

# Materials for an Updatable Holographic 3D Display

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**Abstract**—Holography is a powerful technique for providing high-resolution, realistic three-dimensional (3D) images without the need for special eyewear. A material that takes full advantage of the potential of holography, including updatability, has not existed. Here, the first updatable holographic 3D display based on a photorefractive polymer is summarized. The performance characteristics of these materials are measured, and how they relate to the development of additional display enhancements such as pulsed writing, white light viewing, and large viewing angle, are discussed.

**Index Terms**—Charge carrier processes, holography, photorefractive materials, three-dimensional (3D) displays.

## I. INTRODUCTION

THREE-DIMENSIONAL (3D) perception is fundamental to the human interaction with the physical world. Our brain is accustomed to viewing and processing information, such as depth and parallax, to accurately create a mental image of the real world. It is unfortunate, therefore, that much of our information about 3D objects is presented to us on fundamentally two-dimensional (2-D) displays.

There are many applications where 3D displays would yield a significant advantage in terms of understanding, including military command-and-control, industrial design, data mining, flight simulators, medical imaging, advertisements, and television and movies, to name a few. Even though considerable research has been dedicated to the development of 3D imaging [1]–[9], we are still lacking an important interface between computer generated images and human perception.

Historically, 3D imaging relied on stereoscopic rendering that required the viewer to use special eyewear. The drawbacks of this technique are user fatigue and the inability to reproduce all of the information from the original object. The methods by which different perspectives are displayed and viewed have advanced from simple prisms and parallax barrier to the more sophisticated liquid-crystal optical switches.

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Holography surpasses the fundamental physical limitations of all of these techniques. It can reproduce the wavefront that the object scattered, greatly increasing the resolution and providing the sensation that the object is actually in front of the user [10]–[12]. Holograms contain a much larger amount of information, which is beneficial for the viewer, but require a correspondingly sensitive recording.

Materials for static holograms, such as dichromated gelatin, silver halide, and photopolymers, can be made up to practically any size, with full-color, full-parallax images [13]. Unfortunately, they are not refreshable, limiting their usefulness for displays applications. Systems utilizing acoustooptic materials [14], [15], liquid crystals [16], or microelectromechanical systems-based recording media [17] allow dynamic recording, but these approaches do not exhibit persistence, and the tremendous information content of holograms currently limits these to small sizes and low resolution. Photorefractive (PR) crystals are dynamic materials with memory but, with the delicate growth conditions, are unable to be scaled up to sizes larger than a few centimeters.

PR polymers have the potential to fulfill these requirements. They have been extensively studied over the last decade [18]–[24] and have a wide range of applications including optical amplification [25], correlation [26], communication [27], and imaging through scattering media [28]. Large diffraction efficiencies have been demonstrated [19] as well as fast response times [29]. Well-developed polymer chemistry and processing principles can be applied to provide large-area thin-film display devices.

In a PR polymer, a refractive index modulation mimics the interference pattern created by two coherent beams. A space-charge (SC) field is built up via charge generation, transport, and trapping in the dark regions. Nonlinear and/or birefringent molecules orient to the SC field, leading to a macroscopic refractive index change. The orientation can be reversed by detrapping the charges using uniform illumination, erasing the hologram.

A large-area PR polymer with fast recording and long persistency has been discussed elsewhere [30], [31]. This is useful for some of the applications discussed above, but, to realize the full potential of PR polymers, further material developments will be needed. Full-color and video-rate holograms will require a material with improved sensitivity bandwidth and faster response times. Viewing for large audiences in ambient lighting conditions will require changes to the writing geometry, as well. A reflection geometry can deliver large viewing angles and white light viewable holograms in full color. The much reduced grating spacing will also allow faster writing and erasing for video-rate image updates, but necessitates a higher trap density.

In this paper, the performance parameters of the previously reported holographic 3D display polymer in transmission are reported and contrasted with a similar sample with additional

sensitization. The physical mechanisms that will guide further improvements, such as pulsed writing and reflection geometry, are introduced.

