

Photorefractive Materials for Optical Storage and Display

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Abstract

Real-time data storage and processing using optical techniques have been considered in recent years. Of particular interest are photosensitive electro-optic crystals which permit volume storage in the form of phase holograms, by means of a charge transfer process. A survey of the state of the art of such holographic memories is presented. The physical mechanism responsible for the formation of phase holograms in such crystals is discussed. Attention is focused on various aspects of materials characterization, development and utilization. Experimental reversible holographic read-write memory systems with fast random access and high storage capacity employing this new class of photosensitive materials have already been demonstrated.

Introduction

The increasing use of electronic computers in recent years has considerably increased the demands on computer memories and memory technologies. Current and projected memory requirements define the goals for such a technology: reliable, flexible, random access, read-write memories with high storage densities, fast access times and high data transfer rates, all at an acceptable cost. Several new technologies are emerging to satisfy these memory requirements.¹⁻⁶ Already the storage capacities of magnetic disc files used in peripheral and mass storage have been considerably increased and are expected to improve further. Magnetic core memories for mainframe storage are making way for the considerably faster MOS semiconductor technology. Charge coupled devices, magnetic bubble memories and electron beam accessed memories which offer slightly higher densities with a tradeoff in access time have been demonstrated (Table 1).

For future computer systems, optical memories appear to be attractive candidates for several important reasons. Optical data storage is capable of providing extremely high packing densities limited only by the wavelength of light. In principle 10^8 bits/cm² for two-dimensional storage is possible, but in practice this is limited to about 10^7 bits/cm². Considerably greater storage and processing capacities are possible in the case of volume storage with a theoretical limit of 10^{12} bits/cm³. The estimated practical limit for such a system is about 10^{10} bits/cm³.⁷ Furthermore, optical techniques permit both fast random access time (μ s or less) and data transfer rates on the order of G bits/sec. The upper limits for access time and throughput are determined by the ultimate capabilities attainable by the various system components. Bit error rates and cost per bit have been speculated to be one error in 10^8 retrieved bytes and 10^{-4} cents/bit respectively. In addition, optical memories offer the advantage of immunity from electromagnetic interference.

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Optical data storage can be accomplished either by bit-by-bit recording or by holographic recording. High storage densities⁸ and minimum information losses from localized defects can be achieved only by holographic recording techniques. Holographic optical memories were first proposed about a decade ago. Although research has been going on in this area ever since, holographic memories in photorefractive media have so far not proved economically practical in commercial storage and display systems. One important reason for this has been the cost factor of developing optical memories as compared to existing memory technology. We can identify a similar long period of development for magnetic bubble memory technology which is only within the last year or so reaching economic viability as a result of technical advances on a broad front. Current optical memories and display systems use several relatively complex, high cost optical system components which present a serious drawback from a manufacturer's point of view. A typical holographic recording arrangement (Figure 1) consists of: a coherent light source, beam deflector, a data input element or page composer,

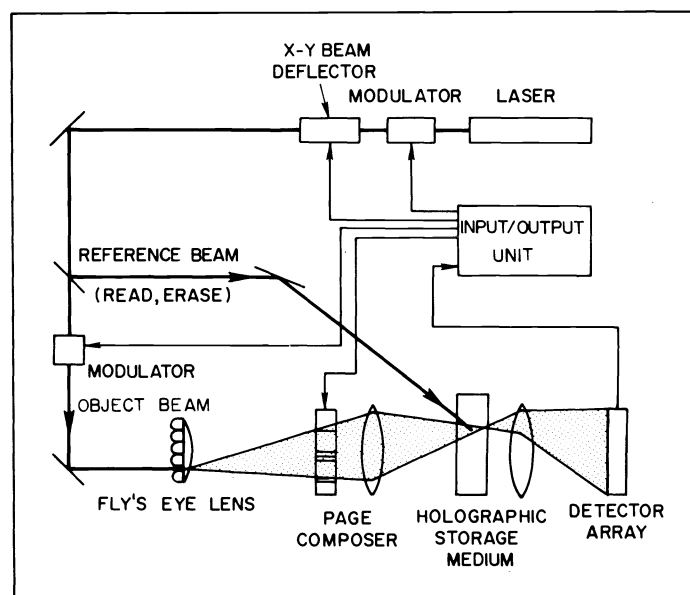


Figure 1. Experimental arrangement for a holographic read-write memory.

a recording medium and a detector matrix. Considerable research has gone into each of these principal components⁹ and considerable progress has been made. This has led to the development of several memory and display prototypes but in order to develop the potentials of holographic optical storage on a commercial scale, further progress needs to be made in the areas of low-cost, coherent light sources, fast and effective beam deflectors, high-speed page composers and optical storage mate-

Table 1. Comparison of Various Memory Technologies

	Access Time	Maximum Storage Density	Maximum Data Rate	Cost Per Bit
1. Magnetic Tapes and Cassettes	5-300 secs.	0.1-1 Mbits/cm ³	10 Mbits/sec	3-300 μ ¢/bits
2. Flexible Discs	0.2-0.5 secs.		2.5 Mbits/sec	100-200 m¢/bit
3. Moving Head Discs	10-50 msec	70 Mbits/cm ³	40 Mbits/sec	1-50 m¢/bit
4. Fixed Head Discs, Drums	3-30 msec.		40 Mbits/sec	50-500 m¢/bit
5. Magnetic Bubbles	0.5-50 msec.	10 ⁵ -10 ⁷ bits/cm ² (projected 10 ⁸ -10 ⁹ bits/cm ²)	100-500 Kbits/sec (projected 10 Mbits/sec)	50-500 m¢/bit
6. Charge Coupled Devices (CCD)	0.05-2 msec.		1-10 Mbits/sec	0.1-0.5 ¢/bit
7. Electron Beam Accessed Memories (EBAM)	0.01-3 msec.	15-60 Mbits/cm ²	10-200 Mbits/sec	0.2-0.3 ¢/bit
8. Magnetic Core	0.1-5 μ sec.		10 Mbits/sec (bit-wise)	0.1-5 ¢/bit
9. Metal Oxide Silicon Random Access Memories (MOS RAM)	0.1-5 μ sec	0.1-1Mbits/cm ²	10 Mbits/sec	0.1-5 ¢/bit
10. Bipolar Random Access Memories (Bipolar RAM)	5-500 nsec.	0.01-0.1 Mbits/cm ²	200 Mbits/sec	0.2-100 ¢/bit
11. Holographic Optical (projected figures)	< μ sec	1-10 Mbits/cm ² 0.1-10 Gbits/cm ³	Gbits/sec	0.1 m¢/bit

rials. Until recently the lack of a convenient storage medium has been a particular hindrance to the practical implementation of holographic read-write memory systems. Although there exist various optical recording materials^{10,11} there are none that exhibit all the desired optical and physical characteristics.

