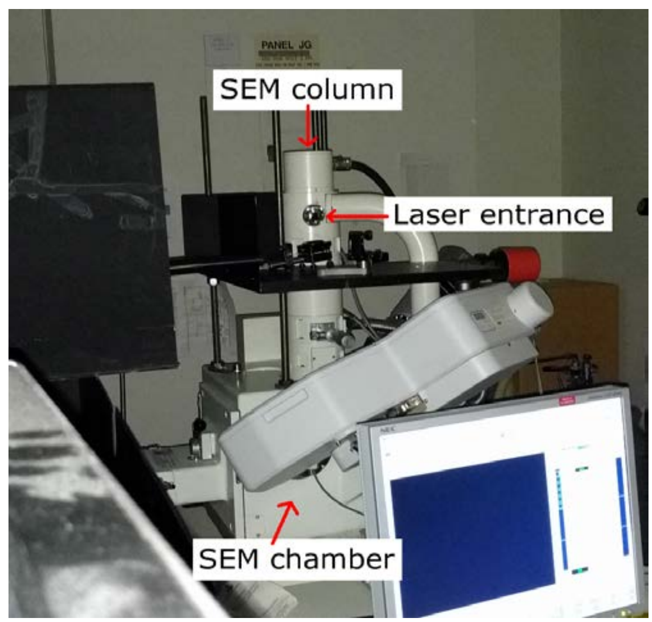

Figure 1 The JEOL 5900 SEM with platform for optical stimulation

In the original configuration the SEM used a thermal source of electrons which were channeled through magnetic lenses and mechanical apertures down to the sample. These different components provided a wide range of possible settings for the electron source which was ideal for the experiments as it provided many methods with which we could tune the output.

In order to generate the extremely short electron pulses, a photocathode system was selected, which is based on Einstein's photoelectric effect [1] [2]. A metal target is illuminated with a laser that has an energy above the metal's work function, exciting electrons in the metal into the vacuum level. Because the electron emission is controlled by the optical beam, it can be quickly and easily modulated at very high frequencies. The electron generation method was previously done at PNNL for the dynamic transmission electron microscope (DTEM) project. For help in designing and optimizing the setup Dr. Andreas W. Schroeder, a professor at the University of Illinois at Chicago, was contracted to help based on his previous work on supporting development of the DTEM.

For most metals, the work function correlates to an energy level in the ultraviolet. To generate short pulses, a Fianium infrared fiber laser capable of 10ps pulses at up to 40MHz and 25W CW equivalent power was selected. The laser beam is then up-converted to UV by frequency quadrupling in a two stage process. The end result is a 266nm UV laser beam that is then transmitted through an optical viewport into the top of the SEM column where the photocathode is located and the electrons are generated. Figure 2 shows part of the optics table with the laser in operation. The wavelength of the laser light was halved twice. Once from infrared to visible (green) and then from visible to ultraviolet (higher energy).

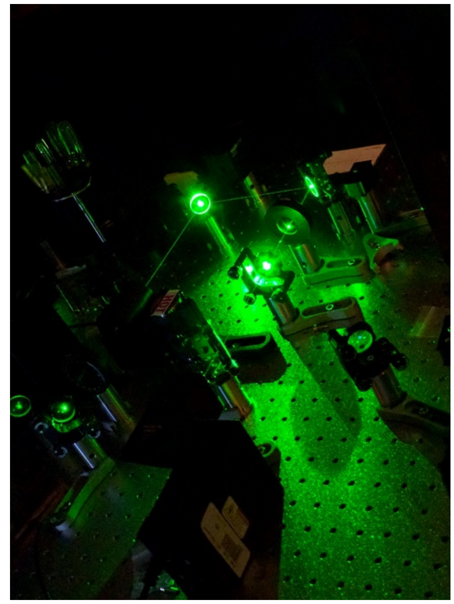

Figure 2 Laser drive system. The green light is from the second harmonic generation

After emission, the electrons were collected into the column with a high electric field. The SEM system is capable of operating between 1 and 30keV, but was operated typically between 5 and 20keV. Below 5keV, the number of electrons that were successfully collected down the column was too small to be usable. Above 25keV, the electrons generated X-rays that were energetic enough to penetrate the viewport, which would require additional shielding and monitoring. Once the electrons are collected into the SEM column, a set of electromagnetic lenses then focuses the electrons down to the sample. It was possible to use the laser-induced electrons to form an image, but the quality was significantly below that which was normal for the SEM since the SEM was optimized for imaging a larger number of electrons.