## II. EXPERIMENTAL METHODS

### A. Material

The PR polymer composites developed and studied contain a copolymer as the hole-transporting host matrix. The copolymer approach is taken to reduce the phase separation typical in guest-host polymer systems with low  $T_g$ , thus allowing increased loading of functional components such as NLO chromophores. The copolymer consists of a polyacrylate backbone with pendant groups tetraphenyldiaminobiphenyl-type (TPD) [32] and carbaldehyde aniline (CAAN) attached through an alkoxy linker (PATPD-CAAN). A fluorinated dicyanostyrene (FDCST) NLO chromophore was added to provide sufficient refractive index change and charge generation at the wavelength of interest (532 nm). The plasticizer N-ethyl carbazole (ECZ) was also used to reduce the glass-transition temperature to approximately room temperature. In some composites, the fullerene derivative [6], [6]-phenyl C<sub>61</sub> butyric acid methyl ester (PCBM) was present to provide improved sensitization.

Samples labeled A were made by melt processing a composite of PATPD-CAAN/FDCST/ECZ (50/30/20 wt%) between two indium-tin-oxide-coated glass slides, with 105- $\mu$ m spacer beads used to set the thickness. Sample B is similar except PCBM was added, making the composition PATPD-CAAN/FDCST/ECZ/PCBM (49.5/30/20/0.5 wt%). The size of the active area varied from 1 cm  $\times$  1 cm to 4 in  $\times$  4 in, depending on the application. Both samples were able to withstand an applied field of more than 70 V/ $\mu$ m for several hours. The samples with PCBM are meant to improve the efficiency for applications in the reflection geometry, as the order-of-magnitude smaller fringe spacing requires more charge generation and trapping to obtain a useful refractive index change. These samples showed no phase separation in an accelerated aging test at 60 °C for seven days. In the 3D display, sample A showed no degradation or damage for several months over hundreds of write/erase cycles. The absorption coefficient at 532 nm of sample A is 90 cm<sup>-1</sup> and sample B is 140 cm<sup>-1</sup>.

### B. Holographic 3D Display System

The hologram for 3D display purposes in transmission geometry is generated using integral image holography. More details on the writing system, including the optical setup, recording parameters, and holographic pixel (hogel) generation are discussed in previous literature [30], [31]. Briefly, dozens of 2-D perspectives of an object are processed on a computer and then optically multiplexed onto the recording medium in a manner such that when reconstructed, the sensation of depth is created via parallax. This allows a 3D image to be constructed from multiple 2-D images without the real object ever needing to be present. A frequency-doubled Nd:YVO<sub>4</sub> laser at 532 nm is split into two paths, a reference beam and an object beam. The object beam is modulated using a 1280  $\times$  768 Holoeye LC-R 720 spatial light modulator (SLM) with the image of a hogel, constructed from the 2-D perspectives. The reference beam is collimated and made to interfere in the sample with the object beam in a Fourier transform geometry. Given the

geometry of the human vision, horizontal parallax only is used, so the object beam is only focused in this direction, creating hogels that are vertical stripes. The hologram is written by successively recording approximately 120 hogels side by side. The total writing intensity is approximately 100 mW/cm<sup>2</sup>. The writing time is limited by the response time of the sample and the vibration relaxation time of stage used to translate the sample, both of these being about 1 s. The overall writing time is then 3–4 min for a 4-in  $\times$  4-in sample, which is much shorter than the decay time of each hogel [31]. After writing, the sample is translated to a new position and the hologram read using a 650-nm LED lamp. The low coherence length results in negligible speckle, and the low absorption at this wavelength reduces detrapping that causes the hologram to decay. Erasing can be done at any time by illuminating the sample with a homogeneous beam at the absorption band wavelength of 532 nm.

For the desired applications, it is beneficial to have a material that can be written quickly but decays very slowly. In order to facilitate this, a technique called voltage kick-off has been developed [30], [31], in which a large voltage (typically 9 kV) is used during the writing to increase speed, while a decreased voltage (typically 4 kV) is used for reading to increase persistency. From the degenerate four-wave mixing (DFWM) dynamics at 9 kV, it takes about 10 s to reach 50% of its steady-state diffraction efficiency. However, an intermediate peak of approximately 10% is observed at the 1-s mark. If at this time the writing beams are turned off and the voltage reduced to 4 kV, the efficiency continues to increase in the dark. This kick-off technique permits a hogel writing time of 1 s in a material with a longer time constant.