The requirements of an ideal optical recording medium include:

- The medium must be a photosensitive material that is physically and chemically stable over long periods of time under normal storage conditions; to enable real-time storage and read-out, the medium should not require complex processing, and it should not impose constraints on the required operating environment, ideally operating in air at normal temperature and pressure; the medium must be easy to fabricate in thick, large area, defect-free sheets with reproducible characteristics, possess a linear exposure dependence, fast response and a large dynamic range.
- Recording of information in the medium should require very small energy densities, i.e., high recording photosensitivity, comparable to the sensitivity of photographic emulsions; in order to realize the full potential of volume holographic recording and readout with high signal-to-noise ratio the medium should be grainless (i.e., low intrinsic noise) and should allow superposition in the same volume of the material of holograms recorded at different angles and different wavelengths without unwanted erasure; the effect utilized for storing information in the medium should have a long relaxation time so that stored holograms have a long lifetime.
- The medium should be reversible so that it can undergo a large number (greater than 10⁹) of write-read-erase cycles without fatigue (i.e., without degradation of the medium) and with short (\approx 1 ms) write-erase cycle time with low energy requirements for erasure.

- The readout should be nondestructive with high diffraction efficiency; the medium should allow resolution of spatial frequencies on the order of 10⁴ lines/mm with the recording sensitivity being independent of spatial frequency.

Several different media and various mechanisms are being investigated for volume holographic storage. In particular certain photosensitive electro-optic crystals appear to be very promising. These crystals exhibit optically induced changes of the refractive index which is known as the photorefractive effect. This paper describes the state of the art of photorefractive materials in holographic data storage, display and processing applications.

The Photorefractive Effect

The photorefractive effect was initially discovered in the non-linear applications of crystals such as LiNbO₃ and LiTaO₃ where it gave rise to undesirable light-scattering and decollimation and hence was referred to as the laser-induced index damage effect.¹² The photo-refractive effect depends upon the optical excitation and subsequent transport and retrapping of electrons which originate from localized centers in such crystals (Figure 2). The optical excitation may be due either to a single photon effect or to a nonlinear multiphoton absorption, depending on the nature of the coherent source used. Photo-excited space charge patterns, resembling the intensity profile of the generating holographic intensity pattern in turn set up electric fields that distort the index of refraction of the crystal via the electro-optic effect. Thus the refractive index modulation which gives a volume phase hologram may be used as a mechanism for information storage. Localized centers which provide the free electrons upon excitation are impurity ions present in significant concentrations even in undoped crystals and substantial improvements in sensitivity have been made by doping electro-optic crystals with transition metal ions.

Phase holograms recorded in electro-optic crystals by means of the photorefractive effect may be experimentally investigated primarily in terms of the holographic diffraction efficiency, η .

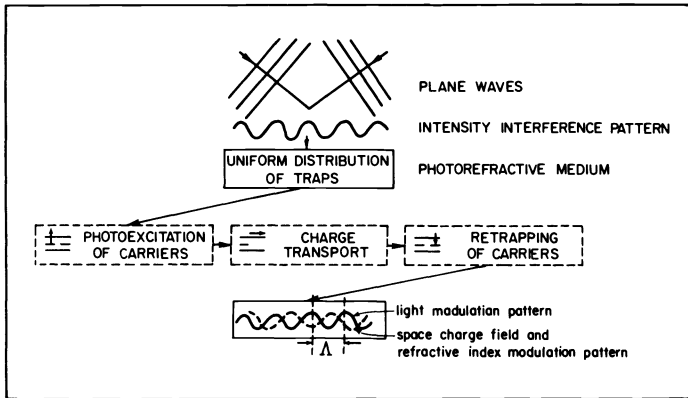


Figure 2. Schematic explanation of the photorefractive effect.

Diffraction efficiency is the fraction of the read beam that is diffracted into the reconstructed image beam. Therefore, a theory for photorefractive volume phase hologram formation must explain the experimentally observed characteristics of the diffraction efficiency of such a hologram. In general, any hologram may be regarded, to a certain extent, to be composed of conceptually simpler sinusoidal grating holograms. Analysis of the diffraction efficiency of a photorefractive hologram has therefore been commonly confined to sinusoidal grating produced by the interference of two plane waves assumed to be infinite in extent. The interference pattern generated by a plane reference and signal wave of wavelength λ and intensity I_0 intersecting at an angle 2θ (Figure 3) is given by

$$I(x) = I_0(1 + m \cos Kx) \quad (1)$$

where $K = 2\pi/\Lambda$ is the grating vector, $\Lambda = \lambda/2\sin\theta$ is the wavelength of the grating, and m is the depth of intensity modulation. Typical fringe spacings are of the order of 1 to 10 μm . We