Photocathode design

When selecting the source for the electrons it was important to select a material which had a work function at a level which matched the incident photon energy. Both tantalum and tungsten were considered as stable refractory materials with usable work functions and reasonable quantum efficiency. Tungsten has a lower quantum efficiency (10^{-6}) than tantalum (10^{-5}), but is very mechanically stable even at high temperature and readily available as it is the standard material for SEM filaments, including for the JEOL 5900.

The standard design for photocathodes is a flat plate that allows the laser to be easily focused and emit electrons in a well-defined directional plume. The curvature of a tungsten or tantalum wire, without such a platform, would spread the emitted electrons in a large arc which would be difficult to focus at the end of the SEM. Based on this, we worked with Energy Beam Sciences of East Granby, CT to design and build Ta filaments with flat surfaces. Two designs were selected, one with a 0.5mm round disc and one with a 100µm flat ribbon bent into a similar shape as a traditional filament. However, despite several attempts, the supplier was unable to successfully manufacture a stable filament that would maintain geometry after heating. They were able to make tantalum wire filaments with the same shape as the standard tungsten filaments. Both tantalum and tungsten wire filaments were used during the course of the project.

Demonstration of photoemission

In order to measure the electron emission, the average beam current was measured using the Faraday cup built into the SEM stage. While each electron pulse has a high instantaneous current, the duty cycle is very low. The effective beam current can be calculated from the pulse density, pulse rate and electric charge as shown in equation 1. For a pulse density of 1×10^5 electrons and a pulse rate of 100kHz, the measured beam current should be 1.6nA. This pulse density is targeted because it was previously determined to be the approximate saturation density for a 150nm diameter exposure spot.

$$current = \frac{pulse\ density \times pulse\ rate}{Coulomb\ per\ electron} \tag{1}$$

Initial testing showed extremely high currents of 10-100nA, far beyond the expected pulse energy. Integrating a lock-in amplifier to the current measuring circuit revealed that there was a large steady-state thermal emission current due to the heating of the filament by the laser beam. Analysis of the beam energy indicated that the filament was likely being heated by no more than 50°C and by reducing the filament heating current, the thermal emission was eliminated. With optimized alignment, steady state beam currents up to ~2.76nA were measured correlating to a per-pulse electron density of 3.45×10^5.

OSL Media

Multiple LiF coated substrates were prepared using electron beam evaporation. In order to be able to investigate how the electron pulse interacts with the LiF coating, media samples were made with three different substrates, two different LiF thicknesses, two different metal coatings, and three different substrate deposition temperatures. In total 124 samples were produced.

Substrate selection

Given the above considerations three different substrates were selected: silicon wafers [3], sapphire wafers, and optically polished polycarbonate. In all cases the surface is polished, which was ideal for the experiments since it minimizes surface roughness contributions.

The silicon substrates were standard electronics grade wafers. Silicon is extremely flat and the semiconductive nature helps reduce charging. The sapphire substrate is a transparent non-conductive oxide with a surface flatness and roughness similar to the silicon substrates. **Figure 3** shows some sapphire substrates after deposition. The polycarbonate substrates were optically polished and represent a typical material for a production data storage disc.