### C. Characterization Setups

The two-beam coupling (TBC) measurements were carried out with a 532-nm DPSS laser, where each p-polarized beam was incident with an intensity of 500 mW/cm<sup>2</sup>. This wavelength is used because it is within the absorption band of both the FDCST and PCBM. The external inter-beam angle was 36° and the external tilt of the sample was 55°. The voltage was ramped up slowly to ensure steady-state conditions as the transmitted intensity of each beam was monitored. The gain coefficient ( $\Gamma$ ) was calculated using [21]

$$\Gamma = \frac{(\cos \alpha_1 \cdot \ln \gamma_1 - \cos \alpha_2 \cdot \ln \gamma_2)}{d} \quad (1)$$

where  $d$  is the thickness of the polymer,  $\alpha$  is the internal angle of the beam with respect to the normal, and  $\gamma$  is the intensity of one beam with the other present normalized to the intensity with the other absent. The polarity of the electric field was set to reduce the beam fanning [33].

Steady-state DFWM measurements in transmission were carried out in the same geometry as for TBC, except the writing beams were s-polarized and a p-polarized reading beam with approximately 100 times less intensity, aligned to be counter-propagating with one of the writing beams, was used to probe the grating.

To modify the TBC and DFWM setups for reflection geometry, the sample was simply rotated so the plane of device was along the bisector of the writing beams (with an offset of a

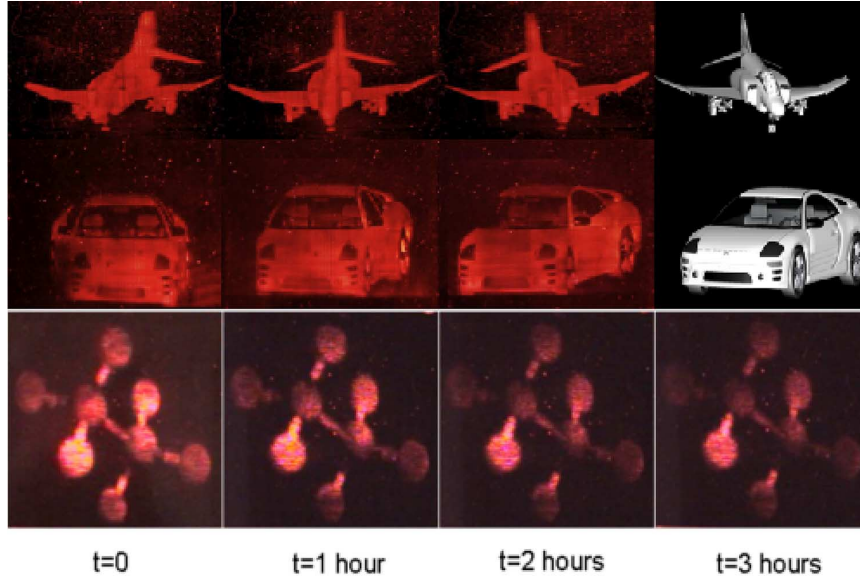


Fig. 1. Digital camera images for the updatable holographic display setup in transmission. The top shows an F-4 Phantom jet, and the bottom shows a Mitsubishi Eclipse. The camera position for each image is rotated to demonstrate occlusion and parallax. The bottom row shows the change in image quality over 3 h of continuous reading.

couple of degrees to separate out the portion of the reading beam directly reflected off the glass).

The formation and decay rates of the SC field depend on many parameters, including photogeneration efficiency, mobility, recombination rate, as well as trap depth, density, and rate [34], [35]. Dark and photoconductivity measurements are useful in probing these processes. A voltage was applied to the sample and this dark current measured until steady-state was reached (typically 10–20 min). Then the sample was illuminated with  $1 \text{ W/cm}^2$  of 532-nm light and the current recorded for another 10 min. For materials where the spot size is different than the electrode area, the dark and photoconductivities ( $\sigma$ ) can be calculated using

$$\sigma_{\text{dark}} = \frac{i_{\text{dark}}}{(EA_{\text{elec}})} \quad (2)$$

$$\sigma_{\text{photo}} = \frac{i_{\text{tot}}}{EA_{\text{beam}}} - \sigma_{\text{dark}} \left( \frac{A_{\text{elec}}}{A_{\text{beam}}} - 1 \right) \quad (3)$$

where  $i_{\text{dark}}$  is the steady-state current without illumination,  $i_{\text{tot}}$  is the same with illumination,  $A_{\text{elec}}$  is the overlap area of the electrodes,  $A_{\text{beam}}$  is the area of the beam, and  $E$  is the applied electric field.