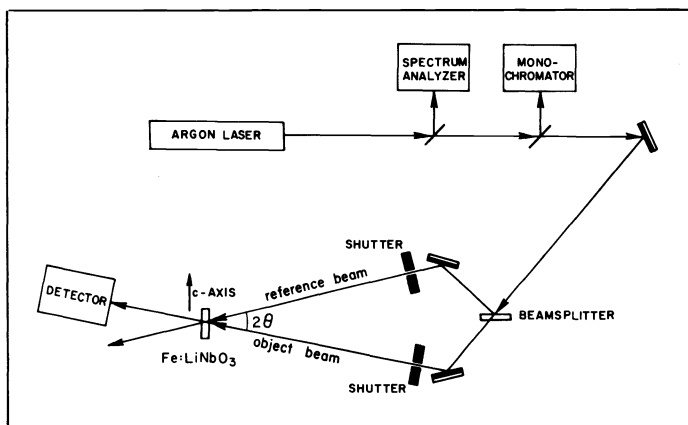


Figure 3. Schematic for recording volume holograms with plane waves.

assume that this interference pattern is incident on a photo-sensitive crystal containing a uniform distribution of localized centers that can donate electrons to the conduction band by photoexcitation at this wavelength. If N is the donor concentration in the crystal, σ the optical absorption cross-section of the electrons and $\hbar\omega$ the energy of the incident photon, then the

photo-electron generation rate may be written as

$$g(x) = g(1 + m \cos Kx), \text{ where } g = \frac{I_0 \sigma N}{\hbar\omega} \quad (2)$$

The continuity equation for free electron concentration n that accounts for photoexcitation, electron transport and trapping with a trapping time τ is given by

$$\frac{\partial n}{\partial t} = g(x) - \frac{n}{\tau} + \frac{1}{|e|} \frac{\partial J}{\partial x} \quad (3)$$

The mechanisms for electron transport in photorefractive materials are not yet fully understood. Early theories described the transport of charge carriers in terms of diffusion or drift, in a constant internal electric field.^{13,14} More recently, Glass *et al*¹⁵ have suggested a new approach, namely that the charge carriers are photogenerated and displaced in a spatially preferred direction. However, for the purposes of analyses of the formation of photoexcited space charge fields, all the different postulated mechanisms other than diffusion may be accounted for by assuming drift in a total equivalent field E . Then the current density J is given by

$$J(x) = e \mu n(x)[E + \mathcal{E}(x)] + e D \frac{\partial n}{\partial x} \quad (4)$$

where μ is the mobility of electrons and D the diffusion coefficient.

A description for the formation of phase holograms may be obtained if one can obtain a time dependent solution for the space charge field $\mathcal{E}(x)$, by solving equations (3) and (4) together with Gauss's Law^{16,17}

$$\frac{\partial}{\partial x} [E + \mathcal{E}(x)] = \frac{\rho}{\epsilon} \quad (5)$$

and the continuity equation

$$\frac{\partial J}{\partial x} = -\dot{\rho} \quad (6)$$

We have recently developed a nonlinear dynamic theory for the space charge field¹⁸ that considers the effect of the photo-generated space charge field on the transport of electrons. The temporal behavior of the space charge field has been obtained explicitly by using the coupled mode approach to analyze the feedback mechanism existing between the photogenerated field and the free electron density. Our solution for the space charge field reads as follows:

$$\mathcal{E}(t) = A(t) \cos [Kx - \varphi_g(t)] \quad (7a)$$

$$A(t) = m[\mathcal{F}_c^2(t) + \mathcal{F}_s^2(t)]^{1/2} \quad (7b)$$

$$\varphi_g(t) = \tan^{-1} [\mathcal{F}_s(t)/\mathcal{F}_c(t)] \quad (7c)$$

Expressions for $\mathcal{F}_c(t)$ and $\mathcal{F}_s(t)$ which have been derived in detail in Ref. 18, determine the evolution in time of the amplitude and phase of the space charge field (Figure 4). As mentioned previously, this space charge field produces a spatial refractive index modulation Δn , through the electro-optic effect.

$$\Delta n(x, z, t) = \frac{1}{2} n_e^3 r \mathcal{E}(x, z, t) \quad (8)$$

where n_e is the extraordinary index of refraction and r is the electro-optic coefficient. In this manner a phase grating is created which is directly related to the original light distribution incident on the crystal.

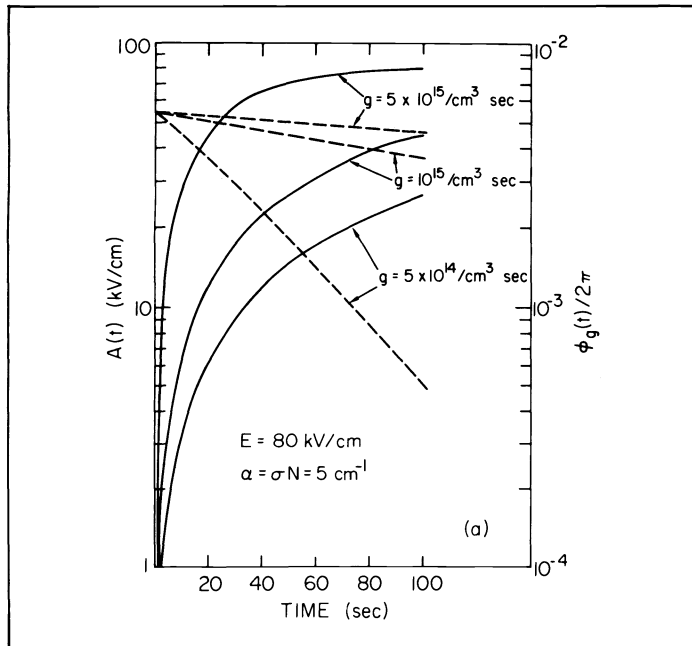


Figure 4. Evolution of the amplitude and relative phase of the photo-generated space charge field.

