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SPECTRAL HOLE BURNING AND HOLOGRAPHIC IMAGE STORAGE

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Abstract Spectral hole-burning allows a large number of holograms to be stored within inhomogeneously broadened absorption bands of dye doped polymer films. Employing an electric field as an additional dimension to address the images, the storage of 25 holograms within the spectral range of one wavenumber has recently been achieved. It is shown that the relative phase between the holograms must be controlled in order to optimize the storage density.

INTRODUCTION

The ratio of inhomogeneous to homogeneous broadening occurring at very low temperatures in the electronic $S_1 \leftarrow S_0$ absorption bands of dye doped polymers can be as large as 10^6. Spectral hole-burning is a spectroscopic technique to improve resolution beyond inhomogeneous broadening limits and has been widely used for the investigation of optical relaxation processes, molecular properties and guest-host interaction in condensed matter[1-3]. The broadening ratio determines the gain in experimental resolution which can be achieved and determines the number of spectral holes which can be burnt into the inhomogeneously broadened band. If the presence of a spectral hole at a specific optical frequency is associated with a digital "1"and the absence of a spectral hole with a digital "0", up to 10^6 bits could be stored at a single spatial spot of μm size[3-7]. To increase the storage density of optical memory devices towards 10^{14} bits/cm^2 would be highly attractive. The enormous gain in storage capacity has initiated many research activities to develop optical storage devices based on spectral hole-burning[8,9] despite serious constraints, such as the need for very low temperatures and high quality light sources. The spectra shown in fig. 1 indicate the potential of this storage technology. The

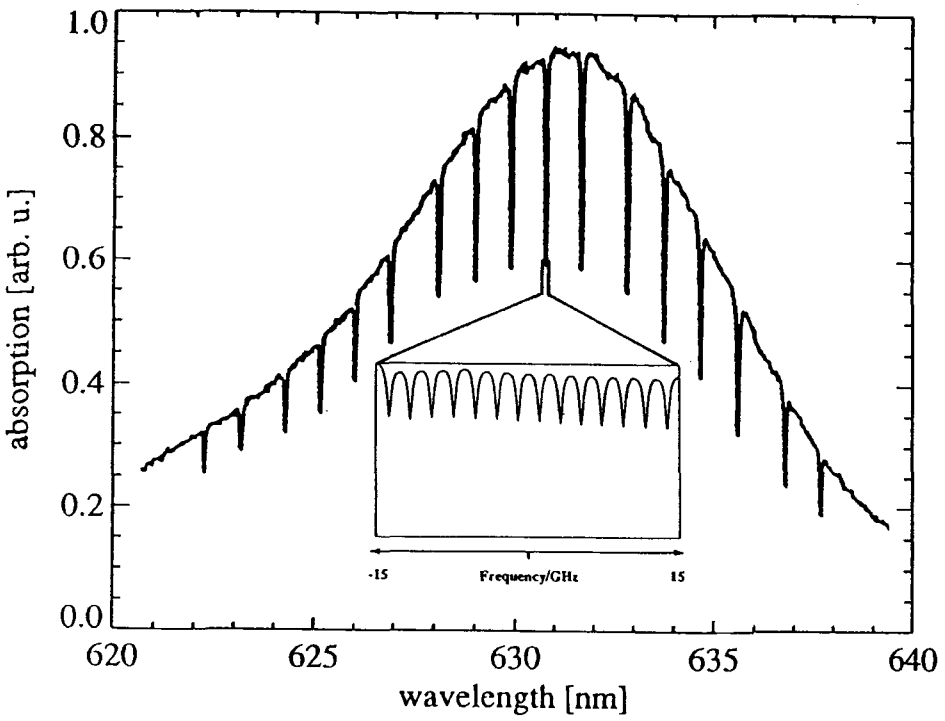

Fig. 1. Lowest singlet absorption band of chlorin in a polyvinylbutyral host detected with low resolution. Each of the 17 spectral holes can be associated with a set of 14 holes detected with high resolution as shown inset.

low resolution spectrum shows the $S_1 \leftarrow S_0$ absorption band of chlorin in a polyvinylbutyral (PVB) host. The seventeen holes distributed across the whole band were detected using a monochromator, resulting in an apparatus limited holewidth of approximately 1 cm^{-1} (30 GHz). This spectral width corresponds to the whole tuning range of the high resolution spectrum with a set of fourteen holes as shown inset. The holewidth in the high resolution spectrum, depending on the temperature, is 0.5 GHz, allowing the holes to be burned at a separation of 2 GHz. Thus, approximately 6'000 spectral holes could be burnt into the spectrum shown in fig. 1. An electric field applied to the sample as an additional dimension facilitates a further increase in storage density[10,11].

