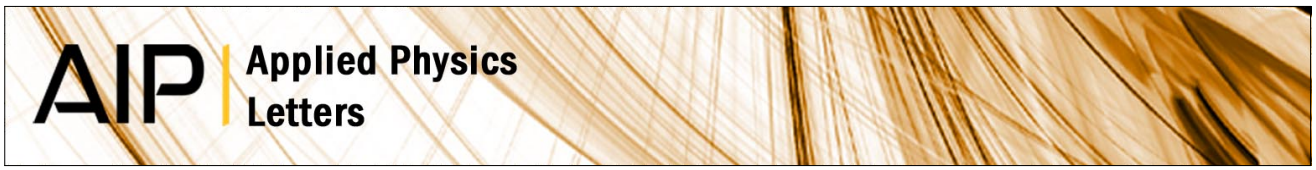

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Photorefractive polymer composite operating at the optical communication wavelength of 1550 nm

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A photorefractive polymer composite sensitized at 1550 nm through direct two-photon absorption has been developed. We show an external diffraction efficiency of 3% in four-wave-mixing experiments and perform holographic reconstruction of distorted images utilizing thin-film devices made of this polymer composite. Amongst other potential applications, the demonstration of accurate, dynamic aberration correction through holography in this all-organic photorefractive device presents an alternative to complex adaptive optics systems currently employed in through-air optical communication links. © *2004 American Institute of Physics*.
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Photorefractive organic composites[1–6] have been widely investigated as recording media for applications such as medical imaging,[7] imaging through scattering media,[8] optically induced focusing-to-defocusing switching,[9] and optical data storage.[10] The inherent compositional flexibility in combination with easy processing and low cost makes polymeric materials a viable alternative to traditionally used inorganic crystals such as $LiNbO_3$. Achieving IR sensitization in photorefractive polymer composites is crucial for imaging and optical communication applications that operate at wavelengths from 800 to beyond 2000 nm. However, this has proven to be a considerable challenge since, in addition to its IR absorption properties, the sensitizer has to be compatible with the polymer matrix with regard to solubility, phase stability, and redox activity with respect to the charge transport material and nonlinear chromophore, which provides the electro-optic functionality.[11,12] Despite recent efforts[7,13,14] in the development of IR sensitive photorefractive polymers, the longest operating wavelength reported so far is 975 nm for an all-organic photorefractive thin film device,[15] far below 1550 nm, the wavelength of choice for optical communications. To address this problem, this letter employs multiphoton sensitization and achieves the desired IR sensitization through direct two-photon absorption in a polymer composite that exhibits negligible linear absorption. This approach can be implemented due to recent developments of organic molecules with large two-photon absorption (TPA) cross sections[16,17] and the evolution of compact, low cost short-pulse lasers. In addition to IR sensitization, the use of TPA provides the advantage that the material is transparent for any cw light beam that reads-out the hologram at the same wavelength.[18] This nondestructive read-out dramatically eases restrictions regarding reading beam intensities and allows for greater film thickness with higher diffraction efficiency. Both features are particularly valuable in imaging applications where high reading intensities and diffraction efficiencies are desired to improve signal-to-noise ratios.

In photorefractive materials, a hologram, a three-dimensional refractive index pattern, is generated by the nonuniform interference pattern formed by two incident coherent beams. This effect is based on the build up of an internal space charge field due to selective transport of the photogenerated charges and a field-induced index change resulting in a phase coding of the incident light distribution.[2] In polymer composites the required photosensitivity, photoconductivity, and electro-optic properties are achieved by combining a number of different functional components. The polymer composite we have developed utilizes a well-optimized chromophore, 4-homopiperidino benzylidinemalonitrile (7-DCST) that exhibits a high figure-of-merit for electro-optic response and a dye (DBM, see Fig. 1) for two-photon sensitization. This represents a different strategy compared to previously reported two-photon photorefractivity in polymers,[18] where the electro-optic dye also served as the sensitizer. Furthermore, an acrylate polymer[19] (PATPD, also shown in Fig.