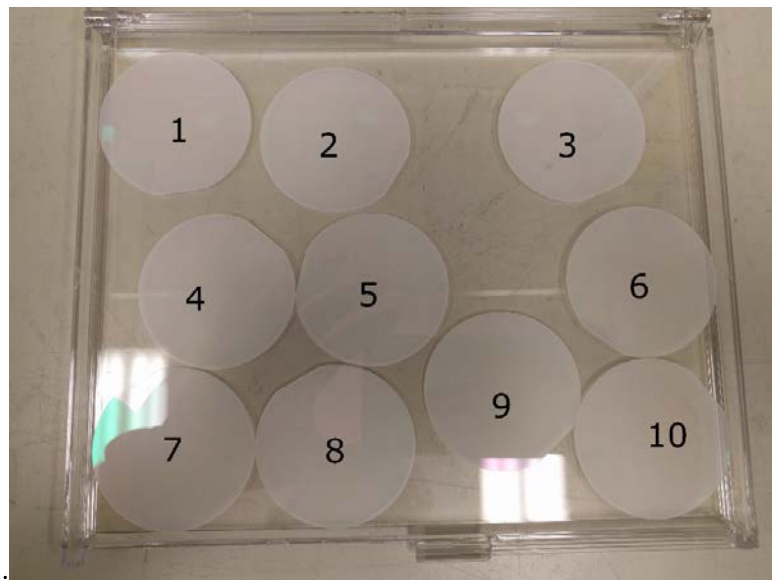

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Figure 3 A box containing sapphire samples coated with LiF. The sapphire disks were 2 inches in diameter. The numbers refer to the sample tracking number.

LiF deposition thickness

Two different target deposition thicknesses were selected, 100nm and 1000nm. **Figure 4** shows the estimated penetration depth of electrons into LiF based on acceleration voltage (theoretical: [4] [5], empirical: [6]). Based on these estimates it was concluded that for low acceleration voltages a 100-150 nm thick LiF layer might be penetrated, but a thick 1000 nm (1 µm) LiF layer would have less interaction with the substrate and therefore would mostly exhibit substrate-independent phenomena. Furthermore, for a low atomic number material, such as LiF the interaction volume is roughly shaped like a pear. The first few nm into the material the volume has the same size as the electron spot, however, the further the electron travels in the material, the higher the interaction probability. Eventually electrons reach a certain depth and are scattered uniformly. This results in a thin neck at the substrate surface and a wider sphere further down. If the material is very thin the electrons never reach the depth at which they are scattered uniformly and therefore a conductive substrate could potentially absorb the electrons that penetrate a thin LiF layer and reduce backscattering. This would enable the affected area to be almost identical to the size of the beam spot. A non-conductive substrate would probably scatter electron back through the LiF layer increasing the dose the area received while also broadening the spot.

