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Spectral hole-burning and holography

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Abstract. A review of recent work in which persistent spectral holes have been burnt as laser induced gratings (plane wave holograms) in the inhomogeneously broadened absorption bands of dye doped polymers is presented. The holographic detection technique is compared to the transmission technique and it is shown that a comparison of transmission and holographic hole widths leads to a direct measurement of homogeneous linewidth. Interference between spectrally adjacent gratings is dependent on the relative phase selected during burning. This consideration is important in the analysis of interference between spectrally adjacent gratings which are split through the application of an external electric field. Finally, the influence of the reconstruction symmetry is described.

Keywords. Spectral hole-burning; holography; interference.

1. Introduction

Absorption bands corresponding to electronic transitions of impurities in solids are inhomogeneously broadened at low temperatures. Highly monochromatic dye lasers allow experiments to be conducted on a sub-population of the impurities through energy selection. For many materials, there is a small probability that the process of excitation and relaxation leaves the impurity in a different ground state, thus changing its absorption energy and modifying the material's overall absorption band through the production of a 'spectral hole'. The width of such spectral holes is directly related to the homogeneous linewidth of the electronic transition and is typically of the order of MHz or GHz. As inhomogeneous bands are often broadened to THz, such persistent spectral holes have proved to be accurate probes of structural and electronic properties and of dynamic processes (Moerner 1988; Völker 1989).

When a highly monochromatic laser beam is split and recombined on such a photosensitive material the interference fringes create absorption and refractive index gratings due to spatially selective hole-burning. Subsequent irradiation with one of the beams causes some of the light to be diffracted colinearly with the other. By scanning the probe laser frequency the profile of the spectral hole may be recorded in either transmission or diffraction and the signals compared. Holography has the advantage of being a zero background technique in that there is no diffracted signal outside the range of the hole width.

In this paper the main findings of theoretical and experimental investigations (Meixner *et al* 1989; Renn *et al* 1990; Holliday *et al* 1990) of the properties of spectral

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holes burnt as laser induced gratings is presented. The experimental techniques are briefly described but the full theory is neglected in favour of a phenomenological approach more appropriate to this brief outline. The parallel field of time domain holography (Saari *et al* 1986) is not considered here.

2. Experimental details

The experimental arrangement necessary to record and reconstruct plane wave holograms using spectral hole-burning is shown in figure 1. The additions required to control and monitor the relative spatial phase of the two recording beams are also indicated. The detection channels were equalised so that both could be used to detect either the transmission or the holographic signal by introducing an appropriate neutral density filter (typically with an optical density of 3) in front of the photomultiplier being used to record the transmission signal whilst detecting the unattenuated holographic signal.

Spectral holes were burned in the S_1–S_0 absorption bands of cresyl violet or chlorin in polyvinylbutyral films immersed in superfluid liquid helium. Plane wave holograms could be burnt in 30 s using 10 μW of light in a 3 mm diameter spot deriving from a

