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LETTERS

An updatable holographic three-dimensional display

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Holographic three-dimensional (3D) displays[1,2] provide realistic images without the need for special eyewear, making them valuable tools for applications that require situational awareness, such as medical, industrial and military imaging. Currently commercially available holographic 3D displays[3] use photopolymers that lack image-updating capability, resulting in restricted use and high cost. Photorefractive polymers[4–9] are dynamic holographic recording materials that allow updating of images and have a wide range of applications, including optical correlation[10], imaging through scattering media[11] and optical communication[12,13]. To be suitable for 3D displays, photorefractive polymers need to have nearly 100% diffraction efficiency, fast writing time, hours of image persistence, rapid erasure, and large area—a combination of properties that has not been shown before. Here, we report an updatable holographic 3D display based on photorefractive polymers with such properties, capable of recording and displaying new images every few minutes. This is the largest photorefractive 3D display to date (4 × 4 inches in size); it can be recorded within a few minutes, viewed for several hours without the need for refreshing, and can be completely erased and updated with new images when desired.

A considerable amount of research has been dedicated to the development of 3D imaging[14–22], because two-dimensional (2D) images give only limited information about an object or a scene owing to their lack of parallax and depth[17]. 3D imaging techniques that rely on special eyewear have unwanted side-effects such as eye fatigue and motion sickness. Holographic 3D displays do not incur these problems because they are viewable with the naked eye (autostereoscopic) and simulate natural human vision. Humans are naturally attracted to holograms, which is why holography has found wide applications in advertisement and entertainment. Current static holographic displays[3] are capable of displaying terabytes of data, and come in practically any size with full colour, full parallax and depth. Previously, dynamic 3D holographic displays based on acousto-optic[23], liquid-crystal displays[24] and microelectromechanical-systems-based recording media[25] have been demonstrated. Unfortunately, these devices do not have memory, and thus do not exhibit persistence of recorded images. The lack of persistence results in the requirement of update rates faster than 30 Hz to avoid image flicker. 3D images exhibit very high information content, so this high refresh rate requirement currently limits real-time holographic displays to small sizes. Photorefractive inorganic crystals are dynamic holographic storage materials that have memory[26], but scaling them to the large sizes needed for 3D displays is challenging. Photothermoplastics provide reversible recording by using surface relief gratings, but they suffer from limited diffraction efficiency and usually require a post-recording developing process. To extend dynamic holographic 3D displays towards practical applications, alternative materials with high efficiency, reversible recording capabilities, memory and significantly larger sizes are needed.

Photorefractive polymers are dynamic holographic recording materials capable of fulfilling these requirements[4–13]. In photorefractive polymers, a 3D refractive index pattern—a phase hologram—replicates the non-uniform interference pattern formed by two incident coherent light fields. This effect is based on the build-up of an internal space-charge field due to selective transport and trapping of the photo-generated charges, and an electric-field-induced index change via the photorefractive effect (ref. 5 and references therein and ref. 4). This process—in contrast to the photochemical processes involved in photopolymer holograms—is fully reversible, because trapped charges can be de-trapped by uniform illumination. The erasibility of the photorefractive gratings allows for refreshing/updating of the holograms. In a typical photorefractive material the holograms are viewed with the help of a reading beam, as long as the initial writing (recording) beams are present. When the writing beams are turned off, the photorefractive hologram decays at a rate determined by the material properties and ambient thermal processes. Photorefractive polymers that have fast recording usually have high decay rates. For updatable 3D displays, however, a material with rapid recording and slow decay (long persistence) is required. A figure-of-merit (FOM) for the design of recording media for spatially multiplexed 3D displays can be the ratio of storage time to the total recording time during which the writing beams are turned on, per holographic element (hogel). In most photorefractive materials the FOM is close to 1, which is far smaller than the FOM > 1,000 required for use in updatable 3D displays with large enough size and resolution.