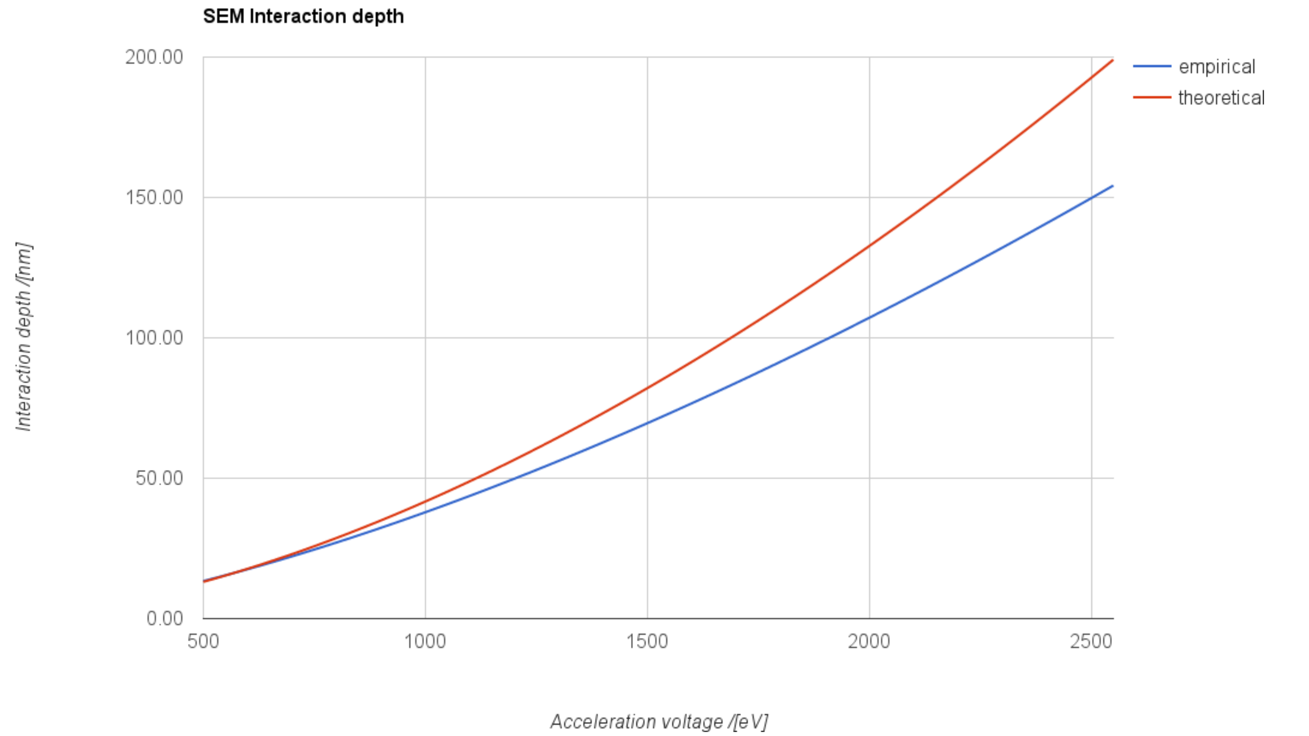

Figure 4 Theoretical and emperical estimates of the penetration depth of electrons.

Additionally, literature suggests [3] that F-centers are more probable at grain boundaries and therefore the grain size of LiF would be relevant. A thin LiF layer would be more sensitive to the grain structure of the substrate and more sensitive to temperature changes in the substrate during deposition.

Metal coating

There were two concerns with regards to the electron pulse: (1) it was too high an energy density and would ablate the LiF upon contact, and (2) the pulse would scatter off the substrate and significantly increase the size of the spot. In order to minimize such contributions two coatings were applied to some of the substrates.

A thick 3 µm aluminum coating was applied to some of the substrates prior to LiF sputtering. This coating is very conductive and very thick so it should absorb penetrating electrons regardless of the substrate.

A thin 500 nm silver coating was applied above the LiF on some of the substrates. This coating has a high atomic number, is conductive, and thin. It was hypothesized that this coating would have a retarding effect on the beam, but would not completely absorb the beam or scatter it completely.

Deposition temperature

Since literature suggests [3] that the grain size would affect the F-center concentration, and therefore the fluorescence, some of the substrates were kept at an elevated temperature during LiF deposition. Some substrates were kept at 150 °C since this was the maximum the heating system could handle and some were kept at 123 °C since this was close to the theoretical evaporation temperature of fluoride.

Demonstration of OSL writing

When writing fluorescent structures with the electron beam there were four different types: squares, lines, multi-pulse spots and single-pulse spots.

The intended spot size for the final product was 100 – 150 nm which is below the resolution limit of the high-end fluorescent microscopes we used to image our results. However, the fluorescent signal can still be detected despite the source being below the resolution limit. Therefore, it was important to locate the written spots correctly in order to differentiate them from possible reflective surface contaminants. This was achieved by writing large squares or rectangles, 200-300 µm on a side. These structures were both distinctive in shape and large enough to be easily identified in the fluorescence microscope. Figure 5 shows an example fluorescent square.

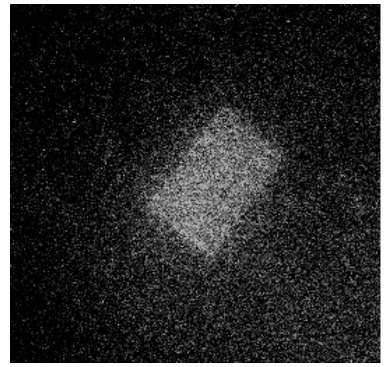

Figure 5 Luminesence pattern from a LiF sample exposed by SEM

In order to measure the approximate beam diameter, the beam was held fixed in a specific location and the sample was moved underneath it. When the movement is slow enough, the pulsed electron beam creates a continuous line of fluorescence in the material (Figure 6). The line width is a good first approximation to the width of the spot. In these samples, the measured linewidth is less than 0.8µm which is the measurement limit of the luminescence microscope.

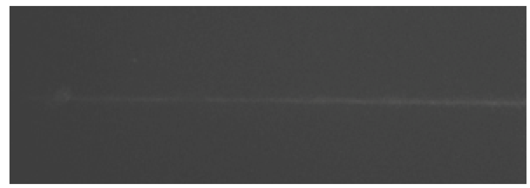

Figure 6 Linear exposure pattern formed by scanning the sample under a pulsed electron beam

A key question that this project was designed to answer was whether the electron beam would ablate the LiF. The electron pulse density correlates to a steady state current well above the damage threshold for LiF, however the extremely short pulse length should mitigate the effect. In the line tests, no signs of damage or heating were observed even though the average energy was much higher than what would be observed in a data-storage application. To further test this, the sample was held fixed under the pulsed beam. Figure 7 shows a single spot created with ~$3x10^6$ pulses by holding the sample still for 30 seconds. The dark spot in the center is due to the ablation of the LiF showing that at quasi-continuous operation, the current density is high enough to cause damage.