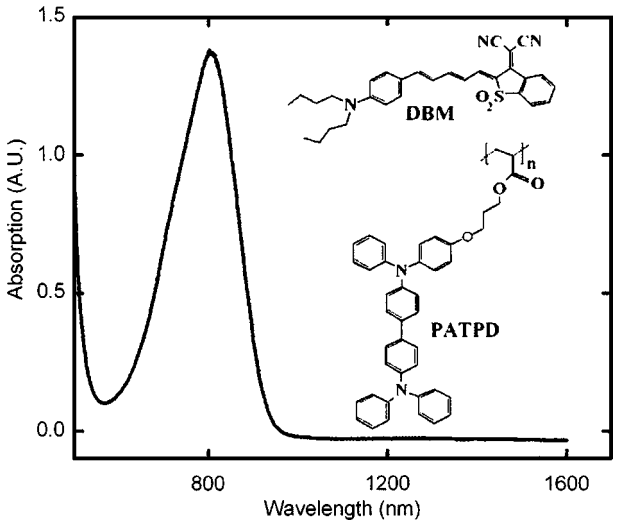

FIG. 1. Linear absorption spectrum of the polymer composite. In addition, the chemical structures of the sensitizer dye (DBM) and the hole transport polymer (PATPD) are shown.

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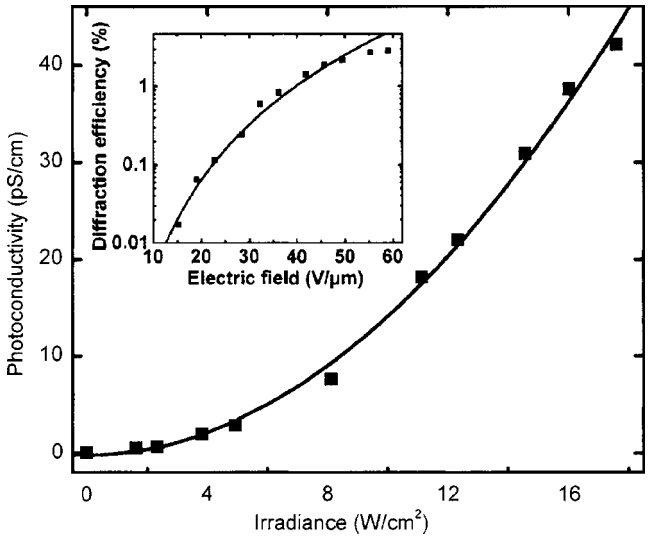

FIG. 2. The photoconductivity measured during irradiation with 130 fs laser pulses at 1550 nm. The photoconductivity is plotted as a function of time averaged irradiance onto a sample that is biased at 57 V/μm. The line is a guide to the eye and is a quadratic function of the average irradiance. Inset: Electric field dependence of the diffraction efficiency measured in a four-wave mixing geometry. To guide the eye a fourth power dependence is plotted together with the experimental data.

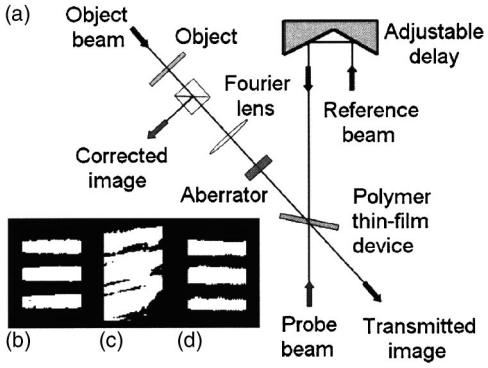

FIG. 3. Correction of an optical image after propagation through an aberrating medium.(a) Holographic imaging setup. (b) Reconstructed image in the absence the aberrating medium. (c) Distorted image transmitted through the aberrator and the polymer thin-film device. (d) Corrected image through readout of the hologram.

1) incorporating a pendant tetraphenyldiaminobiphenyl (TPD, a well-known hole-transport agent) group through an alkoxy side-chain linkage was used for charge transport. Redox characteristics exhibited in cyclic voltammetry indicate that charge generation and transport are feasible through the respective constituents. In order to obtain a large electro-optic response in the material through the molecular rotational nonlinearity, the glass transition temperature (T_g) of the composite was brought close to room temperature by the addition of a plasticizer, *N*-ethylcarbazole (ECZ). The composite with PATPD: 7-DCST: DBM: ECZ ratios at 50: 25: 10: 15 wt % exhibited a broad T_g close to room temperature in differential scanning calorimetry. Uniform films of 105 μm thickness were fabricated by sandwiching this composite between two ITO deposited glass electrodes.[20]