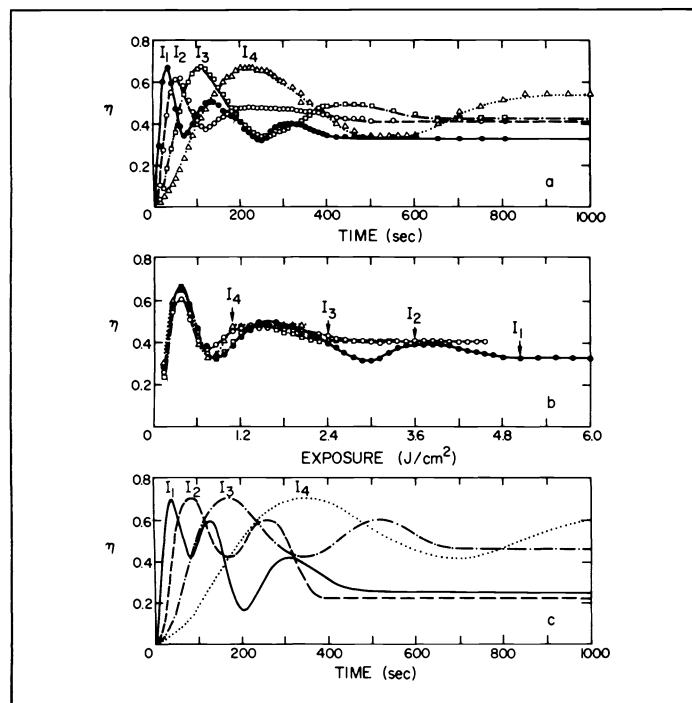


Figure 5. (a) Experimental curves of diffraction efficiency η as a function of writing time for four different intensities: $I_1 = 2I_2 = 4I_3 = 8I_4 = 12 \text{ mW/cm}^2$. (b) The same η curves plotted as a function of exposure ($W = It$). (c) Theoretical diffraction efficiency curves plotted as a function of time for the four intensities used in (a).

For a thick, sinusoidal grating phase hologram Kogelnik¹⁹ has derived an expression for the diffraction efficiency

$$\eta = \sin^2 \left(\frac{\pi d \Delta n}{\lambda \cos \theta} \right) \quad (9)$$

where d is the grating thickness. Hence $\eta \propto (\Delta n)^2$ or $[\mathcal{E}]^2$. Uchida²⁰ has extended this theory to apply to attenuated gratings. More recently we have obtained²¹ a time and space dependent solution for the diffraction efficiency that is in good agreement with the experimentally observed behavior of the diffraction efficiency of photorefractive holograms (Figure 5). Furthermore, this analysis can also provide a novel technique for determining the nature and magnitude of the total equivalent electron drift field E .²¹

Materials Development and Characterization

Important material parameters that influence the photorefractive effect include (a) a large linear electro-optic coefficient, (b) suitable absorption characteristics, (c) an optimal concentration of traps, and (d) a suitable carrier transport mechanism. Although this effect has been observed in several pyroelectric crystals such as LiNbO_3 , LiTaO_3 , BaTiO_3 , SBN, KTN and PLZT, their relatively low recording and erasure sensitivities compared to other optical recording media has been a considerable practical drawback. At this time, the most promising of these crystals is LiNbO_3 , whose photorefractive sensitivity can be increased several orders of magnitude by optimizing the charge generation and transport mechanism. The photorefractive sensitivity is defined as the amount of incident light energy density required to produce a phase hologram with a given diffraction efficiency or with a given refractive index modulation. Recently, Micheron *et al*²² have reported very high hologram recording sensitivities comparable to the photographic plate photosensitivities in $\text{Bi}_{12}\text{SiO}_{20}$ and $\text{Bi}_{12}\text{GeO}_{20}$, but so far with relative short storage lifetimes (~ 24 hours).

The effectiveness of the three processes, electron photo-generation, transport and trapping, responsible for the photorefractive effect in pyroelectric crystals is determined by the nature and concentrations of the impurity ions present in the crystals. An ion in one charge state may act as an electron donor while the same ion in a different charge state may act as an acceptor. Thus their concentrations affect the rates of electron generation and trapping. Furthermore, their concentrations may determine the efficiency of the electron transport mechanisms. Photorefractive sensitivity is therefore strongly influenced by the nature and concentration of impurities present in the crystal whether intentionally or inadvertently.

Several transition metal elements such as Fe, Co, Ni, Cu, Mn, Cr, Rh and U have been used for doping LiNbO_3 and other photosensitive crystals in an attempt to enhance the photorefractive sensitivity. One of the best materials so far in terms of the photorefractive sensitivity is Fe:LiNbO_3 . In Fe:LiNbO_3 , Fe^{2+} ions are believed to be responsible for generation of electrons while Fe^{3+} ions appear to be electron acceptors. The photorefractive sensitivity of Fe:LiNbO_3 therefore depends on the relative concentration of the Fe^{2+} and Fe^{3+} ions. Hence, it is desirable to optimize the relative concentrations of Fe^{2+} and Fe^{3+} ions in Fe:LiNbO_3 crystals by controlling the oxidation-reduction state of such crystals.

The physical and chemical properties of doped pyroelectric crystals important in holographic applications have been studied and characterized by several different spectroscopic techniques.²³ Spectral measurements include x-ray emission spectra, IR absorption spectra, optical absorption and EPR measurements. Optical absorption spectra were recorded for Fe:LiNbO_3

crystals (Figure 6). The intensity of the 424.5 nm band has been ascribed to changes in Fe^{2+} concentrations which in turn have been identified by both EPR spectrometry and magnetic susceptibility measurements. Recently, a direct confirmation of the existence of a Fe^{2+} state in LiNbO_3 has been made by investigating the Mössbauer spectra of $\text{Fe}:\text{LiNbO}_3$.²⁴

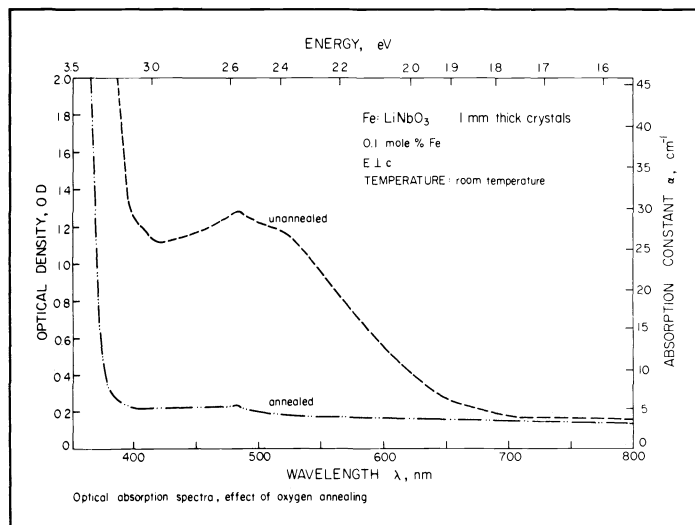


Figure 6. Optical absorption curves for $\text{Fe}:\text{LiNbO}_3$ crystals with the same Fe concentration but with different heat treatments.