### III. RESULTS AND DISCUSSION

#### A. 3D Holographic Images

Sample A was used to create a large-area PR display. Holograms were recorded in a 4-in  $\times$  4-in active area device, some images of which can be seen in Fig. 1. The camera position for each image was rotated around the sample to demonstrate the occlusion and parallax indicative of the 3D effect. The top row images are from a model of an F-4 Phantom fighter jet, and the bottom row is from a model of a Mitsubishi Eclipse. Note that these 2-D snapshots are only a modest reproduction of the actual experience of viewing the 3D hologram. A modified version of the voltage kick-off technique was used for display holograms. Here, 9 kV was applied to the whole polymer during recording

and was reduced to 4 kV only after this 3-min process. Thus, the first hogels suffer a reduced diffraction efficiency due to the faster decay, however this does not seem to introduce an observable brightness variation across the width of the image. With this process, the image could be continuously viewed for hours or erased within 30 s. Sample B is not used in the transmission display because the increased absorption of PCBM samples reduces the brightness of the reconstructed image. The kick-off technique also does not work with this sample, as discussed below.

#### B. Material Parameters

The quantitative performance of the different samples was measured in order to gain a better understanding of the physical processes involved and to guide developments. Fig. 2 shows the results of TBC measurements in transmission and reflection. The most interesting feature is the increased magnitude and the opposite direction of the gain for sample A compared with B. The sign of gain can reverse if the phase shift between the SC field and interference pattern shifts by  $180^\circ$ , which can happen if either the sign of the charge carrier or the polarity of electric field in the polymer reverses. Either charge buildup at the polymer/electrode interface or local screening could reduce the field in the sample, but would never cause it to reverse on a large scale [36]. This suggests the possibility that the sign of the majority charge carrier in sample A is opposite to that of B.

When ionized, PCBM can act as a hole trap [37], so presumably the number of trapped holes in sample B is larger than in sample A. If increased hole trapping leads to positive gain, then the negative gain in sample A is perhaps due to the electrons being the majority charge carrier. The decreased magnitude with additional hole trapping also suggests a competing grating effect, though further studies are necessary to confirm this.

The change in sign of the gain for both samples in reflection relative to transmission is consistent with the theoretical geometrical factor for an index modulation due to birefringence [38], though the actual sign depends also on the nature of the SC

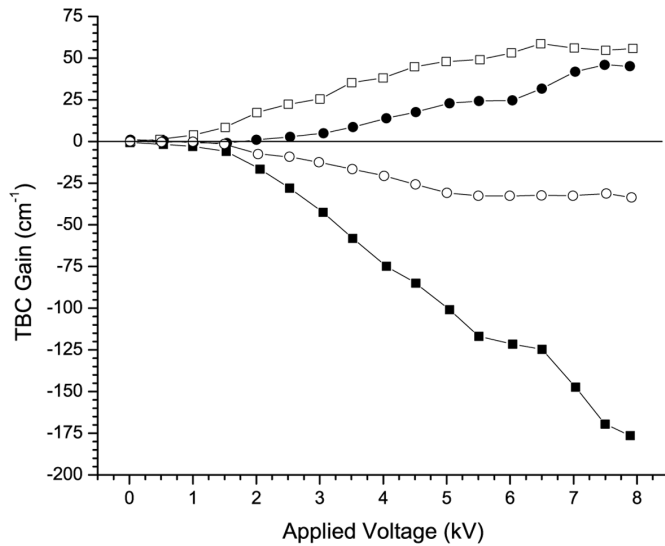


Fig. 2. Steady-state TBC gain at 532 nm. Closed symbols are in transmission, and open symbols are in reflection. The direction for sample A (squares) is opposite that of sample B (circles).

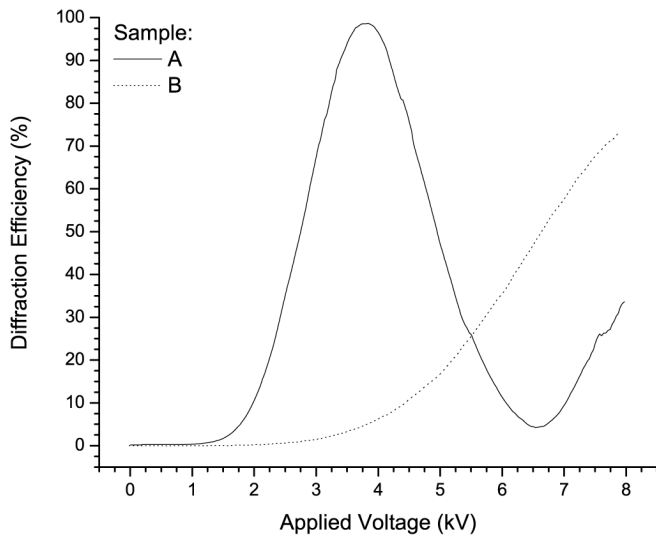


Fig. 3. Diffraction efficiency from DFWM in transmission geometry. Sample A shows maximum efficiency at 3.8 kV, while the addition of PCBM increases the over-modulation voltage.