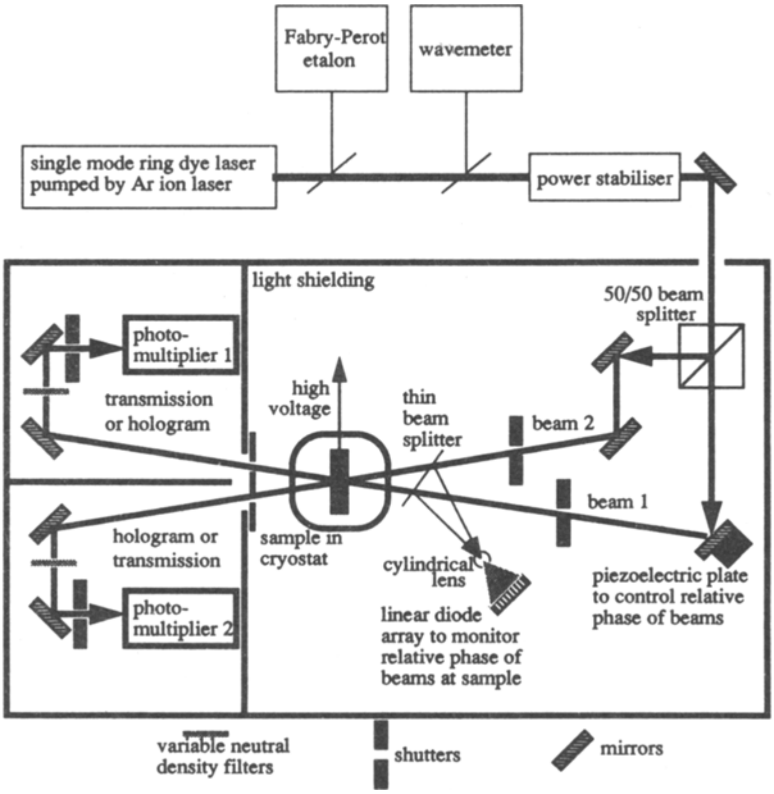

Figure 1. Experimental arrangement for recording and reconstructing holograms through spectral hole-burning.

single mode ring dye laser. Pairs of holes could be burnt in the frequency domain, typically separated by 10 GHz, or in the electric field domain, typically with field strengths of $\pm 2 \times 10^5\ \mathrm{Vm}^{-1}$. The hole widths were approximately 1 GHz and reduced the optical density from about 0·7 to 0·5 at the hole peak. The holograms were reconstructed using each beam with the laser power reduced by a factor of 10 to prevent significant degradation of the gratings due to additional hole-burning during the reconstruction process. Three-dimensional representations of the hologram efficiencies and transmission strengths were produced by stepping the electric field between $\pm 2{\cdot}5 \times 10^5\ \mathrm{Vm}^{-1}$ and the laser over a 15 GHz range. The signal strengths were measured simultaneously for transmission and hologram channels for 50 ms for each of the 64×64 data points.

3. Results and discussion

The parameters governing the creation of single plane wave holograms produced as spectral holes have been investigated experimentally and explained theoretically (Meixner *et al* 1989). As burning time increases the hologram efficiency increases linearly whilst the spatial distribution of removed impurities is sinusoidal, corresponding to the intensity of the interference fringes of the exciting laser. As the absorbing concentration of impurities decreases, however, the grating becomes increasingly more anharmonic. After a well-defined period the holographic efficiency at the exciting frequency reaches a maximum and begins to decrease.

The homogeneous linewidths of electronic transitions reveal much about dephasing mechanisms but are notoriously difficult to measure with certainty (Völker 1989). To extract the genuine homogeneous linewidth from spectral hole-burning measurements the hole width must be extrapolated to zero power and time. It was shown (Meixner *et al* 1989) that when the holographic efficiency reaches a maximum the frequency broadening of the diffracted signal is twice the equivalent parameter for the transmission case. By burning a hole to the maximum holographic efficiency at the laser wavelength and measuring the relative widths of the transmission and holographic signals, the homogeneous linewidth of free base chlorin in a polyvinybutyral film was determined to be 170 MHz. Such a technique determines homogeneous linewidths from deep holes with good signal to noise ratios and precludes the measurement of very weak holes as is necessary when extrapolating to zero fluence.

A study of the diffraction properties of spectrally adjacent holograms (Renn *et al* 1990) revealed the phase dependence of the interference caused by the overlap of hole wings. Plane wave holograms burnt as spectral holes may be considered to be composed of absorption and refractive index gratings each of which causes a diffracted beam $\pi/2$ out of phase with the other. When hologram wings spectrally overlap the diffraction efficiency is determined by the vector sum of the absorption and refractive index components. If the adjacent holograms are burnt with the same relative phase, the vector sum is of absorption parts on the 'real' axis and refractive index parts on the 'imaginary' axis. When the phase of one of the holograms is altered, the vector addition is correspondingly modified causing absorption and refractive index parts to interfere on both axes and thus to change the diffraction efficiency of the holograms. The result is a phase dependent diffraction efficiency for spectrally adjacent holograms, illustrated for two cases in figure 2.