Classical detection methods such as transmission detection and fluorescence detection give easily evaluable signal shapes but in many cases suffer from light source (laser) instabilities and interference effects and become ineffective when very small relative changes in absorption are to be measured. Generally, rela-

tively small signals are observed against a large background which in most cases is not shot noise limited. A very elegant detection method, where the background subtraction is performed optically, is based on laser induced grating techniques or holography[12]. Holographic detection of spectral holes was developed in order to improve the detection sensitivity[13] and was subsequently used for the investigation of the temperature dependence of optical dephasing[14] and for the study of electric field effects on spectral holes[15,16]. Making use of the imaging properties of holography, images can be stored as spectrally narrow holograms. Laser frequency or electric field selective recording of images in a dye doped polymer film (oxazine-4 in polyvinylbutyral) has recently been demonstrated[17,18]. An important property of holography due to interferometric recording is the storage of the relative phase between reference and object beam. In this paper we show how the storage capacity i.e. the number of images in a given spectral range (1 cm^{-1}), can be improved when an electric field is used in combination with the frequency dimension.

HOLOGRAPHIC DETECTION

In a laser induced grating experiment the sample is illuminated by the interference pattern created by two crossed laser beams. This is shown in fig.2 for the most simple hologram, two plane waves of wavelength λ, crossing at an angle of 2Θ.

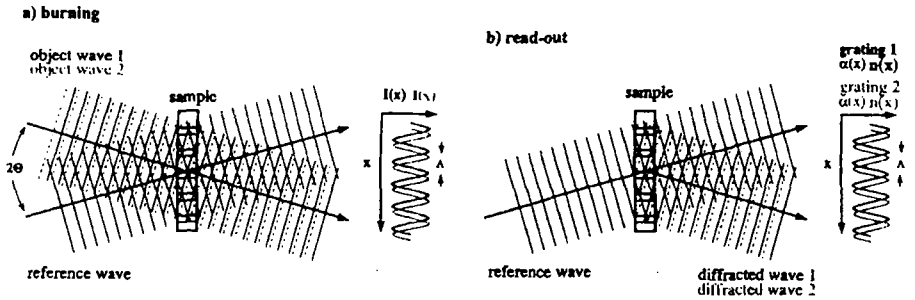

Fig. 2. Fundamental principle of holographic detection and illustration of the phase of a hologram. The sample is illuminated with the reference pattern of two plane waves (a) indicated by the solid lines. A phase shift in one of the beams gives rise to the corresponding phase shift of the interference pattern at the sample. When the reference wave is diffracted from two different gratings (b) the two diffracted waves may interfere constructively or destructively depending on the relative phase of the two gratings.

The intensity pattern, see fig. 1, is described by

$$I(x) = (I_1 + I_2)[1+V\cos(2\pi x/\Lambda + \varphi)] \quad (1)$$

with the fringe contrast, $V = 2\sqrt{I_1 I_2}/(I_1 + I_2)$, depending on the intensities of the two beams, I_1 and I_2, and the fringe spacing, $\Lambda=\lambda/2\sin\Theta$. The hologram phase, φ, describes the spatial position of the interference pattern along the x-axis. φ is connected directly to the relative phase of the interfering light beams. When the phase of one of the beams is shifted by a fraction of a wavelength the interference pattern of the two waves at the sample moves by the same fraction of the fringe spacing. This is indicated by the dashed lines in fig. 2. It is clear that the reproducibility in recording of single holograms depends very strongly on the stability of the fringe pattern at the sample.

