Optical storage in LiNbo3 : Fe with selective erasure capability
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1. Introduction. — Three dimensional storage using phase hologram recording in transparent media is a technique which allows high storage density and capacity, since the minimum bit size may be as small as the cube of the optical wavelength [1]. Photosensitive electrooptics belong to the most promising class of three dimensional optical storage materials [2]: illumination causes mainly a change in refractive index, their spatial resolution is diffraction limited, they don’t require any revealing process and they are reusable. Some of them require low recording light energy (100 µJ/cm² for K(Ta,Nb)O₃ using a two photons process [3], 3 mJ/cm² for Sr₀.75Ba₀.25Nb₂O₆ [4]), or show strongly assymmetric recording-erasure cycles, which allow multiple hologram superimpositions (more than 500 holograms superimposed in LiNbO₃ : Fe [5]).

In the most cases, the storage time is found experimentally equal to the dielectric relaxation time (some seconds to some months), and this is the reason why photoinduced change of refractive index is attributed to photoinduced electric fields, and not to polar defects reorientations or photoinduced dipole moments. Indeed, it was demonstrated that photocarriers excited from impurity centers can move on macroscopic distances in these crystals before being trapped, which generate space charge fields and refractive index changes via the electrooptic effect.

The carrier transport processes can be isotropic diffusion [6], or anisotropic diffusion in polar materials which give rise to a bulk photovoltaic effect [7], or drift under the influence of an applied field [8, 9], or combinations of these three effects. They generate a current in an external circuit connected to the crystal under illumination such as:

\[ J = q D \frac{dn(x)}{dt} + \alpha I(x) + [\sigma_0 + \beta I(x)] E(x) \]  

The diffusion current exists only for non uniform illuminations \( I(x) \), which cause a gradient of photocarriers concentration \( n(x) \).

The photovoltaic current is proportional to the absorbed light power \( \alpha I(x) \) and can be measured only parallel to the polar axis of the crystal. The coefficient \( K \) is odd function of the spontaneous polarization, and depends upon the nature and value of the impurity ions. The third term in the current \( J \) accounts for the crystal conductivity and photoconductivity (photocarriers drifts) in presence of the electric field \( E(x) \).

The field \( E(x) \) inside the crystal is given by the electrical displacement \( D \)

\[ D = \int_{0}^{r} J \, dt \]  

and the photoinduced refractive index change is obtained using the appropriate electrooptic coefficients.

Using this model, we derive the operating conditions which provide the best use of the full dynamic range in refractive index changes, especially for holograms superimpositions and coherent erasures, in Fe doped LiNbO₃ crystals (0.015 % Fe, 3 mm thickness, reduced in Ar atmosphere for 50 % absorption at \( \lambda = 514 \) nm).
2. Uniform illuminations of Fe doped LiNbO₃ crystals. — The LiNbO₃ crystal is first uniformly illuminated, and the short circuit current \( i_{cc} \) is measured between two electrodes perpendicular to the polar c axis (Fig. 1a). Using eq. (1), \( i_{cc} \) is given by

\[
i_{cc} = A \cdot \alpha K I \tag{3}
\]

where \( A \) is the electrodes area. Since no photovoltage can be generated in this short circuited crystal, it is expected from the previous model that the refractive index remains unchanged. The refractive index was measured using a low power He-Ne laser and crossed polarizers, and no index change was observed under illuminations by an Ar laser at \( \lambda = 514 \) nm, up to incident powers of 100 mW/cm² (the experimental arrangement limits the measure to index changes larger than \( 10^{-6} \)). At higher powers, or long exposure times, an increasing scattering is observed in the transmitted beams, whereas no scattering effect can be detected with the same energy density, using an incoherent light source. This anomalous scattering effect under coherent illumination can be attributed to surface defects or bulk inhomogeneities: the coherent incident light beam is slightly scattered by the defects at the beginning of illumination, and the randomly scattered beams interfere inside the crystal. They cause a randomly non uniform repartition of illumination, which is therefore recorded as new refractive index inhomogeneities.

This effect limit to some \( J/cm^2 \) the energy density which can be absorbed during operations at room temperature. Nevertheless, the crystal recovers its low scattering state after heating at 200 °C for some minutes: the increase in conductivity decreases the dielectric relaxation time \( \tau \), therefore, complete erasure of all the recorded refractive index changes occurs by heating during times larger than \( \tau \).