We have developed photorefractive polymer composites that combine these properties, suitable for use in updatable 3D displays. The composite consists of a copolymer with a hole-transporting moiety and a carbaldehyde aniline group (CAAN) attached through an alkoxy linker. The copolymer approach is adopted to minimize the phase separation between the functional components usually seen in homopolymer photorefractive composites. A copolymer with a polyacrylic backbone was used to attach pendant groups, tetraphenyldiaminobiphenyl-type (TPD) and CAAN in the ratio 10:1 by the synthetic modification of the polyacrylate TPD (PATPD) polymer[12]. The host PATPD-CAAN copolymer provides the optical absorption and charge generation/transport at the writing wavelength (532 nm). A plasticizer, 9-ethyl carbazole (ECZ) was added to the composite. The non-linear optical (NLO) properties were achieved by adding a fluorinated dicyanostyrene (FDCST) chromophore. The composite PATPD-CAAN:FDCST:ECZ (50:30:20 wt%) was formed into thin-film devices by melting it between two indium-tin-oxide-coated glass electrodes with a thickness of 100 μm set by glass spacer beads. This composite showed no phase separation in an accelerated ageing test

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at 60 °C for 7 days. The photorefractive thin-film devices show near 90% diffraction efficiency at an applied voltage of 4 kV in steady-state four-wave mixing measurements (Fig. 1a). The two-beam coupling gain coefficient Γ for these devices at 5 kV is around 200 cm^{-1} (inset to Fig. 1a). Figure 1b shows a 4×4-inch active area thin-film device made from this composite. The device shows no degradation or dielectric breakdown for extended periods of usage (several months) in display recording experiments, with hundreds of write/erase cycles every month at high applied voltages (9 kV) and optical intensities around 100 mW cm^{-2}.

The holograms recorded in the photorefractive polymer thin-film devices can persist for up to 3 h in the dark (without writing beams) at an applied voltage of 4 kV, while continuously being probed with a red (633 nm) laser beam. We have developed a new technique to improve the writing speed of organic photorefractive materials that is based on manipulation of the applied voltage, which we call the 'voltage kick-off technique'. In conventional holographic recording of photorefractive polymers, a constant external voltage is applied across the polymer to dynamically pole the NLO chromophores (ref. 5 and references therein and ref. 4). In the kick-off approach, we apply an increased voltage (9 kV) across the polymer to increase the writing speed during hologram recording, and then reduce the voltage to its optimum value of 4 kV after recording is complete. The temporarily increased voltage facilitates efficient separation of electron–hole pairs, and improves the drift characteristics, forcing the charges to travel faster, and increases the orientational order parameter and speed of the NLO chromophores. The reduction of the voltage to its optimum value after recording ensures hologram persistency. The overall benefit of the voltage kick-off is the reduction of the writing time per hogel to less than a second, by fine-tuning of the applied voltage. We have achieved a diffraction efficiency of 55% using a total writing time (the time during which the writing beams are turned on) of only 0.5 s at 1 W cm^{-2} irradiance with this technique (Fig. 2), much higher than the 1.5% efficiency achieved with writing for 0.5 s at 4 kV without using voltage kick-off. With the several hours of persistence time of holograms in this composite, this corresponds to a FOM $>$ 10,000 without the need for thermal[27] or other fixing methods, which is a significant step in the development of photorefractive polymers for holographic storage and display applications.