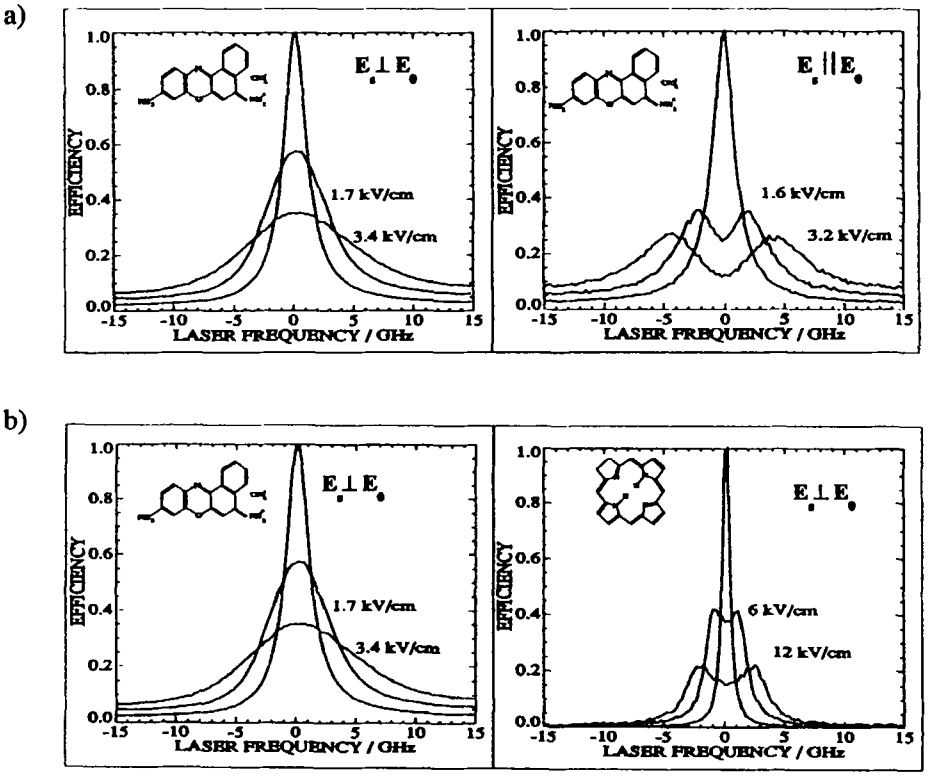

Fig. 3. Influenc of experimental geometry and molecular properties on the shapes of spectral holes. (a) one molecule, two different geometrical arrangements: electric field E_s perpendicular and parallel with respect to the light polarization E_0; (b) two different molecules, cresyl violet and chlorin, one experimental geometry ($E_s \perp E_0$).

The modulated intensity pattern creates an excited state grating and, due to the hole-burning process persistent gratings of the absorption coefficient and refractive index are formed. The hologram efficiency of such a grating was recently investigated based on Kogelnik's coupled wave theory[19]. For shallow holes (weak burning) the frequency dependence of a spectral hole is given by[20]

$$\eta(\omega) = \exp[-\alpha_0(\omega)d)]\,[\alpha_1(\omega_b)\cdot d/4]^2 \cdot (\Gamma/2)^2 / [(\omega_b - \omega)^2 + (\Gamma/2)^2] \qquad (2)$$

Eq. 2 describes a spectrally very narrow hologram. The amplitude is given by the maximum hole depth at the burning position, $\alpha_1(\omega_b)$, and the frequency dependence follows exactly the Lorentzian hole shape as observed in conventional detection methods. $\eta(\omega)$ describes a zero background signal since it approaches zero far away from the burning position, ω_b; for this reason, holography can be regarded as an ideal technique for the detection of spectral holes.