field. The smaller magnitude is due to a reduction in the grating spacing relative to the trap density.

DFWM was also performed, as the efficiency does not depend on the phase of the SC field and it is a more useful quantity to know for display applications. Fig. 3 shows the steady-state curves, indicating sample A is over-modulated at 3.8 kV. It is this behavior that allows the voltage kick-off to be effective, since decreasing the voltage for reading actually increases the efficiency and decay time. The increased over-modulation voltage for B could be caused by  $C_{60}$ , which has been shown to inhibit chromophore orientation [39], [40]. However, this effect would not also explain the reversal of gain, so is only partially responsible. The large over-modulation voltage prohibits the use of a kick-off technique for sample B in a transmission display.

Conductivity is one of two major factors contributing to the formation and decay of the grating (the other being chromophore orientation). The results of these measurements are

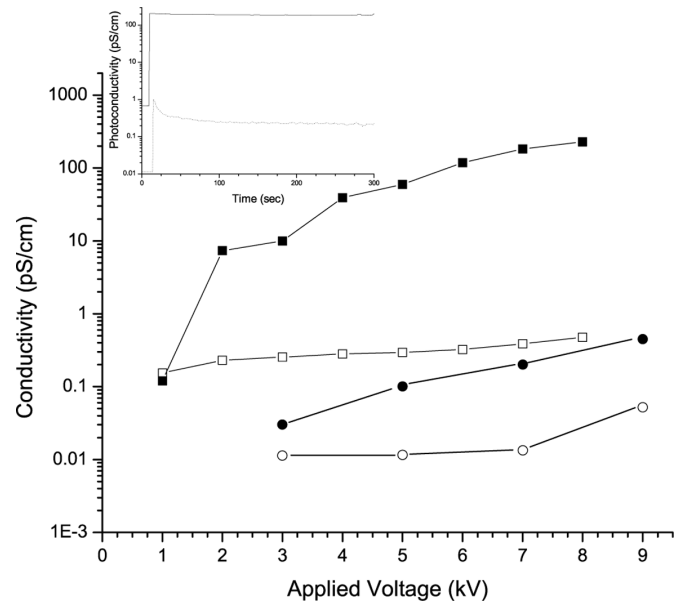


Fig. 4. Dark (open symbols) and photoconductivity (filled symbols) at 532 nm for samples A (squares) and B (circles). PCBM greatly reduces the conductivity, more significantly for the photoconductivity than the dark. Inset: Photo-transients for A (solid line) and B (dotted) show the degradation over time due to deep traps.

shown in Fig. 4. The PCBM decreases the dark conductivity by about an order of magnitude. This reduction is not beneficial for a video rate hologram, but is necessary for persistency in reflection to avoid fast image decay from the reduced SC field.

PCBM reduces the photoconductivity by much more than in the dark, which is caused in part by hole trapping anions. This happens despite the increased absorption, because the sensitizer increases both charge generation and trapping [41], [42]. The large conductivity in A is useful for video-rate applications using a pulsed laser and will translate into a faster writing and erasure rate. This is necessary to avoid overlapping images. However, the increased sensitizer density in B is necessary for pulsed writing, because as many charges as possible must be generated during the brief time the sample is illuminated. For this application, a balance will need to be sought between charge generation and PR speed.

This reduction in conductivity due to traps is supported by the transient photoconductivity (inset in Fig. 4). Samples with a sufficiently high density of deep traps will show reduction of the photocurrent over time [34]. Indeed, sample A decays 25% and sample B by 80% over about 4 min. With more traps, charge transport into dark regions necessary to fully establish the SC field takes longer, and in display holograms, sample B does exhibit an increased response time. It is expected that in reflection the smaller fringe spacing will counteract the reduced photocurrent leading to a faster recording time than in transmission.