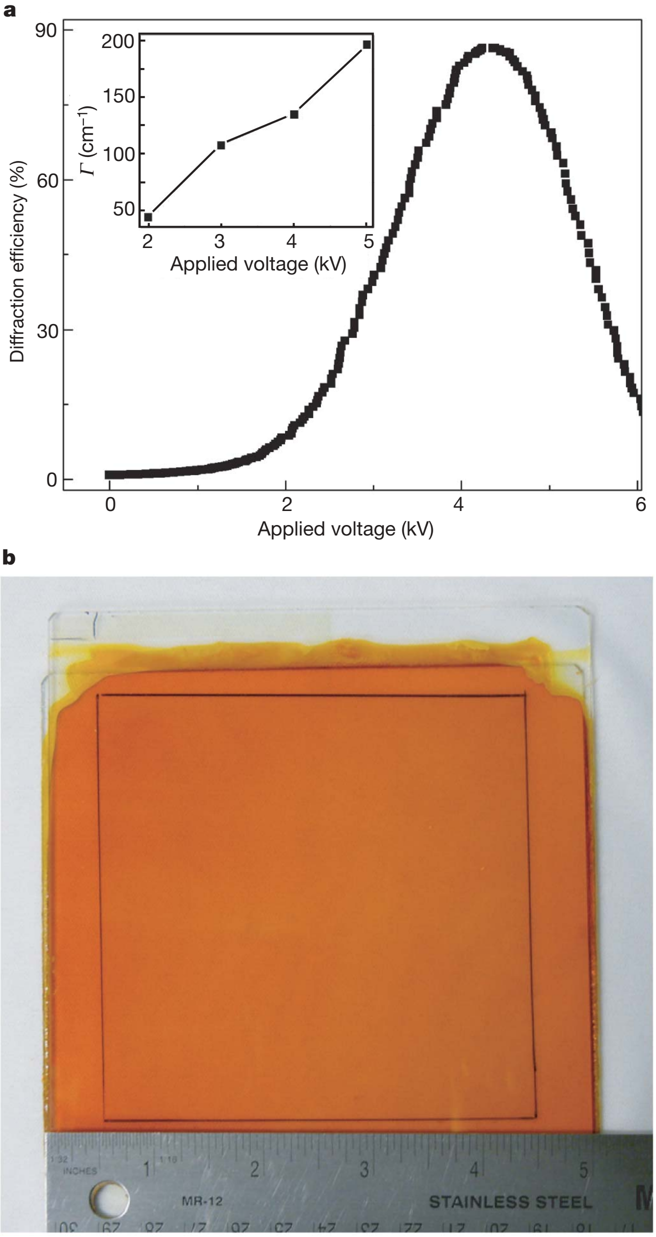


Figure 1 | Diffraction efficiency and the photorefractive polymer thin-film device. a, The steady-state diffraction efficiency of the 100-μm-thick polymer composite is measured using writing beams at 532 nm with a total irradiation of 1 W cm^{-2} and a reading beam at 633 nm. Inset, two-beam coupling gain versus applied voltage. **b**, Picture of a 4×4-inch photorefractive polymer thin-film device.

Holographic stereography—a technique based on optical multiplexing of a limited number of viewpoints (perspectives) onto different parts of a recording medium—is a widely used technique for producing 3D imagery and displays[14,28,29]. We have built a fully automated, computer-controlled 3D holographic printer/display based on holographic stereography using the photorefractive polymer devices described above. The 3D display is recorded onto the entire photorefractive polymer device with an active area of 4×4 inches (Fig. 3). First, 2D perspective views of the object of interest are generated from a 3D computer model. The 2D perspectives can also be generated using methods like magnetic resonance imaging,

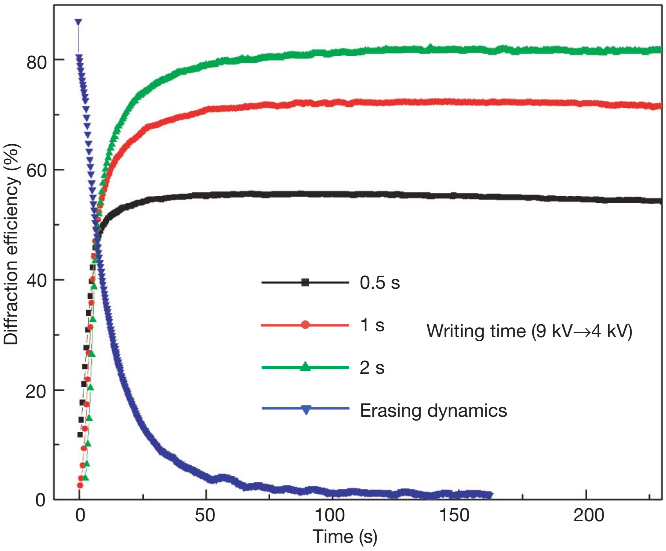


Figure 2 | Recording dynamics of the photorefractive polymer and voltage kick-off. The writing beams at 532 nm with 1 W cm^{-2} irradiance are turned on at an applied voltage of 9 kV for a few seconds (writing time), and then turned off. The voltage is then reduced to 4 kV to ensure hologram persistence and high diffraction efficiency. The maximum diffraction efficiency achieved increases with increasing writing time. A modified version of this technique was used for the 3D display recording of Fig. 4. Also shown is the erasing dynamics of the photorefractive hologram at 9 kV. The erasing beam is a spatially uniform 532 nm laser beam at an irradiance of 1 W cm^{-2}.