When an electric field is applied to the sample the energy levels of the guest molecules are shifted due to the interaction of the molecular dipole moments with the external field. The transition frequency of a specific guest molecule is thus shifted by (neglecting quadratic Stark effect)

$$\Delta\omega = -(1/\hbar)\ \vec{\Delta\mu} \cdot \vec{E}_s \qquad (4)$$

The frequency shift $\Delta\omega$ depends on the difference of dipole moments, $\vec{\Delta\mu} = \vec{\mu}_1 - \vec{\mu}_0$, between the excited ($S_1$) and the ground state ($S_0$) and on the local field, $\vec{E}_s$. Due to the isotropic orientation of the dye molecules the spectral hole shows a broadening

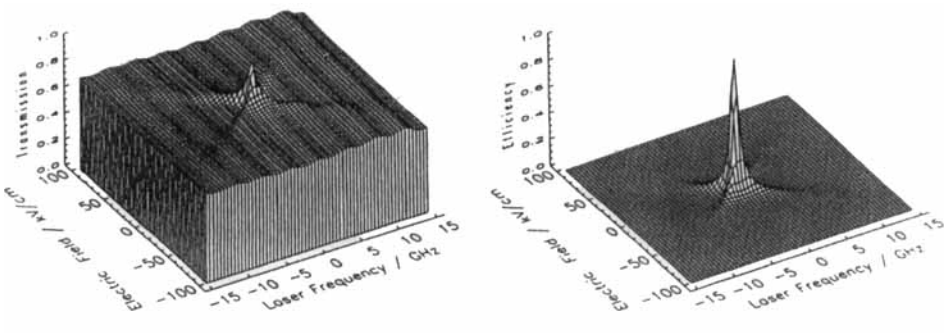

Fig. 4. Diffraction efficiency of a single spectral hole plotted as a function of the read-out laser frequency and the electric field applied to the sample.

or a splitting depending on the molecular and experimental geometry[15]. From the electric fields influenced hole shapes the molecular dipole moment difference, $\Delta\mu$, can be derived[21-25]. In fig. 3, the dependence of the shapes of spectral holes on experimental geometry, i. e. the direction of the light polarization, $\vec{E}_0$, with respect to the electric field, $\vec{E}_s$, is shown. Hole spectra obtained with the dye cresyl violet in a polyvinylbutyral host are shown for perpendicular and parallel orientations of $\vec{E}_s$ and $\vec{E}_0$. In fig.3(b), hole spectra are shown for one geometry ($\vec{E}_s \perp \vec{E}_0$) but for two different molecules, cresylviolet and chlorin. The direction of the dipole moment difference between the ground and excited state with respect to the transition moment is clearly established.

In fig. 4 the electric field dependence shown before for chlorin is plotted as a three dimensional representation. The hole was simultaneously detected in transmission (a) and by means of holography (b). Both, the transmitted and the diffracted intensity are plotted as a function of the probing frequency of the laser and the electric field applied to the sample. The hole was burnt in the center of the tuning ranges (30 GHz, 200kV/cm) and appears as a very narrow peak.Many holes can be stored in such a two dimensional plane, ideal for optical storage. The difference between the two detection techniques is impressively shown; whereas the transmission signal appears against a considerable background, the holographic signal arises from a zero background plane.

CONTROL OF THE GRATING PHASE

Efficient use can be made of interference effects between two light waves diffracted from two different gratings when the position of the pattern can be monitored and accurately controlled . The influence of the grating phase on the interaction of spectrally adjacent holograms has recently been studied[26]. In a hole-burning experiment one hologram, with a given phase, is stored at a burning frequency, ω_1, and a second hologram, with a different phase, at burning frequency, ω_2, as indicated in fig. 2a. Due to their Lorentzian spectral shape, the holograms will overlap. If the sample is illuminated with a reference beam having a frequency in the range between ω_1 and ω_2, two coherent beams are diffracted from the two gratings. This is indicated in fig. 2b. These two diffracted beams interfere constructively or destructively depending on their relative phase . Such interference effects must be