The steady-state diffraction efficiency in reflection is shown in Fig. 5. The efficiency was observed to increase as the angle of the reading beam was offset relative to counter-propagation. This is because, as the chromophores align with the applied dc field, the average index modulation changes, which alters the Bragg matching and requires an offset angle to restore [43]. The peak efficiency is about 16% at 6° offset and 3.6 kV for sample A and about 40% at 3° and 9 kV for sample B. In [43] and [44], over 60% was obtained with a PVK-based composite,

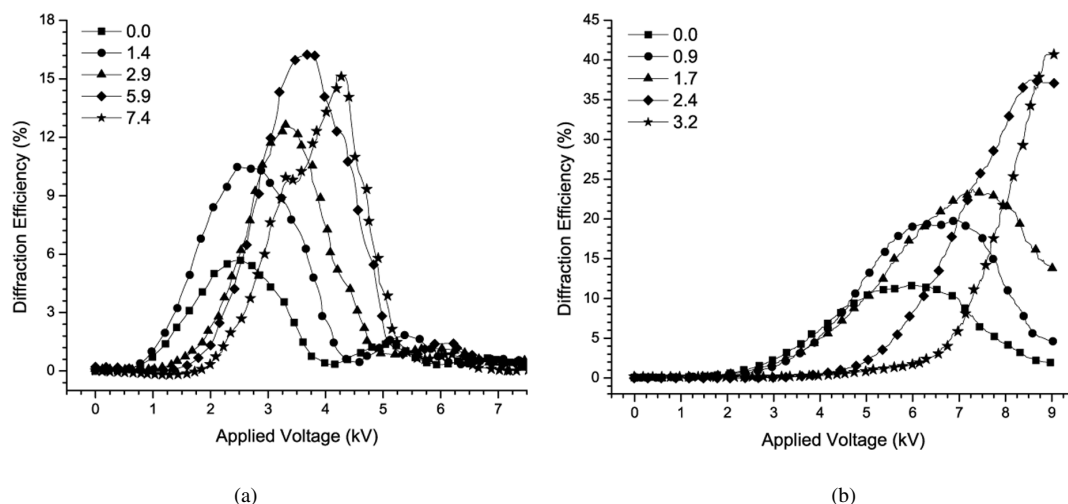


Fig. 5. Steady-state diffraction efficiency in reflection geometry for (a) sample A and (b) sample B. The numbers in the legend show the degree offset of the reading beam from counter-propagating with the writing beam. PCBM increases the peak efficiency by more than a factor of 2.

though this polymer does not lend itself well to making large area devices and the hole mobility is less than in the PATPD copolymer used here. The fact that the peak efficiency in B occurs at a higher voltage and smaller offset angle is consistent with the reduced birefringence observed in transmission.

These results are in contrast to transmission, where the PCBM did not increase the efficiency. One possible explanation is that the charges do not need to travel as far before they are trapped in a dark region, and thus avoid recombination. This effect is greater for sample B, where the PCBM can trap holes (when ionized) or electrons (when neutral).

This also means that the charges do not have to travel far to recombine in the dark. Indeed, in a reflection display setup, the fast decay of the hologram limited the usefulness of the sample for large area displays. Trap densities commensurate with the order of magnitude smaller grating spacing are required, not only to improve the efficiency, but also to provide the SC field enough resolution to accurately capture the spatial variation of the inhomogeneous interference pattern of the hologram, and PCBM shows significant improvement. New chromophores and sensitizers may provide sensitivity inline with those of chemical emulsions, which is necessary for wider applicability. Other novel techniques will likely have to be explored, such as layered photoconductive polymers [45] and nanoparticle sensitization [46], [47], which have already shown promise.

#### IV. CONCLUSION

The performance of the previously reported PR polymer device used in an updatable holographic 3D display has been reported. The device shows high efficiency at 4 kV and a large TBC gain magnitude. However, the direction of the gain reverses when PCBM is added, which also reduces the magnitude, and is perhaps due to competing gratings of different sign. The increased sensitizer density is beneficial for video rate displays, however the reduced conductivity is beneficial for the reflection geometry, where a twofold increase in the efficiency is observed. Further increases to trap density and sensitivity are needed to develop displays at video rates and in reflection.

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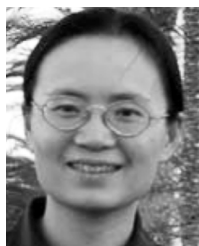
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