Considering now the arrangement of figure 1b, where the crystal is uniformly illuminated in open circuit, i.e. \( J = 0 \). A space charge field \( E_{sc} \) can be developed by photovoltaic effect, which saturation value is given by

\[
E_{sc} = -\frac{\alpha K I}{\sigma} \tag{4}
\]

where \( \sigma \) is the conductivity under illumination. An homogeneous photoinduced change of refractive index is now observed, as previously reported by Glass [7], and the same scattering effects appear as shown in the short circuit configuration.

3. Non uniform illumination with holographic gratings. — Using the previous results, one can now deduce the spatial index distributions during hologram gratings recording, for open circuit and short circuit conditions. The spatial illumination repartition is

\[
I(x) = I_0 (1 + m \cos kx) \tag{5}
\]

where \( k = 2 \pi /\Lambda \), and \( \Lambda \) is the fringe spacing. The fringes are perpendicular to the polar axis.

In the LiNbO₃ : Fe crystals used, both diffusions current at \( \Lambda = 10 \) μm and photoconductivity current can be neglected compared to the photovoltaic current. This is not the case for LiNbO₃ : Fe crystals with higher ratios Fe³⁺/Fe²⁺ (less reduced crystals) in which photoconductivity must be taken into account.

Therefore, eq. (1, 2) reduce to

\[
j(x, t) = \frac{\alpha K I}{\sigma} E(x, t) + \varepsilon \frac{\partial E(x, t)}{\partial t} \tag{6}
\]

open circuit configuration is described by the limiting condition :

\[
j(x, t) = 0 \tag{7}
\]

and solution for \( E(x, t) \) is therefore

\[
E(x, t) = \frac{\alpha K I_0}{\sigma} (1 + m \cos kx) \left(1 - \exp \left(-\frac{\sigma t}{\varepsilon} \right)\right) \tag{8}
\]

Both continuous and modulated terms in the holographic grating are recorded: during hologram superpositions, the different continuous terms are added, which causes an increase of the average change of refractive index (and would contribute to saturate the refractive index change in photoconductive crystals). Optical erasure by uniform illuminations causes erasure of the modulated component only, and increases the average change of refractive index.

In the short circuit configuration (Fig. 2b), the limiting condition is

\[
\int_0^t E(x, t) \, dx = 0 \tag{9}
\]

solution of eq. (6) is therefore (\( A \ll \) crystal length \( \ell \))

\[
E(x, t) = \frac{\alpha K I_0}{\sigma} m \cos kx \left(1 - \exp \left(-\frac{\sigma t}{\varepsilon} \right)\right) \tag{10}
\]

The continuous term in the holographic grating is no more recorded, and therefore, the short circuit configuration allows multiple hologram superimposition and provides a true optical erasure. This will be shown in the next section, for the particular case of coherent optical erasure.
4. Selective erasure of superimposed volume holograms. — Information pages can be superimposed at the same location of the storage medium by slightly changing the beam incidence outside of the Bragg selectivity range. Erasure of such recorded holograms is usually performed by uniform illumination of the storage medium at the recording wave-length (or smaller) or by heating. Both of these techniques cause a bulk erasure process which apply indiscriminately to the different holograms and lead to limitations in applications of such a storage process [10]. Selective erasure of any stacked hologram or information bit is demonstrated by using a coherent subtraction method.

The storage crystal used for these experiments is iron doped and reduced LiNbO$_3$-0.015 % Fe — which shows a high resistance to optical erasure by uniform illumination — (asymmetric Recording-Optical erasure cycle) [11]. The crystals having such a cycle are suitable for multiple storage, since the recording beams of a new hologram cause a weak erasure of the previously recorded ones in the same volume. The coherent erasure concept applies to such crystals providing a compact system with a greatly increased storage density and maintaining the versatility of erasure of any stacked hologram or bit in a page.

4.1 OPEN CIRCUIT RECORDING. — When recording the Fourier hologram of a transparent object having a nearly uniform spectrum, the refractive index change at spatial frequency $k$ is deduced from eq. (8), in open circuit recording conditions, using the linear electro-optic coefficients (Fig. 3a):

$$n(x, y, z) = n_0 + \delta n \cos [kx + \varphi(x, y, z)]$$

$n_0$: bulk photoinduced index change developed by d. c. term of holographic pattern (low spatial frequency),

$\delta n$: index modulation characteristic of fringes modulation (high spatial frequency).