computer-assisted tomography, confocal microscopy or aerial and satellite imaging. The perspectives are then divided or 'sliced' into multiple 2D image planes. The image planes are re-organized using a computer algorithm into 2D matrices (the hogel data), which are then uploaded to a spatial light modulator (SLM). The SLM that is illuminated with a 532 nm laser beam displays the hogel data in sequence with the translation stages and an electro-optic laser shutter. The laser beam modulated by the SLM (object beam) illuminates the predefined hogel area on the polymer device. A coherent reference beam simultaneously illuminates the same area, which facilitates the recording of the hogel through interference with the object beam and the photorefractive effect. After one hogel is recorded the shutter turns off the laser beams, the polymer device is translated to the next hogel position, and new hogel data are uploaded to the SLM (see Supplementary Video 1 for display operation). The holographic display is viewed using light from an expanded, low-power helium neon (633 nm) laser beam in transmission geometry (Fig. 3).

In many applications, horizontal parallax only (HPO) imaging[29] is an effective approximation to full-parallax imaging, because humans perceive depth using horizontally offset eyes. The use of HPO recording helps in significantly reducing the number of hogels in a 3D display, resulting in shorter total writing times. We have recorded 3D displays (4 × 4 inches in size) with complex and high-quality images (Fig. 4) within a few minutes using HPO imaging (see Supplementary Video 2). The total recording time used per hogel (0.8 × 101 mm in size) varies from 0.5 to 2 s, and the total irradiance (sum of both beams) used is 0.1 W cm^{-2}.

Here, we used a modified version of the voltage kick-off technique, in which a constant high voltage (9 kV) was applied to the entire polymer device during recording of the hogels. Once recording of all of the hogels is completed, which takes around 2.5 min, the voltage was reduced to its optimal value of 4 kV, which ensures long persistence with maximum diffraction efficiency. The first few recorded hogels suffer a small reduction of diffraction efficiency owing to the high applied voltage during recording of the later hogels, but this lower diffraction efficiency does not create a noticeable brightness variation across the display (see Supplementary Videos 1 and 2). For larger displays the variation may be significant, but this can be avoided by the use of patterned electrodes that allow for individual control of the applied voltage for each hogel.

The 3D display exhibits a total horizontal viewing angle of 45° with uniform brightness, and a resolution that is comparable to NTSC (National Television System Committee) television. The Bragg mismatch usually observed in non-degenerate four-wave mixing that results in intensity variations across the horizontal view-zone is minimized by using a vertical reference/reading beam geometry. The images are viewable for up to 3 h directly on the photorefractive thin-film device without the need for intermediate projection tools or magnification between the recorded image and the viewer (Fig. 4b).

The pictures of the holograms in Fig. 4, which were captured using a video camera, are only modest facsimiles of the effect experienced upon direct viewing. This is principally due to the astigmatism introduced by the HPO recording technique and electronic artefacts such as saturation, to which the human visual system is relatively insensitive. The images can be completely erased within minutes by uniform illumination of the display using a 532 nm beam (Fig. 4c), and new images can be recorded when desired. There is no technological limit to the achievable display size, because large thin-film devices can be fabricated and even tiled together. Moreover, the persistence and efficiency of the material make it the leading candidate for future full-parallax displays, which typically require two orders of magnitude more information content than HPO displays. For larger, full-parallax displays a combination of short pulsed recording[30] and thermal fixing[27] can be used, which is a future route for holographic 3D display development.