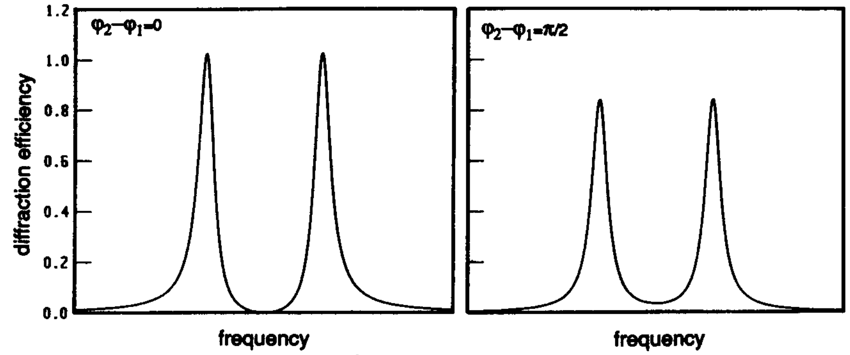

Figure 2. Normalised diffraction efficiencies calculated for the cases of zero phase change between holograms and for a phase change of $\pi/2$.

Application of an electric field to some materials causes a linear Stark shift of the resonant optical transition energy which may split the spectral hole. Split components of spectrally adjacent holes may be overlapped by applying a particular electric field resulting in a superposition of the associated gratings. Cresyl violet was an appropriate impurity for electric field induced interference investigations as the small angle between the transition moment and the dipole moment difference between ground and excited states, and the large magnitude of the dipole moment difference gives rise to an appreciable hole splitting when the electric field vector of the light field is parallel to an applied electric field (Meixner *et al* 1986).

A rigorous derivation of the frequency and electric field dependence of the diffraction efficiency, $\eta(\omega_p, E_s)$, of a pair of spectrally adjacent holograms was obtained (Renn *et al* 1990b). It is given by

$$\begin{aligned}\eta(\omega_p, E_s) = {} & A_1(\omega_p, E_s)^2 + B_1(\omega_p, E_s)^2 + A_2(\omega_p, E_s)^2 + B_2(\omega_p, E_s)^2 + \\ & + [A_1(\omega_p, E_s)\cdot A_2(\omega_p, E_s) + B_1(\omega_p, E_s)\cdot B_2(\omega_p, E_s)] \times \\ & \times \cos(\varphi_2 - \varphi_1) + [B_1(\omega_p, E_s)\cdot A_2(\omega_p, E_s) - \\ & - A_1(\omega_p, E_s)\cdot B_2(\omega_p, E_s)]\cdot \sin(\varphi_2 - \varphi_1). \end{aligned} \qquad (1)$$

This expression contains terms, A_1 and A_2, which describe contributions due to modulation of the absorption coefficient caused by holograms 1 and 2 respectively and terms, B_1 and B_2, which describe contributions due to modulation of the propagation constant (proportional to the refractive index) (Meixner *et al* 1989). The nature of the interference between the gratings when the Stark components are overlapped is determined by the spatial phase difference, $\varphi_2 - \varphi_1$, between the holograms, selected during burning. Good agreement between experiment and theory was obtained for pairs of holograms burnt at different frequencies and zero electric field and for pairs of holograms burnt at the same frequency but at different electric field strengths. Figure 3 illustrates the latter case for a pair of holograms burnt with a phase difference of $\pi/2$. Regions of constructive and destructive interference may be observed in the contour plots as areas containing many and few lines respectively, between the two densely shaded circular regions which correspond to the unperturbed spectral holes. Small differences between experimental data and numerical simulations are explained by slight phase fluctuations during burning and due to matrix induced contributions to the dipole moments not accounted for in the computer model.