The linear absorption spectrum of the photorefractive polymer composite in the near infrared wavelengths (Fig. 1) is dominated by a feature centered near 800 nm due to optical absorption by the sensitizer, while no significant linear absorption is observed beyond 1100 nm. Valuable information regarding charge generation by nonlinear absorption is provided by the photoconductivity measurements shown in Fig. 2. In this experiment, the sample was irradiated by 1 kHz repetition rate, 130 fs optical pulses with a central wavelength of 1550 nm. When the charge generation is through TPA, the photoconductivity of the polymer composite is given by

$$\sigma(E) = \Phi(E)\frac{e\mu\tau\beta I^2}{2\hbar\omega}. \quad (1)$$

Here $\Phi(E)$ is the photogeneration quantum efficiency, e is the electron charge, μ is the mobility of holes, τ is the carrier lifetime in the excited state, β is the TPA coefficient, I is the irradiance of the incident light, and $\hbar\omega$ is the single photon energy. In Fig. 2, the photoconductivity shows a quadratic dependence on the time-averaged irradiation in agreement with Eq. (1), which is a strong evidence for carrier generation through TPA. The measurements also emphasize almost two orders-of-magnitude contrast between dark ($\sim$0.5 pS/cm) and photoconductivities, which is favorable for the build up of space charge fields in these composites.

To demonstrate IR photorefractivity and study the refractive index gratings in the polymer films, four-wave-mixing experiments were performed in standard tilted-sample geometry.[21] Volume holograms were recorded that resemble the interference pattern of two writing beams generated at 1550 nm in an optical parametric amplifier pumped by a femtosecond Ti-sapphire laser. The inset to Fig. 2 shows the diffraction of 1550 nm (cw) laser light incident on the photorefractive grating written in the present polymer composite as a function of the applied electric field at a time-averaged irradiance (sum of both writing beams) of 9.9 W/cm^2. The data were taken at the steady state of the diffraction efficiency with maximum overlap of the writing pulses. In agreement with Kukhtarev's model applied to orientational photorefractivity in polymers in the space-charge limit,[2] the diffraction efficiency increases with the fourth power of the applied external field. For the highest fields we observed almost 3% internal diffraction efficiency. The grating recording process was fully reversible, i.e., the gratings could be erased by illumination with either one of the writing beams. When using high-power pulsed reading beams, the photorefractive gratings were erased at an increasing rate beyond their intrinsic thermally induced decay. In striking contrast, the application of 1550 nm cw reading beams, with the same average power as the pulsed ones, did not affect the decay of the gratings. The insensitivity of the photorefractive grating decay with respect to the cw reading beam intensity provides a clear signature of nondestructive readout with this polymer composite.

One of the main difficulties in free-space optical communication systems has been to find an effective but simple way to correct the distortions induced by turbulent atmospheric conditions. To demonstrate the capability of our polymer composite as a media for optical correction of image-bearing beams in free-space laser communication, an experiment based on phase-conjugated read-out of the photorefractive hologram at 1550 nm was performed (Fig. 3). Optical phase-conjugation has important potential applications in free-space data links when employed in the retromodulator/conjugator configuration.[22] An image carrying femtosecond laser beam (object beam) passed through an

imaging lens and a phase distorting aberrator. The Fourier transform of the distorted image was formed in the sample and interfered with a collimated reference beam from the same laser source. A cw probe beam counterpropagating to the reference beam read the recorded hologram and a wavefront-reversed replica of the object beam was generated through diffraction by the hologram. The diffracted signal propagated once again through the distorting aberrator and was inversely Fourier transformed. In the plane of a CCD device a reconstructed image was obtained and compared to a simply transmitted image.

Figures 3(b)–3(d) illustrate the accuracy of this dynamic aberration correction scheme. Without an aberrator (distorting medium), the reconstructed image in Fig. 3(b) showed all the features of the original object and our simple thin film device was able to produce an image with almost 97% of the pixels reconstructed correctly. As shown in Fig. 3(c), the insertion of an aberrator strongly disturbed the transmitted image, in this case to a degree where close to 50% of the camera pixels were in the wrong state. However, the reconstructed image [Fig. 3(d)] with the same aberrator in place resembles the original object within 6% of all pixels, a number that almost equals the reconstruction error without any aberrator. In comparison with adaptive optics techniques that require a distortion compensation actuator, a wave front sensor and a control unit as well as associated electronics, the use of a photorefractive polymer hologram for beam cleanup is simple, low cost, and all-optical.

In conclusion, we have extended the wavelength range of organic photorefractive materials to the optical communication window around 1550 nm. At this important wavelength we demonstrated the recording of photorefractive holograms and holographic image correction using an all-polymeric thin-film device. Nondestructive readout is an inherent advantage of sensitization through nonlinear absorption. Applications of our devices in through-air communications are possible and our approach might also find an important place in IR imaging applications. Future research will focus on optimizing both sensitivity and response time of our polymer composites.

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