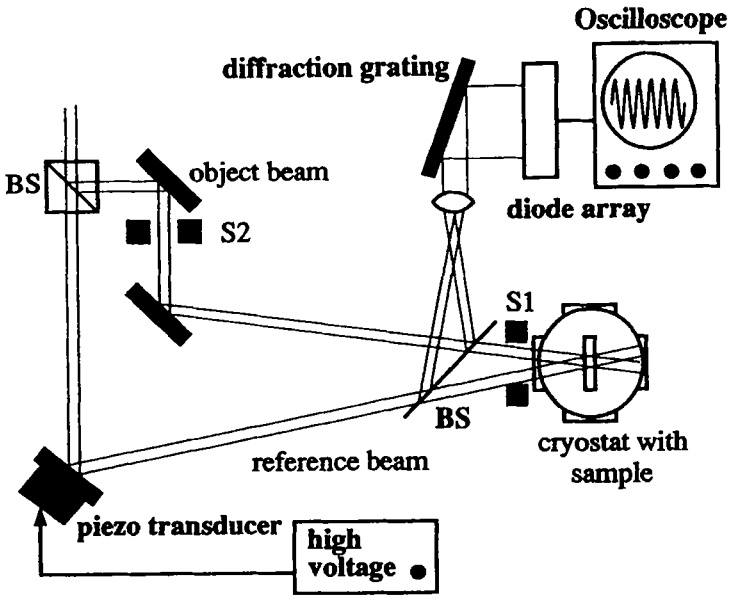

Fig. 5. Experimental arrangement used for observation and adjustment of the phase of the holograms.

taken into account when the spectral density of holograms is to be maximized and crosstalk effects between the individual holograms are to be minimized.

In order to achieve accurate phase control the experimental arrangement[17] has been slightly modified. This is shown in detail in fig. 5. A beam splitter mounted directly in front of the cryostat created a second interference pattern. This fringe pattern expanded using a cylindrical lens and a grating used in grazing incidence. The enlarged fringe pattern was detected by a Reticon photodiode array and could be displayed on an oscilloscope. The adjustment of the grating phase was accomplished by moving the reference beam mirror which was mounted on a piezoelectric transducer. The experimental results suggest a good correlation between the phase changes observed on the oscilloscope and the phase differences of the holograms stored at the sample.

MULTIPLE STORAGE OF HOLOGRAMS

In fig. 6, experimental results are shown for twenty-five spectral holes burnt as a 5x5 matrix in a frequency range of 30 GHz and an electric field range of 200 kV/cm. The holographic signal depends strongly on the relative phases between the holograms during burning. All holograms depicted in fig. 6(a) were burnt with the same phase. Comparing to fig 2b, the holographic signal appears to be no longer background free. The holograms arise against a background, induced by the constructive interference of the long ranging refractive index contributions to the hologram efficiency. This background increases with the number of stored holograms. For the holograms shown in fig. 6(b) a phase difference of π was maintained for all nearest neighbours in the frequency and the electric field domains. The dispersive background has practically vanished due to destructive interference, and the twenty-five holograms arise well resolved from zero background. All the data shown in fig. 6 has been obtained without image information in order to investigate the basic dependence of background and crosstalk on the hologram phase. Based on this information it was subsequently possible to store 25 images at a single spatial position within one wave number, making use of electric field and laser frequency multiplexing. Sequential reconstruction of the stored information by addressing the appropriate values of the electric field and laser frequency re-

a) b)

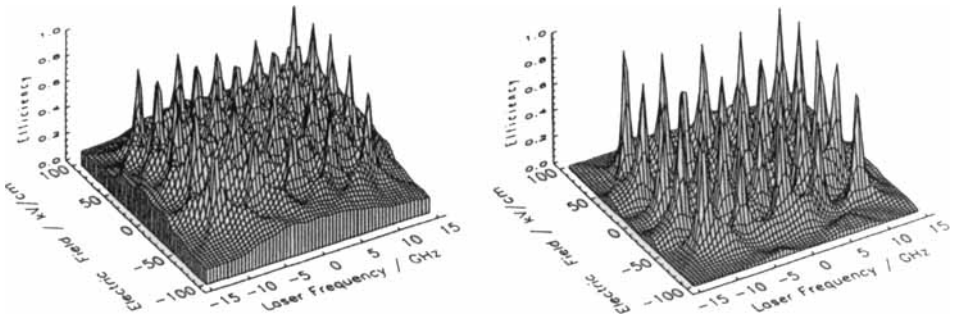

Fig. 6. Experimental results showing a 5x5 matrix of holograms recorded in a plane spanned by the laser frequency and the electric field. The holograms were stored with the same phase (a) and with phase difference π (b) with respect to the nearest neighbours.

