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Photochemically accumulated photon echoes and Stark effect

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We investigate photon echoes stimulated by diffraction from photochemically accumulated gratings in the presence of an externally applied electric field. In the experiment carried out with picosecond laser pulses using a chlorin-doped polyvinylbutyral film at 1.6 K we observe an oscillatory behavior of the diffracted signal intensity depending on the strength of the applied electric field. The unusual oscillating behavior is due to a linear Stark effect, which influences the frequency domain profile of the photochemically accumulated gratings.

The burning and probing of narrow spectral holes with a tunable single-frequency laser in combination with the application of a variable external electric field yields detailed information about the impurity centers [1–4] and also has potential applications in parallel optical data processing and holography [5–8].

The effects associated with the application of an external electric field manifest themselves not only in connection with a single, narrow spectral hole, but also in the case of more complicated spectral structures generated by hole burning such as spectral gratings and time and space domain holograms. In this paper we present the first experiments where the influence of an external electric field upon a grating-like spectral structure is studied using a photochemically accumulated stimulated photon echo technique.

The molecular hole burning system we investigate is a polymer (polyvinylbutyral) doped with chlorin molecules at a concentration of 10^{-3} M. The sample is prepared by pressing a 100 μm thick film between two heated glass plates covered with semitransparent conducting electrodes.

At a temperature of 1.6 K the maximum of the 0–0 absorption of chlorin occurs at 633 nm. Prior to the laser irradiation the optical density of the sample in the used spectral range of 632–636 nm is 1.0–1.5.

In the experiment we use a picosecond dye laser (DCM) synchronously pumped by a frequency-doubled mode locked Nd : YAG laser. The dye laser output pulses have a duration of 5 ps and a repetition rate of 76 MHz. The spectral width of the dye laser pulses of 1.5 Å (120 GHz) is much less than the inhomogeneous bandwidth of our hole burning system (5.6 THz), which allows us to make several different exposures on one sample by choosing different wavelengths of the dye laser. The time resolved echoes are detected by the method of time correlated single photon counting. The apparatus has been described earlier in refs. [9–11].

Our setup is shown in fig. 1. The incoming dye laser beam is expanded by a telescope and split into a reference and an object beam. The reference and the object beams overlap at an angle of 10° on the sample, which is positioned inside a liquid-He immersion cryostat. The intensity of each beam is 10 mW/cm^2. The delay of the object beam with respect to the reference beam is varied between different exposures in the range of 0–500 ps. During each exposure the optical path difference between the two beams is kept constant with a precision of a fraction of a wavelength.

As the first step we open both beams and let them illuminate the sample simultaneously for 10

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s. The photochemical hole burning alters the absorption of the medium and results in a spatial and spectral domain grating-like structure or hologram [12] with a frequency domain period of the grating inversely proportional to the time delay between the reference and the object pulses.

After the exposure a shutter closes the object beam and the reference beam is attenuated by insertion of a neutral density filter. It has been shown [12] that illumination of the spatial and spectral domain gratings with the reference beam alone gives rise to a time-delayed diffracted signal that propagates in the direction of the object beam and which is an analogue of an accumulated stimulated photon echo.

As next step we change the value of the externally applied electric field and pass the attenuated reference beam through the sample. We record the time integrated echo signal intensity by focussing the diffracted signal through a pinhole and by using a microchannel plate in combination with the electronic photon counting system.

The electric field dependence of the diffracted signal obtained from a single narrow spectral hole in the same material shows a monotonic decreasing behavior due to the linear Stark effect [4]. However, our experiment which is carried out with time delayed picosecond pulses reveals a completely different behavior. The results are summarized in fig. 2. The horizontal axis of the plot corresponds to the strength of the electric field applied across the sample.The vertical axis represents the logarithm of the intensity of the diffracted signal and is expressed in arbitrary units.

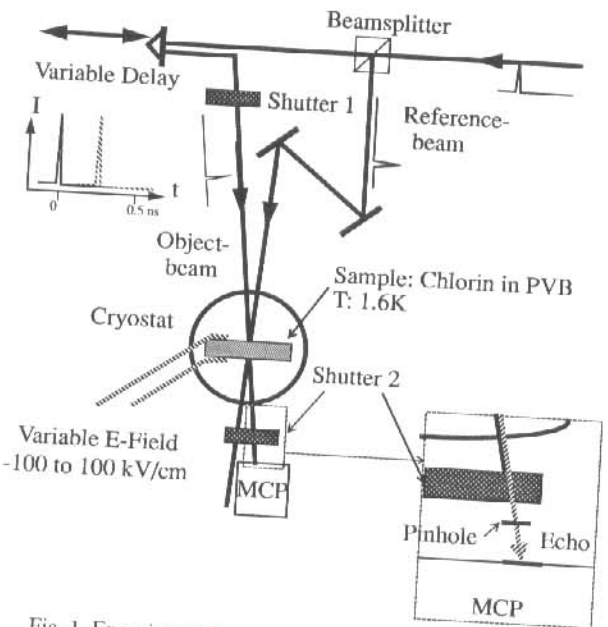

Fig. 1. Experimental setup to store time domain holograms.