From the optical and EPR data we can not determine the absolute concentrations of Fe^{2+} and Fe^{3+} ions for crystals with the same dopant concentrations but with different heat treatments. We find that the photorefractive sensitivity increases almost linearly with the Fe^{2+} concentration but after a certain

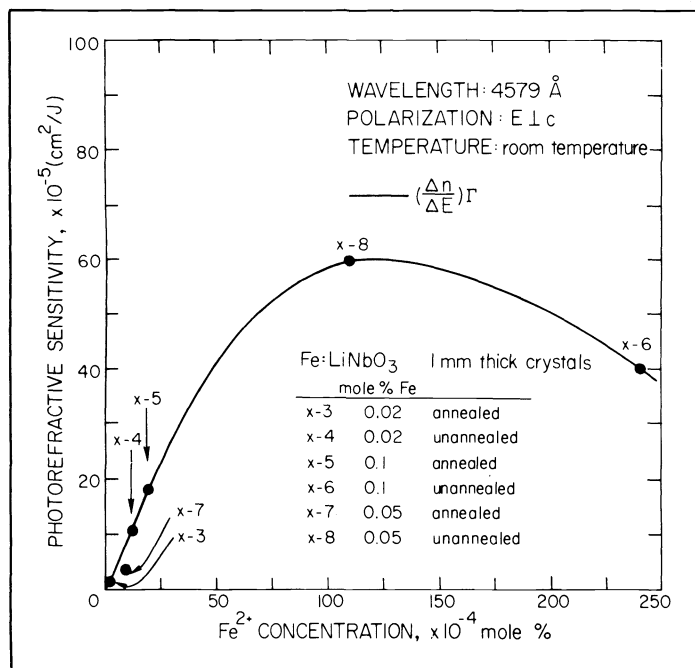


Figure 7. Dependence of sensitivity for photorefractive recording on Fe^{2+} concentration in $\text{Fe}:\text{LiNbO}_3$.

point begins to decrease with further increase in Fe^{2+} concentration (Figure 7). Furthermore, the decrease in photorefractive

sensitivity upon oxygen annealing follows a pattern very similar to that of the decrease in Fe^{2+} concentration (Figure 8), clearly indicating the dependence of sensitivity on Fe^{2+} concentration. For a crystal of given thickness with a high electro-optic coefficient, such as LiNbO_3 , that has been doped with the appropriate transition metal such as Fe there exist optimum values of iron and its valence states Fe^{2+} and Fe^{3+} at which the process of electron generation, transport and trapping and hence the CW photorefractive sensitivity are optimized. Any further increase in the photorefractive sensitivity is limited by the accompanying increase in optical absorption. For 1 mm thick $\text{LiNbO}_3:\text{Fe}$ crystals, we find the optimum values of Fe, Fe^{2+} and Fe^{3+} to be: 0.05 mole % Fe with $\text{Fe}^{2+}/\text{Fe} \sim 20\text{-}25\%$ and $\alpha \sim 13.5 \text{ cm}^{-1}$.²³

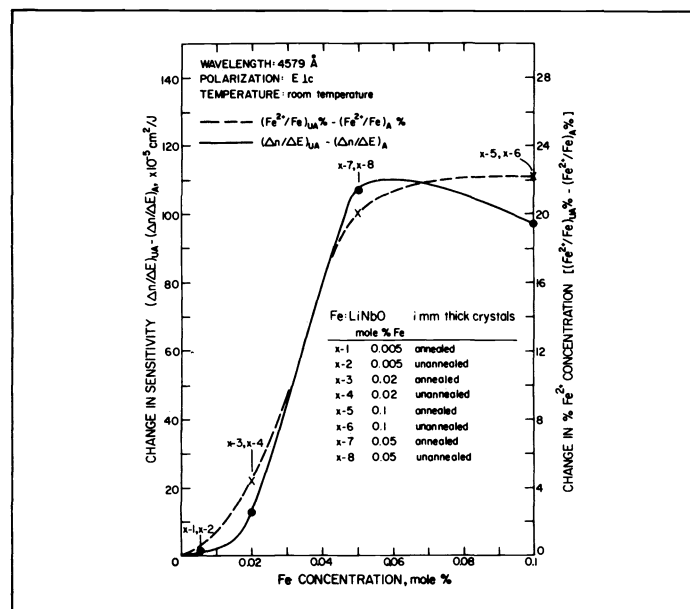


Figure 8. Comparison of change in photorefractive sensitivity and change in percentage of Fe^{2+} concentrations in $\text{Fe}:\text{LiNbO}_3$ crystals.

Techniques

In order to realize the full potential of photorefractive materials for volume recording and to demonstrate the feasibility of holographic optical memories several techniques have been investigated.

Recording with High Intensity Pulses

For CW recording the photorefractive sensitivity is based on a single photon effect. Von der Linde *et al*⁶ have obtained greatly increased photorefractive sensitivity by utilizing multiphoton absorption in both doped and undoped LiNbO_3 . Recording holograms with 500 MW/cm² pulses from a mode-locked and doubled Nd:glass laser, they observed a photorefractive frequency sensitivity 130 times higher than the CW photorefractive sensitivity of $\text{LiNbO}_3:\text{Cu}^{2+}$ at approximately the same wavelength (0.53μ) and writing energy density. The increased photorefractive sensitivity was attributed to two-photon absorption. Similar experiments with KTN⁶ yielded even higher sensitivities. However, the optical quality of presently available KTN is as yet not very good. Substantial reduction in the pulse intensities required for the photorefractive recording were also observed using two-step excitation via a real intermediate state in $\text{LiNbO}_3:\text{Cr}^{3+}$ and $\text{LiTaO}_3:\text{Cr}^{3+}$.