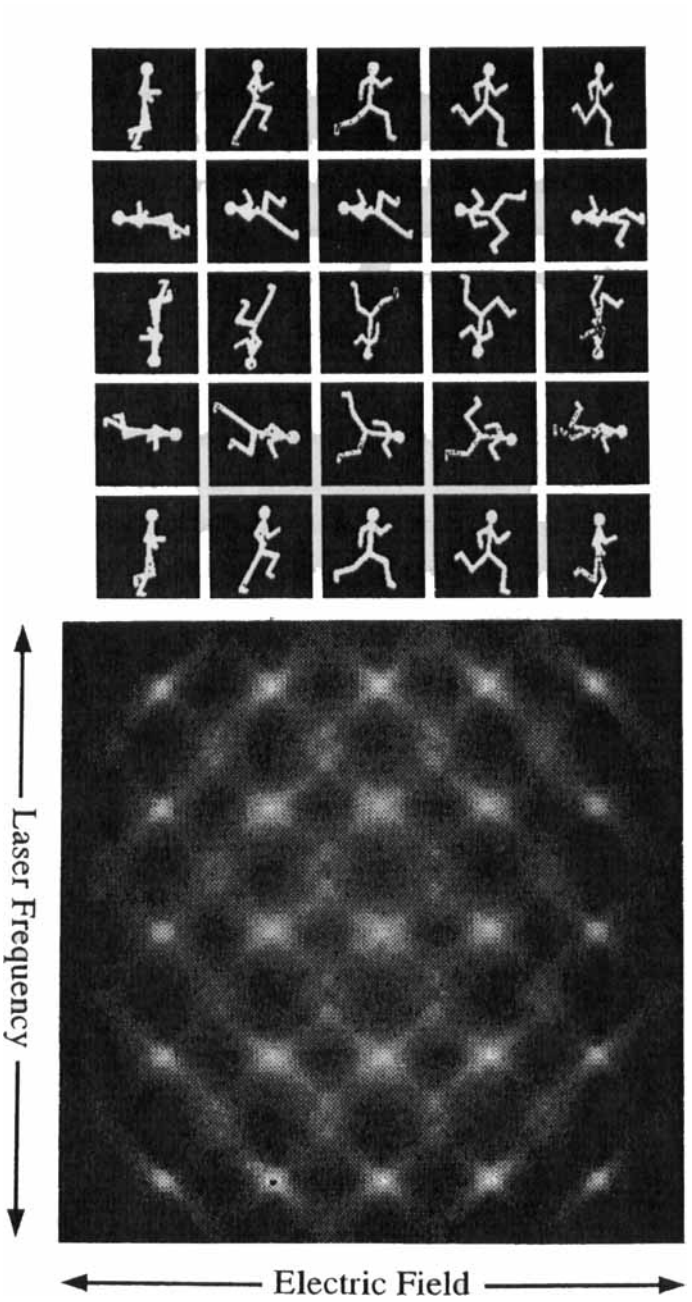

Fig. 7. Density plot of the diffraction efficiency of 25 holograms stored in the laser frequency - electric field plane. Each of the bright spots corresponds to a retrieved image. The different images are shown above.

sults in a 25 image movie. In fig. 7, a density plot of the 25 holograms is shown together with the image information corresponding to each of the holograms. The set of holograms consists of rows of five different stickmen stored in the field dimension with different orientations (frequency). Sequential addressing of the different rows of "image positions" gives the impression of a stickman running in a box. Extrapolating the number of stored images from one wavenumber to the usable range of the inhomogeneously broadened band (about 400 cm^{-1}) this results in approximately 10'000 images, corresponding to a seven minute video movie.

CONCLUSIONS

The present work demonstrates how image storage based on spectral hole-burning and holography can be optimized by controlling the phase of the holograms. An improved reproducibility of the data has been achieved and the storage density can be increased by simultaneously using both the electric field and the laser frequency as dimensions for storing data.

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