4.2 SHORT CIRCUIT RECORDING. — From the analysis performed in the previous section, recording and selective erasure operations should be done with the crystal short circuited. With such conditions eq. (10) shows that the index change is established around the original value (Fig. 3b).

$$n(x, y, z) = \delta n \cos [kx + \varphi(x, y, z)]$$
and by coherent erasure process, the new photo-
induced change is:

\[ n'(x, y, z) = -\delta n \cos [kx + \phi(x, y, z)] . \]

After such a Recording-Coherent erasure cycle, the
initial index value is really recovered (Fig. 3b) and the
number of cycles should not be limited. Nevertheless,
the number of cycles is limited to a tenth by the scatter-
ing effect, which increases the noise after each cycle.
The non recording of the d.c. terms of hologram
patterns also provides an increase in the number of
superimposed images since in practice a relatively low
fringe modulation is used for high quality image
reconstruction \((m \ll 1)\).

5. Experimental results. — Complete erasure of an
information page can be performed using the coherent
erasure process. Selective erasure of any information
block or bit can be also achieved by recording of a
partially masked transparency with \(\pi\) shift. The
common parts of the two transparencies are erased.
This corresponds the logical operation Exclusive OR.
Experimental confirmation is demonstrated in figure 4
(measured recording sensitivity : 250 mJ/cm\(^2\)\(\eta = 1\%\)),
which shows complete erasure, selective and only bit by
bit erasure in a single recorded hologram.

![Figure 4](image)

Three holograms of the same binary page have been
superimposed by changing the reference beam inci-
dence on the crystal. These angle changes are provided
by step mechanical translation of reference beam and
converted into rotation by a lens (Fig. 5). The experi-
mental sequence is the following:

- Recording of three stacked holograms of the
  same data plane (Bragg angles \(\theta_1, \theta_2, \theta_3\))
  (hologram diameter : 1.7 mm; Efficiency \(\eta = 1\%\);
  Beam ratio : 13).
- Selective erasure of one single bit in the page
  (Fig. 6b).
- Repositionning of reference beam at Braggs
  angle \(\theta_2\) and selective erasure in the page (Fig. 6b).
- Repositionning the reference beam on Bragg
  angle \(\theta_1\), and complete erasure of the third hologram
  (Fig. 6b).

Figure 7 demonstrates the possibility of logical ope-
rations between two binary transparencies \(A\) and \(B\)
(metallic grids for avoiding phase distortion). In the
particular situation where \(B\) is a uniform object trans-
parency (\(B = 1\)) the reconstructed image has a reversed
contrast. The experimental sequence of figure 7 shows
the logical operation \(A + B\).

6. Comments. — For these experiments, precau-
tions must be taken to avoid mechanical vibrations of
the components. The laser stability and \(\pi\) shift are
continuously controlled by projection of a magnified
portion of the fringes on a vidicon with display on a TV
monitor. The phase shift has been obtained either with
an Electro-optic modulator, or by switching a Babinet
plate between two positions. The problem encountered
with our crystals is photoinduced scattering which
rapidly degrades the signal to noise ratio of reconstruc-
ted images. After several Recording-Coherent erasure
cycles, we observe the scattered light at the same level
as that obtained by continuously reading one holo-
gram with the reference beam during the same period
of time. This effect limit actually in our crystals the
number of superimposed images an selective erasure
cycles to about 10 and the potential large dynamic
range of the crystal is not used. As mentioned in [5]
recording with heated could reduce the build up of this
coherent scattering.

From system consideration, the ideal material would
be a storage crystal with a completely dissymmetrical
cycle and no photoinduced scattering. Information
to be erased should be readed on the photodetector
matrix and rewritten on the page composer for selec-
tive erasure.
SELECTIVE ERASURE

Fig. 5. — System configuration for selective erasure and processing in stacked holograms.
SELECTIVE ERASURE IN STACKED HOLOGRAMS

Fig. 6. — Selective erasure of stacked holograms: Fig. 6a: images reconstruction from three stacked holograms; Fig. 6b: selective erasure in three stacked holograms with reference beam repositioning $\theta_1$, $\theta_2$, $\theta_3$, single bit erasure (Bragg angle $\theta_3$), partial erasure (Bragg angle $\theta_2$), complete erasure (Bragg angle $\theta_1$).
FIG. 7. — Logical operations between two binary transparencies \( A \) and \( B \) (operation \( \overline{A + B} = A \overline{B} \)).

References