These experiments open the possibility of using tunable, high peak power, CW or pulsed, mode-locked lasers for enhancing the

photorefractive sensitivities of suitably doped pyroelectric crystals via multiphoton processes.

Avoiding Unwanted Erasures

One of the basic requirements for a read-write memory or optical processing is that the storage medium should allow easy and fast erasure. This is possible in the case of a photorefractive material. Photorefractive recording relies upon optical excitation and redistribution of electrons into a pattern closely resembling the incident intensity interference pattern due to the recording beams. Therefore, illuminating such a charge pattern with a uniform optical beam results in a uniform redistribution of charge, which results in erasure of the stored information. The time required for erasure can be made small by choosing crystals with high photoconductivity. Distortionless reconstruction of a hologram containing many spatial frequencies requires that the same wavelengths be used for reconstruction as used for recording. Since the reconstruction of readout process is similar to the erasure process the readout of a photorefractive hologram can result in its unwanted erasure. Therefore, a technique is needed to avoid unwanted erasure of a hologram when reading a hologram or when reading or recording one or many superimposed holograms.

Several different techniques have been considered to avoid unwanted erasure of photorefractive holograms:

Low Erasure Sensitivity. By controlling the concentration of the impurity ion and its valence states, crystals have been prepared that exhibit high storage sensitivity but low sensitivity for erasure.²⁵

Fixing. Fixing a hologram implies replacing the electronic charge pattern by a more stable ionic charge pattern. This may be accomplished either thermally or electrically. In thermal fixing²⁶ the crystal in which a hologram is stored is heated to a temperature at which the ions in the lattice become relatively mobile while the trapped electronic charges remain thermally stable (between 100 and 200 C). The ions are transported due to drift in the space charge fields until the trapped electronic charge is neutralized. Upon cooling the crystal the electronic charge is redistributed by incoherent illumination leaving behind a permanent ionic pattern. In electrical fixing techniques²⁷ local changes of spontaneous polarization in ferroelectric crystals are caused by the electronic space charge fields of a hologram in the presence of externally applied fields smaller than the ferroelectric coercive field. The resulting drift of ions cancels the internal space charge fields. Upon uniform illumination with no applied field, the electrons are redistributed and an ionic pattern remains behind. Fixed holograms may be used for optical display or for read-only memories.

Multiphoton Excitation. The above mentioned multiphoton photorefractive recording techniques for improving optical recording sensitivities are useful in eliminating unwanted erasure.^{6,28} This may be explained as follows. Suppose $\hbar\omega$ is the energy required to excite an electron from the trap to the conduction band via a single photon effect. Then at sufficiently high light intensities the same transition can be caused to occur via a real or virtual intermediate level by using two frequencies ω_1, ω_2 such that $\omega = \omega_1 + \omega_2$. If now two beams of frequencies ω_1 are made to interfere in the medium in the presence of a third beam with frequency ω_2 a hologram will be generated in the medium. The spatial frequency of the hologram is determined by ω_1 yet light at frequency ω_2 is essential for the formation of the hologram. If the hologram is read at ω_1 there is no erasure of the hologram since there is no photoexcitation of the electrons to the conduction band, whereas the hologram

can still be erased by simultaneous exposure to light at frequencies ω_1 and ω_2 .

Multiple Hologram Storage. A particular attractive feature of photorefractive crystals is that they lend themselves to volume recording. Holograms may be recorded throughout the thickness of the medium. The Bragg condition for thick holograms allows such holograms to be accessed only within a certain angular and frequency bandwidth around the angle and frequency of light used for recording them. This angular and wavelength selectivity of volume holograms can be used for superposing in a given volume of the medium holograms recorded at different angles and wavelengths.²⁹ Recently Staebler *et al.*³⁰ have reported the superposition of 512 holograms in Fe:LiNbO₃ by angular superposition. They simultaneously recorded and fixed the holograms by maintaining the crystal at 160 C and rotating the 1 cm thick sample by 0.1° after each recording. They observed good readout quality of the holograms with diffraction efficiencies ranging from 2% to 25% from the last to the first hologram recorded. Such an experiment demonstrates the high density information storage capability of photorefractive materials.

Selective Hologram Erasure

Bulk optical or thermal erasure of superposed holograms will indiscriminately erase all the different stacked holograms. A technique is therefore needed that will selectively erase one or portions of many superimposed holograms. Huignard *et al.*³¹ have demonstrated selective erasure using a coherent optical erasure technique. The principle underlying this technique is as follows: Let H be one of many superposed holograms. If H is to be erased, one records on H at the same wavelength and angle as used for H, a complementary hologram of H. This is done by recording H with a phase difference π introduced in the reference beam. If part P of the hologram H is to be erased then recording P on H with a phase difference of π introduced in the reference wave will erase P.

Device Characteristics

The ultimate aim in understanding the physical processes, improving the material characteristics and devising new techniques for photorefractive holography is to develop a memory device with optimum characteristics for a specific optical information processing application. Considerable progress has been made in realizing practical devices with presently available technology.

Diffraction Efficiency

For plane wave recording diffraction efficiencies very close to 100% have been observed in some LiNbO₃:Fe crystals and values of diffraction efficiency above 50% are not uncommon.