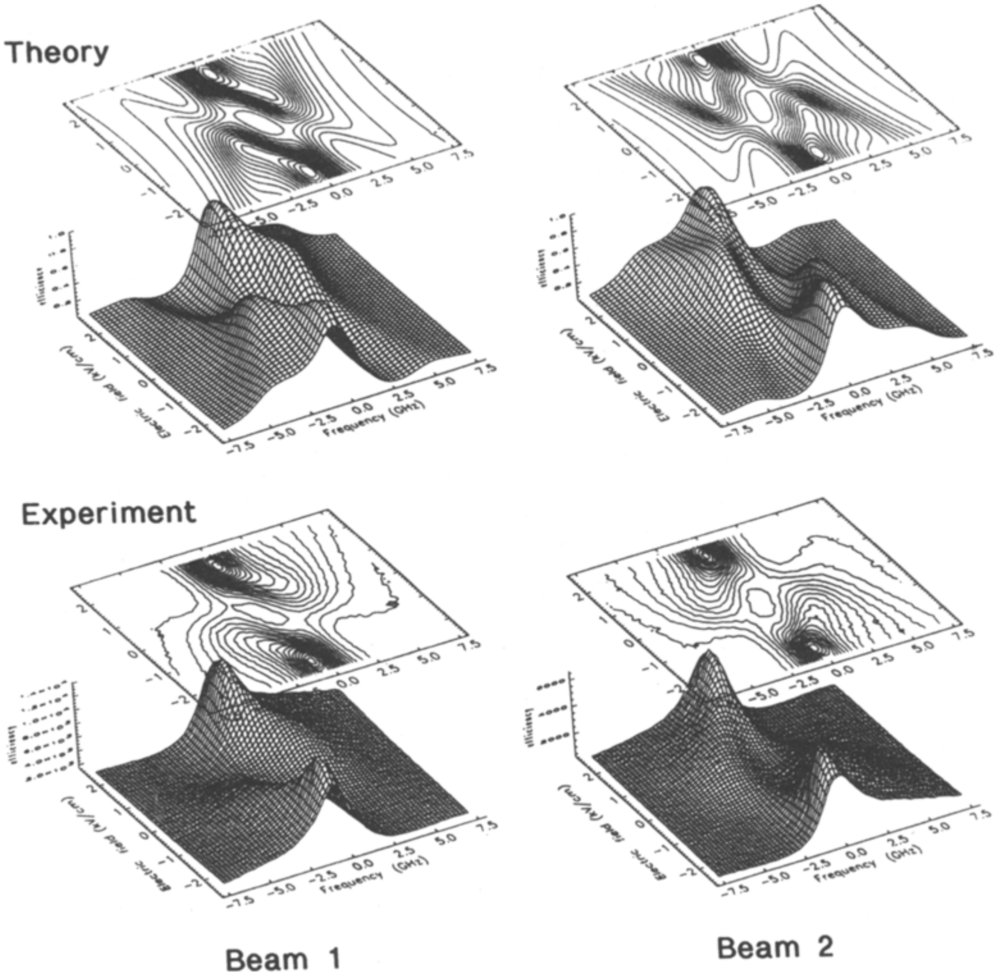

Figure 3. Comparison of unsmoothed experimental and theoretical data for a chosen spatial phase change of $\pi/2$. The data are plotted as a contour plot and a surface plot with arbitrary units of holographic efficiency.

Figure 3 also illustrates the dependence of the holographic efficiency on reconstruction beam (Holliday *et al* 1990). To understand the origin of the dependence of holographic efficiency on the choice of reconstruction beam consider equation (1). The beams are incident upon the gratings at angles to the normal of θ and $-\theta$ respectively, where $|\theta|$ is typically 10°. The effective spatial phase difference between the holographic gratings is therefore reversed when the reconstruction beam is changed. For one beam the spatial phase difference is $\varphi_2 - \varphi_1$, whilst for the other it is $\varphi_1 - \varphi_2$.