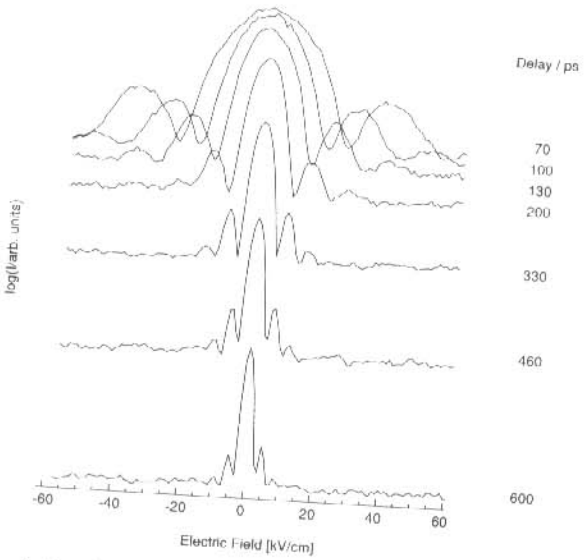

Fig. 2. Results of Stark field dependent echo signals stored with different pulse delays.

Our experimental data are understood by analyzing the spectral response (the Fourier transform of a pulse pair with a time delay) in terms of a set of spectral holes. A spectral profile of a hole undergoes splitting and broadening in a Stark field [4]. At a certain value of the splitting the contrast in the spectral domain vanishes and so also the intensity of the detected echo signal. With further increase of the Stark splitting of the holes the contrast in the spectral domain increases again and the photon echo signal reappears.

The sensitivity of the echo signal with respect to the applied Stark field increases with the resolution in the spectral domain. This is clearly seen from our experimental data: for the shortest pulse delay (70 ps) we observe a signal oscillation with

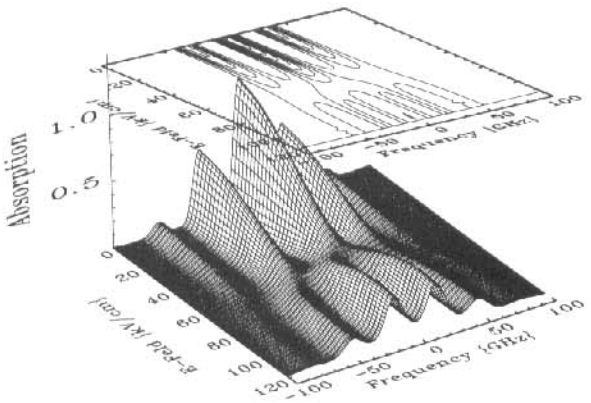

Fig. 3. Simulated data for the E-field dependency of the spectral grating: delay 30 ps.

a minimum at ± 25.0 kV/cm and a second maximum at ± 37.5 kV/cm with an intensity of 10% of the maximum at 0 V/cm. With increasing delay the resolution in the spectral domain is increased and the signal oscillation period along the electric field axes decreases to ± 2.50 kV/cm for the maximum delay of 600 ps.

Figure 3 presents a simulation of the pulse response in the spectral domain corresponding to our experimental situation. We have assumed a delay value of 30 ps and have taken the known Stark splitting parameters of the chlorin molecule [4]. One can clearly see that at a certain value of the electric field strength the spectral pattern tends to smear out. At larger field values the periodic structure reappears.

In conclusion, we have shown that the application of an external electric field affects not only single narrow spectral holes, but also changes the diffraction properties of complicated spectral structures over broad spectral intervals. In principle, this allows the investigation of fine spectral effects caused by an external electric field without using single-frequency tunable lasers. A similar approach could be applied to the effects caused by other types of external fields such as a magnetic field, hydrostatic and uniaxial pressure.

References

[1] M. Maier, Appl. Phys. B 41 (1986) 73.
[2] F.A. Burkhalter, G.W. Suter, U.P. Wild, V.D. Samoilenko, N.V. Rasumova and R.I. Personov, Chem. Phys. Lett. 94 (1983) 483.
[3] U.P. Wild, S.E. Bucher and F.A. Burkhalter, Appl. Opt. 24 (1985) 1526.
[4] A.J. Meixner, A. Renn, S.E. Bucher and U.P. Wild, J. Phys. Chem. 90 (1986) 6777.
[5] A. Renn and U.P. Wild, Appl. Opt. 26 (1987) 4040.
[6] A.J. Meixner, A. Renn and U.P. Wild, J. Chem. Phys. 91 (1989) 6728.
[7] U.P. Wild, A. Renn, C. De Caro and St. Bernet, Appl. Opt. 29 (1990) 4329.
[8] U.P. Wild and A. Renn, J. Mol. Electron. 7 (1991) 1.
[9] U.P. Wild, A.R. Holzwarth and H.P. Good, Rev. Sci. Instr. 48 (1977) 1621.
[10] S. Canonica and U.P. Wild, Anal. Instr. 4 (1985) 331.
[11] E.C. Meister, U.P. Wild, P. Klein-Bölting and A.R. Holzwarth, Rev. Sci. Instr. 59 (1988) 499.
[12] A. Rebane, R.K. Kaarli, P.M. Saari, A. Anijalg and K. Timpmann, Opt. Commun. 47 (1983) 173.