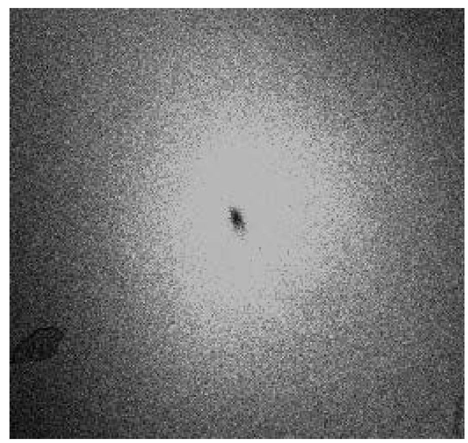

Figure 7 Ablation damage from an overexposed sample

Patterning of discrete spots

In order to write spots with a pulsed electron source it is necessary to move the substrate fast enough to have a reasonable spatial separation between each pulse. For a 100kHz pulse rate, a 5µm spacing requires a linear translation speed of 0.5m/s or 50cm/s. To reach 200nm spacing at the peak frequency of 40MHz, a 20m/s translation speed is needed. To demonstrate this spacing, a high speed translation setup was built, shown in Figure 8. In the original project schedule, a rotational stage was planned, however this was changed to a linear stage for two reasons. First, was the difficulty in obtaining a rotational motor suitable for integration inside the SEM sample chamber at a reasonable price. While multiple motor options were identified, they typically required either a new feedthrough in the SEM system or had an unreasonable price

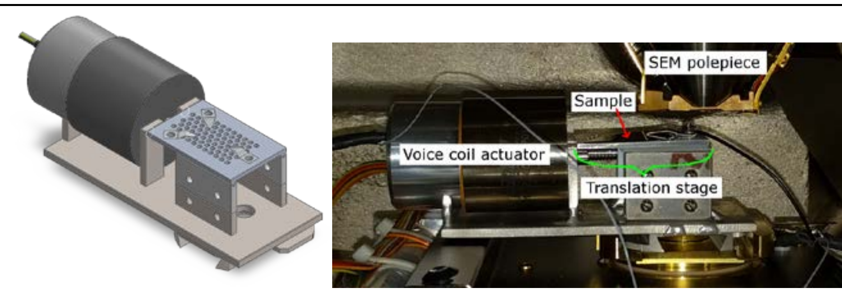

Figure 8 The high speed translation stage. The left figure is a 3D model and the right figure is a photograph of the system in the SEM sample chamber.

and delivery schedule for a single motor. The second reason was the ability to easily mount substrates in a variety of sizes and shapes. A rotational system requires a round sample and precise mounting to eliminate vibration.

In the left image of Figure 8 the large cylinder at the back is the voice coil actuator which moves the perforated stage at high speed. It was important to minimize the mass of the sample in order to achieve such high velocities, which is why the translation stage is perforated. The image on the right shows the setup when mounted inside the vacuum chamber of the SEM. The sample stage is capable of a peak velocity of 0.5m/s and was typically operated at 0.25m/s.

Figure 9 shows a composite image of a line of patterned dots written using photoemission electrons generated at 50kHz.