Image-updating capability can significantly extend the applications of holographic 3D displays and reduce the cost of 3D imaging. We have developed photorefractive polymer devices that combine exceptional properties such as large size, high efficiency, fast recording, image persistency, long lifetime and resistance to optical and electrical damage, satisfying many of the major requirements for use in holographic 3D displays. These advances have allowed us to

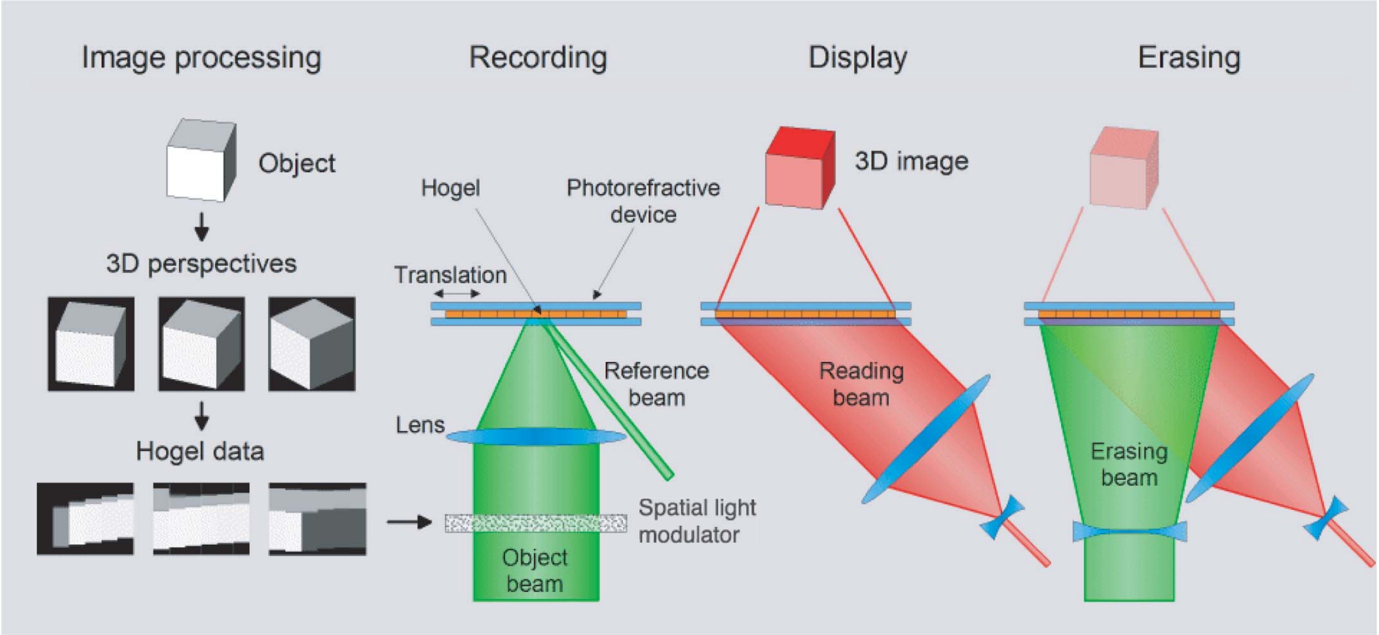
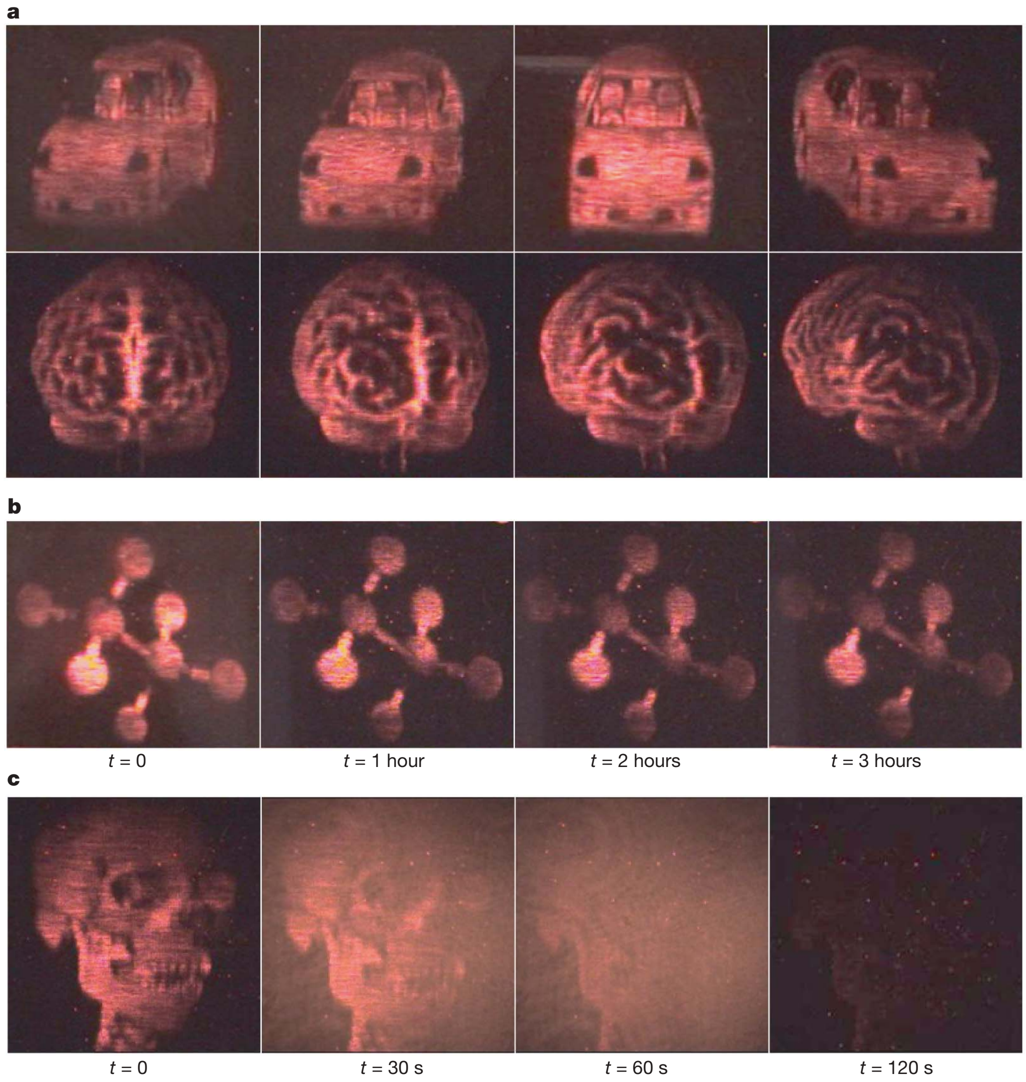