The absorption coefficient and the propagation constant modulations are physical properties of the gratings and are therefore not influenced by the choice of reconstruction beam. When the gratings are asymmetrically displaced from each other, however, they appear different to radiation incident from opposite sides of the normal. When the gratings are displaced symmetrically, for $\varphi_2 - \varphi_1 = 0$ or π, they appear

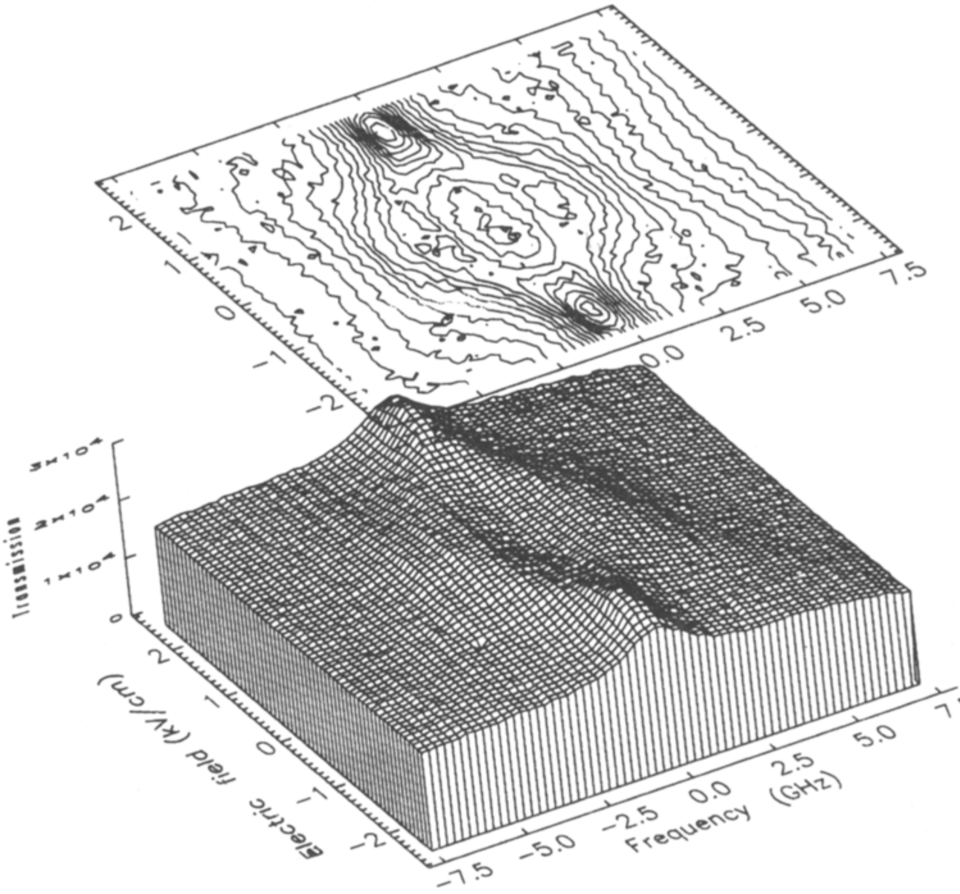

Figure 4. Experimental plot of transmission strength (arbitrary units) measured concurrently with the holographic efficiency plot shown in figure 3 for a spatial phase change of $\pi/2$ and with beam 1.

identical to each beam and the diffraction efficiency as a function of electric field and frequency is equivalent regardless of which is chosen for reconstruction (Holliday *et al* 1990).

A typical transmission signal is shown in figure 4. Interference effects arise from the properties of the holographic gratings and are therefore manifested only in the holographic signal. The transmission signal therefore has the same symmetry properties regardless of reconstruction beam or grating properties. The large background signal associated with the recording of spectral holes in transmission is clearly seen.

A full understanding of the reconstruction properties of, and the interference effects between, holographic gratings is essential to the development of multiple image storing holographic systems (Renn *et al* 1989) and for the realisation of a working 'molecular computer' (Wild *et al* 1990). Such a device combines data or images, stored holographically using spectral hole-burning, by overlapping split Stark components of holes. Addition and subtraction may be performed by selecting conditions which result in constructive or destructive interference respectively.

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