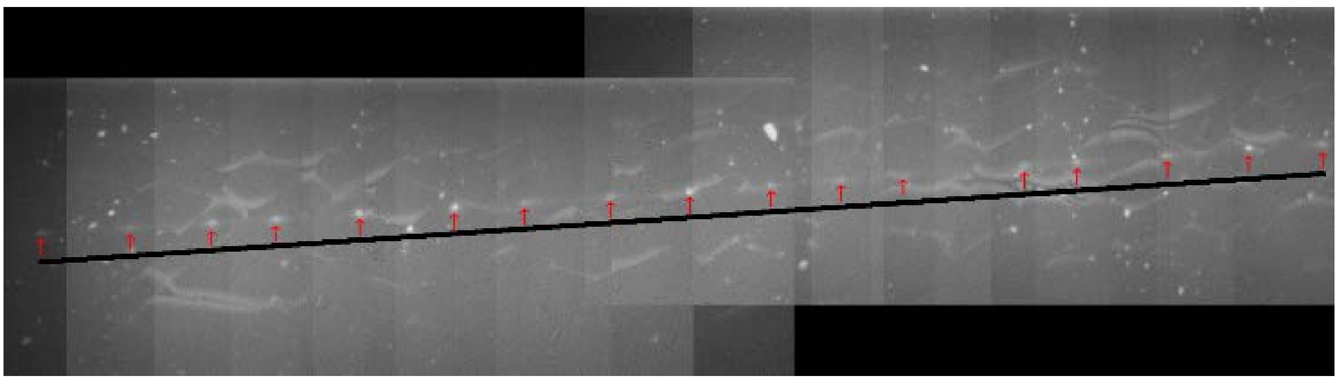

Figure 9 Composite image of discrete point patterning. The red arrows highlight the individual exposed spots

Data storage analysis

The advantage of OSL for data storage is the increased data storage capacity, and while the primary goal of this program was to demonstrate the ability to generate OSL spots at high speed, we did work on improving the estimates of storage capacity (see Figure 10). Based on a saturation of approximately 10^5 electrons, the theoretical maximum level of gray scales would be between 50,000 and 100,000 or 1-2 electrons per level. This correlates to a bit depth of 16 bits/spot. A more realistic maximum is 12 bits, or 4096 gray scales, with 20 electrons/level, though this would require significant advances in both electron control and sensing.

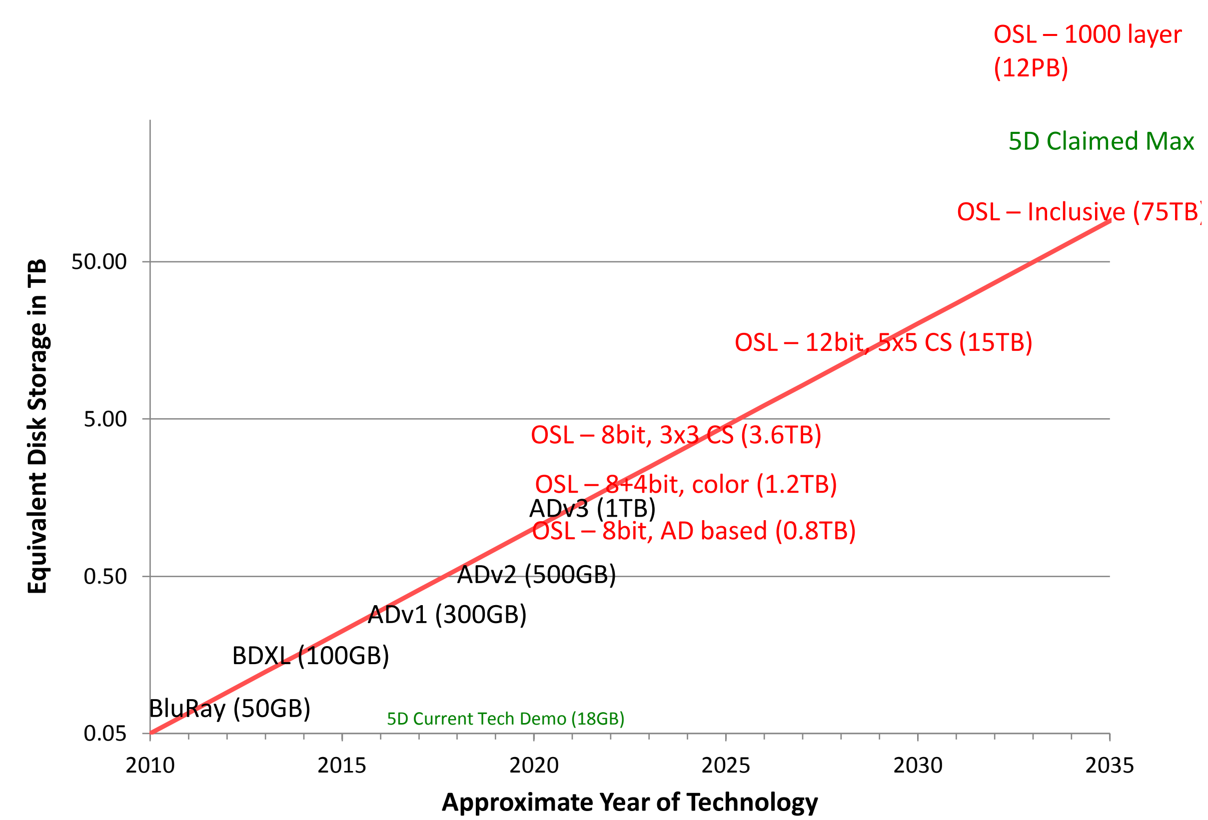

Figure 10 Storage capacity roadmap for OSL products showing BluRay, Archive Disc (AD), and OSL.