Figure 3 | Image processing, hologram recording and display. The 2D perspective views of the object are generated using a 3D computer model or a video camera moving on tracks around the object. The perspective images are re-organized (hogel data) and uploaded to the SLM. The SLM modulates the object beam, which is focused to the photorefractive polymer and recorded in the Fourier transform geometry. The completed display can be viewed using a reading beam. The result is realistic 3D imagery with parallax and depth. The holograms can be erased by uniform illumination at the writing wavelength.



$t = 0$ $t = 1$ hour $t = 2$ hours $t = 3$ hours

$t = 0$ $t = 30$ s $t = 60$ s $t = 120$ s

Figure 4 | Images from the updatable holographic 3D display. The display uses a single photorefractive thin-film device with an active area of 4 × 4 inches. Here, a modified version of the voltage kick-off was used to avoid using patterned electrodes. A constant voltage (9 kV) was applied across the entire polymer. Once recording of all of the hogels was completed, the voltage was reduced to its optimal value of 4 kV. **a**, A 3D hologram of a sports car was written, displayed, and then erased and a new hologram of a human brain was recorded onto the same area. The images were captured from a distance of 75 cm from different angles to demonstrate the 3D effect using a video camera moving around the display. **b**, The persistence of the hologram, in this case a 3D model of an ethane molecule, is demonstrated by capturing pictures at different times after recording. **c**, Erasure of a 3D image, a human skull, using uniform exposure is demonstrated.

demonstrate the largest updatable photorefractive holographic 3D display so far, scalable to full parallax and colour.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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