Recording Sensitivity

Sensitivity for photorefractive recording may be defined as

$$S = \left(\frac{\Delta n}{\Delta E} \right) \Gamma \quad \text{where,} \quad \Gamma = \left(\frac{1 - e^{-\alpha L}}{\alpha L} \right)$$

and ΔE is the incident energy density required to produce a refractive index modulation with amplitude Δn . The magnitude of S depends on the total electric field acting on the charge carriers or the effective drift length $L = \tau \mu [E + \mathcal{E}(x)]$. The best value for CW photorefractive sensitivity is approximately 6×10^{-4} (cm²/J). The values of sensitivity for recording with short pulses in LiNbO₃:Cu is approximately 2×10^{-4} (cm²/J) whereas for KTN, it is approximately 3 orders of magnitude

better.⁶ $\Delta E/\Gamma$ is the average energy density inside the crystal, α , L being the attenuation coefficient and crystal thickness, respectively.

Erasure Sensitivity

Erasure sensitivity may be defined as the reciprocal of the light energy density required to erase the hologram to 1/e of its initial diffraction efficiency. Erasur sensitivities up to 100 (cm^2/J) have been reported in $\text{Fe}:\text{LiNbO}_3$ ²⁵ with erasure times up to several seconds.

Resolution

For a 1 cm thick crystal of LiNbO_3 a spatial resolution exceeding 1600 lines/mm has been observed.³² SBN, BaTiO_3 and PLZT have also been used to reproduce holograms with resolution greater than 1000 lines/mm.

Storage Lifetime

The thermal relaxation time of holographic fields in ferroelectric materials such as LiNbO_3 is determined by the conductivity and the dielectric constant of the material. At room temperature and in the absence of light the conductivity is very low and lifetimes of several weeks to months have been found in practice. At lower temperatures (close to 0 °C) the thermal relaxation time can be increased to several years.¹⁴

Storage Density

As mentioned earlier the experimentally realizable range of storage density for volume storage is between 10^8 and 10^{10} bits/ cm^3 . With 10^4 bits per page, 10 holographic pages have been superposed in a 1 cm thick $\text{LiNbO}_3:\text{Fe}$ crystal, to yield a bit density on the order of 10^5 bits/ cm^3 .⁷ In another experiment over 500 holograms have been written in $\text{Fe}:\text{LiNbO}_3$ by angular superposition.³⁰

Signal-to-Noise Ratio (SNR)

The ultimate storage capacity of a photorefractive crystal will be limited by the SNR required to read out the image. Recently Burke *et al.*³³ have investigated the noise sources intrinsic to volume holography in $\text{LiNbO}_3:\text{Fe}$. They conclude that statistical noise due to granularity in the electronic space charge, scatter noise due to medium imperfections and noise due to intermodulation distortion are not significant. Cross-talk noise between superposed holograms can be reduced considerably at angular spacings of 0.1°, allowing superposition of a large number of holograms.

Conclusions

Holographic optical memories offer definite advantages for information processing on account of their high storage density and fast access time capabilities. Photorefractive materials are attractive candidates for the storage medium in read, write, random-access holographic memories and displays. Among the desirable properties of photorefractive materials are the following: (1) real-time write, read and erasure capability, (2) high storage density due to volume recording and superposition of holograms, (3) high diffraction efficiency phase holograms, (4) high resolution and low noise readout, (5) long lifetime of information storage and (6) *in situ* thermal, optical or electrical erasure.

The feasibility of holographic read-write memories with a photorefractive material as a storage medium has been experimentally demonstrated by several groups.^{7,30} For example,

Ref. 7 describes a system in which an array of 5×5 $\text{LiNbO}_3:\text{Fe}$ crystals is used as an optical memory. Transparent electrodes are provided to allow local thermal erasure of each crystal. Erasure times are of the order of several tens of seconds. High density of storage is achieved by superposition of holographic pages by angular rotation of the reference beam. Ten holographic pages, each containing 10^4 bits were stored in each location of the memory plane. A page in a block of superimposed holograms was accessed by rotating the reference beam with an acousto-optic beam deflector that allowed an access time of roughly 3 μ sec. With further improvements in the performance of beam deflectors, page composers and the photorefractive storage crystal, much higher densities and faster access times should be feasible. In particular, research is needed in (1) understanding the charge transport mechanisms and in identifying electron acceptors and donors in photorefractive materials, (2) improving recording and erasure sensitivities, (3) reducing record-erasure cycle time, (4) avoiding unwanted erasure of a hologram during readout or storage of a superposed hologram, (5) allowing superposition of a higher number of holograms with low noise and high spatial resolution, and (6) assuring long information storage lifetimes.