Data storage – Compressive Sensing

In order to improve the storage density, we developed a new read-back approach based on a technique commonly called compressive sensing. In the OSL process, the write limits are set by the electron beam, which can be focused to a much smaller spot, potentially down to 10-20nm in diameter. This would enable both a 10x reduction in track pitch and a 10x reduction in linear pitch, increasing the areal density by 100x. To realize this, compressive sensing technology could be used to extract out a signal from the multiple OSL pixels illuminated by a standard 405nm laser. This is a more powerful variation of the technology currently used in hard drives to distinguish the targeted bit from the surrounding field (see Figure 11).

Compressive sensing takes advantage of the temporal variations in a signal to reconstruct the original image. An extreme case of this are what are called single pixel cameras, were a single large photodetector and an array of randomly varied micromirrors (to provide temporal variation) can be used to extract out the original image. In the case of OSL with a spinning disc, the temporal variation is already built into the system. It is only very recently that computer systems and algorithms have become fast enough to do this analysis in near real time. With a 5-10 year time horizon, as well as the much simpler image that is being analyzed, real time data processing will be easily achieved for this technology.

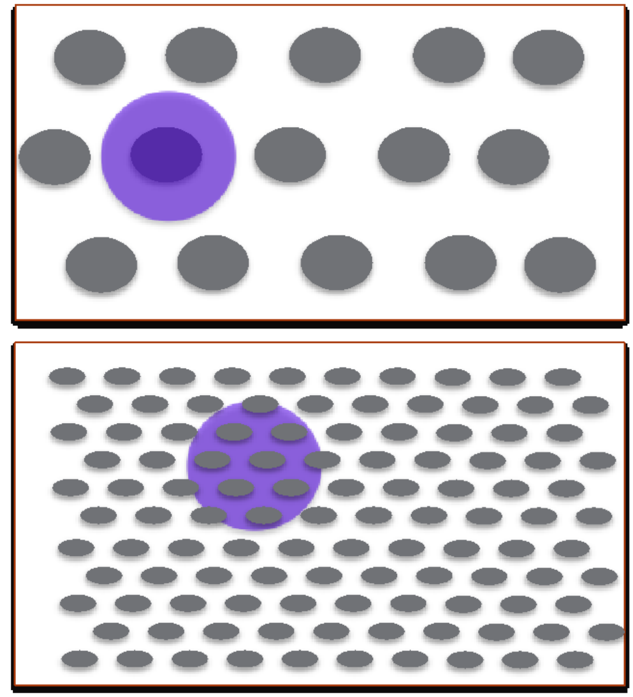

Figure 11 Diagram of sensing laser spot (purple circle) and how it overlaps with data spots in BluRay (top) and OSL (bottom)

Summary

The goal of this program was to evaluate if optically stimulated luminescence spots could be generated with an electron density and pulse length that would scale to data storage. We successfully built a photoemission based electron gun capable of generating 10ps pulses of up to 10^6 electrons and used that to write lines and discrete spots on LiF without damaging the substrate.

We also investigated multiple paths to further increase the storage density. First, we used variable accelerating voltages to vary the relative intensities of the red and green emission bands. This enables storing of additional information in a single spot by independently varying more than one emission line. Second, we developed a new concept for improving the areal density by using oversampling and compressive sensing techniques to enable reading of data pixels smaller than the excitation laser spot diameter. This enables exploiting the ability of the writing e-beam to be focused to a much smaller spot than the read back excitation laser. IP has been filed on both of these concepts as part of the program.

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Optically Stimulated Luminescence Data Storage

David W. Gotthold ◆ **We are developing a new approach for data storage using optically stimulated luminescence (OSL) that can be scaled to high data densities with excellent data lifetime and integrity.**◆

To meet the ever-increasing data storage need for cloud computing, scientists and engineers are constantly pushing the limit of data storage density. Hard disk drive (HDD) has been the dominant computing data storage device for the past 50 years, with over 800 million units sold in 2012. Until recently, HDD density increased by 100% every year, but advancing technology has decreased this rate to 25%. Solid-state hard drive (flash) is the storage method based ferroelectricity, where direct wiring enables immediate access to the bit. However, flash suffers memory wear and thus can be used only as short-term storage. Although CD, DVD, and Blu-ray are popular optical data storage methods, they have a significantly lower density (under 10% of HDD). Still another recent advanced storage method is holography, a promising technology because of its large density and fast data access rate, reading a page each time instead of a bit (like flash). However, sensitivity to vibration and extremely low speed have prevented this technology from succeeding.

We propose to develop a new data approach that is capable of making a paradigm change in storage technology. OSL has been in use for many years as a radiation dosimetry technology. OSL media has an extraordinary linear optical response (over eight orders of magnitude for LiF) that will enable ternary (higher-level) encoding of data in a single bit, thus storing more data in each defined location. The objective of this project is to prove the concept of OSL data storage by demonstrating multi-values data encoding. This information is a primary concern for potential development partners and will be key in identifying the potential market opportunities for this new technology.

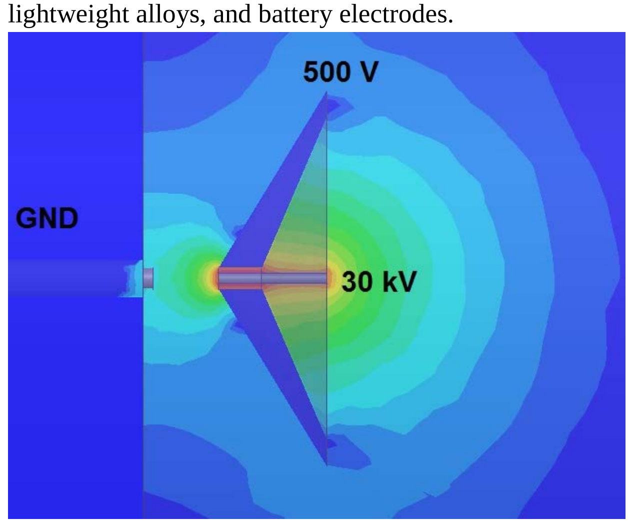
Based on work done in previous years, the lack of knowledge about the OSL process at very high power levels as identified as the critical technology gap. To address this issue, a new project was started to develop a very short pulse electron source capable of producing the ionizing radiation at and energy density and pulse length necessary to evaluate the dynamic formation of the F-centers responsible for the OSL signal.

To develop the ability to measure the formation of OSL centers, a high energy pulse source of ionizing radiation needed to be developed. After evaluating a range of options including pulse x-ray, gamma and electron beams. It was determined that a variable intensity electron beam would be the easiest to generate and control, so development work this year has been focused on building a picosecond pulsed electron beam source with variable intensity.

In order to achieve the targeted short time, high intensity pulses, a laser pumped photocathode was determined to be the best approach and the design and build of this system has been the focus of this year's project. Based on prior work developing a dynamic TEM, a laser driven photocathode system is being developed that will be able to generate electron pulses with up to 10^5 electrons/pulse in as little as 10ps. This system will be integrated with an existing JEOL 5900 scanning electron microscope to enable focusing of the electron pulse and control of the energy density. The drive laser is a Fianium HYLASE with a frequency quadrupling crystal system in order to provide pulse of 266nm that will be directed into the SEM column and on to a tantalum photocathode. The field energies around the photocathode and along the SEM column have been modeled to understand the pulse energy propogation through the system.

In addition, F-centers are strongly influenced by crystal defects and grain boundaries. By growing the LiF films under different process parameters we can attempt to quantify this relationship and generate predictive tools.

In the next fiscal year, the build of the system will be completed and LiF samples will be exposed at a range of pulse energies and speeds in order to probe the F-center formation process. This will enable a more complete understanding of the nature of the OSL effect, the formation of the F-centers, and the limitations of dose rate for this material. We will also have a high speed pulsed electron source coupled with an SEM that is expected to be useful for probing a range of ultra-fast processes in critical areas such as catalysis, structure evolution in lightweight alloys, and battery electrodes.



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