References

1. Haughton, K. E., "An Overview of Disk Storage Systems," *Proc. IEEE*, Vol. 63, pp. 1148-1152. Aug. 1975.
2. Bobeck, A. H., Bonyhard, P. I. and Geusic, J. E., "Magnetic Bubbles - An Emerging New Memory Technology," *Proc. IEEE*, Vol. 63, pp. 1176-1195. Aug. 1975.
3. Hughes, W. C., Lemmond, C. Q., Parks, H. G., Ellis, G. W., Possin, G. E. and Wilson, R. H., "A Semiconductor Nonvolatile Electron Beam Accessed Mass Memory," *Proc. IEEE*, Vol. 63, pp. 1230-1240. Aug. 1975.
4. Chen, D. R. and Zook, J. D., "An Overview of Optical Data Storage Technology," *Proc. IEEE*, Vol. 63, pp. 1207-1230. Aug. 1975.
5. Feth, G. C., "Memories: Smaller, Faster and Cheaper," *IEEE Spectrum*, Vol. 13, pp. 36-43. June 1976.
6. von der Linde, D. and Glass, A.M., "Photorefractive Effects for Reversible Holographic Storage of Information," *Appl. Phys.*, Vol. 8, pp. 85-100. 1975.
7. d'Auria, L., Huignard, J. P., Slezak, C. and Apitz, E., "Experimental Holographic Read-Write Memory Using 3-d Storage," *Appl. Opt.*, Vol. 13, pp. 808-818, April 1974; Optical Systems and Photosensitive Materials for Information Storage, *Optical Properties of Solids*, ed. B. O. Seraphin, 1976, pp. 851-922.
8. Vander Lugt, A., "Design Relationships for Holographic Memories," *Appl. Opt.*, Vol. 12, pp. 1675-1685. July 1973.
9. Special Issue on Optical Storage of Digital Data, *Appl. Opt.*, Vol. 13, April 1974.
10. Kurtz, R. L. and Owen, R. B., "Holographic Recording Materials - A Review," *Opt. Eng.*, Vol. 14, pp. 393-400. Sept.-Oct. 1975.
11. Special issue of RCA Review, Optical Storage and Display, Vol. 33, March 1972.
12. Ashkin, A., Boyd, G. D., Dziedzic, J. M., Smith, R. G., Ballman, A. A., Levinstein, H. J. and Nassau, K., "Optically Induced Refractive Index Inhomogeneities in LiNbO_3 and LiTaO_3 ," *Appl. Phys. Lett.*, Vol. 9, pp. 72-74. July 1966.
13. Chen, F. S., "Optically Induced Change of Refractive Indices in LiNbO_3 and LiTaO_3 ," *J. Appl. Phys.*, Vol. 40, pp. 3389-3396. July 1969.
14. Amodei, J. J. and Staebler, D. L., "Holographic Recording in LiNbO_3 ," *RCA Rev.*, Vol. 33, pp. 71-93. March 1972.
15. Glass, A. M., von der Linde, D. and Negran, T. J., "High-Voltage Bulk Photovoltaic Effect and the Photorefractive Process in LiNbO_3 ," *Appl. Phys. Lett.*, Vol. 25, pp. 233-235. Aug. 1974.
16. Amodei, J. J., "Analysis of Transport Processes During Holographic Recording in Insulators," *RCA Rev.*, Vol. 32, pp. 185-198. June 1971.
17. Young, L., Wong, W. K. Y., Thewalt, M. L. W., and Cornish, W. D., "Theory of Formation of Phase Holograms in Lithium Niobate," *Appl. Phys. Lett.*, Vol. 24, pp. 264-265. March 1974.
18. Kim, Dae M., Shah, Rajiv R., Rabson, T. A. and Tittel, F. K., "Nonlinear Dynamic Theory for Photorefractive Phase Hologram Formation," *Appl. Phys. Lett.*, Vol. 28, pp. 338-340. March 15, 1976.
19. Kogelnik, H., "Coupled Wave Theory for Thick Hologram Gratings," *Bell Syst. Tech. J.*, Vol. 48, pp. 2902-2947. Nov. 1969.

20. Uchida, N., "Calculation of Diffraction Efficiency in Hologram gratings Attenuated Along the Direction Perpendicular to the Grating Vector," *J. Opt. Soc. Am.*, Vol. 63, pp. 280-287. March 1973.
21. Kim, Dae M., Shah, Rajiv R., Rabson, T. A. and Tittel, F. K., "Study of the Equivalent Electron Drift Field Characteristics in LiNbO_3 by Phase Holography," *Appl. Phys. Lett.*, Vol. 29, pp. 84-86. July 15, 1976.
22. Micheron, F., Peltier, M. and Huignard, J. P., "High Photosensitivity Volume Hologram Recording in $\text{Bi}_{12}\text{SiO}_{20}$ and $\text{Bi}_{12}\text{GeO}_{20}$ by Efficient Charge Transport Process," *IX International Conf. on Quant. Elec.*, Amsterdam. June 1976.
23. Shah, Rajiv R., Kim, Dae M., Rabson, T. A. and Tittel, F. K., "Characterization of Iron Doped Lithium Niobate for Holographic Storage Applications," *J. Appl. Phys.* Vol. 47, pp. 5421-5431. Dec. 1976.
24. Keune, W., Date, S. K., Dezsi, I. and Gonser, U., "Mossbauer Effect Study of Co^{57} and Fe^{57} Impurities in Ferroelectric LiNbO_3 ," *J. Appl. Phys.*, Vol. 46, pp. 3916-3924. Sept. 1975.
25. Staebler, D. L. and Phillips, W., "Fe-doped LiNbO_3 for Read-Write Applications," *Appl. Opt.*, Vol. 13, pp. 788-794. April 1974.
26. Staebler, D. L. and Amodei, J. J., "Thermally Fixed Holograms in LiNbO_3 ," *Ferroelectrics*, Vol. 3, pp. 107-113. 1972.
27. Micheron, F. and Bismuth, G., "Electrical Control of Fixation and Erasure of Holographic Patterns in Ferroelectric Materials," *Appl. Phys. Lett.*, Vol. 20, pp. 79-81. Jan. 1972.
28. von der Linde, D., Glass, A. M. and Rodgers, K. F., "Optical Storage Using Refractive Index Changes Induced by Two-Step Excitation," *J. Appl. Phys.*, Vol. 47, pp. 217-220. Jan. 1976.
29. Gaylord, T. K. and Tittel, F. K., "Angular Selectivity of Lithium Niobate Volume Holograms," *J. Appl. Phys.*, Vol. 44, pp. 4771-4773. Oct. 1973.
30. Staebler, D. L., Burke, W. J., Phillips, W. and Amodei, J. J., "Multiple Storage and Erasure of Fixed Holograms in Fe-doped LiNbO_3 ," *Appl. Phys. Lett.*, Vol. 26, pp. 182-184. Feb. 1975.
31. Huignard, J. P., Herriau, J. P. and Micheron, F., "Coherent Selective Erasure of Superimposed Volume Holograms in Fe-doped LiNbO_3 ," *Appl. Phys. Lett.*, Vol. 26, pp. 256-258. March 1975.
32. Chen, F. S., La Macchia, J. T. and Fraser, D. B., "Holographic Storage in Lithium Niobate," *Appl. Phys. Lett.*, Vol. 13, pp. 223-224. Oct. 1968.
33. Burke, W. J., Sheng, P. and Weakliem, M. A., "Intrinsic Noise Sources in Volume Holography," RCA Final report - prepared for ONR. Dec. 1975. ⊗