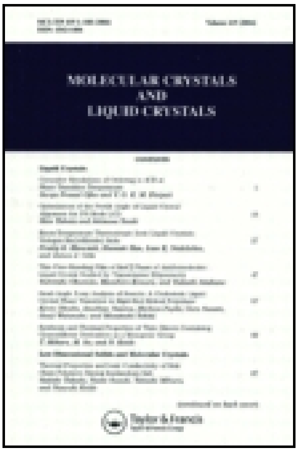

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TIME-RESOLVED NONLINEAR OPTICAL STUDIES OF A FABRY-PEROT CAVITY CONTAINING PTS-POLYDIACETYLENE

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Abstract We have made off-resonant nonlinear optical measurements of PTS polydiacetylene in a Fabry-Perot cavity. The measurement was made by a pump-probe technique using 90 ps pulses at 1.06 μm wavelength. The change in transmission at specific bias points on the F-P fringe, was measured as a function of time delay between the pump and probe. Large (~50%) changes in transmission with a pulse-with-limited response time, were observed at low optical intensities. The magnitude of n_2 as measured (~10^{-5} cm^2/MW) and the negative sign are consistent with our Z-scan measurement of PTS at this wavelength. The results of the measurement on the F-P cavity clearly demonstrate possibilities of applications of PTS in ultrafast (sub-picosecond) all-optical switching and logic operations using this device geometry. This is the first time that all-optical switching with time resolution has been demonstrated for an organic material.

INTRODUCTION

Polydiacetylenes are conjugated polymers with electronic and optical properties dominated by delocalized π-electrons confined in one-dimension. These materials are important in nonlinear optics because of their large off-resonant third order nonlinearity and subpicosecond response time[1,2]. These properties make them suitable for ultrafast all-optical device applications. In this report we present experimental results of a time resolved measurement of a nonlinear Fabry-Perot cavity containing PTS-polydiacetylene. These measurements have provided informations about the magnitude and sign of the off-resonant nonlinearity of PTS-polydiacetylene, with picosecond time resolution. Z-scan measurements are presented as an alternative technique to measure the sign and magnitude of the nonlinearities. Finally a Kramers-Kronig (K-K) calculation is used to understand the mechanisms involved in the off-resonant nonlinear behavior.

NONLINEAR FABRY-PEROT CAVITY

Nonlinear Fabry-Perot cavities are among the most promising devices to control light with light, because of logic as well as memory capabilities[3, 4]. We have constructed a Fabry-Perot cavity containing PTS-polydiacetylene and used it in the experimental arrangement shown in Fig.1. This was a pump-probe experiment used to obtain information about the dynamics of the nonlinear process for off-resonant excitation. A piezoelectric drive was used to control the cavity length. The pump and probe were collinear with a controllable delay between them. A Nd:YAG laser at 1064 nm wavelength and with pulses of 90 ps duration (82 MHz) was used to perform this experiment. A pockels modulator was used to control the average power in the beam.

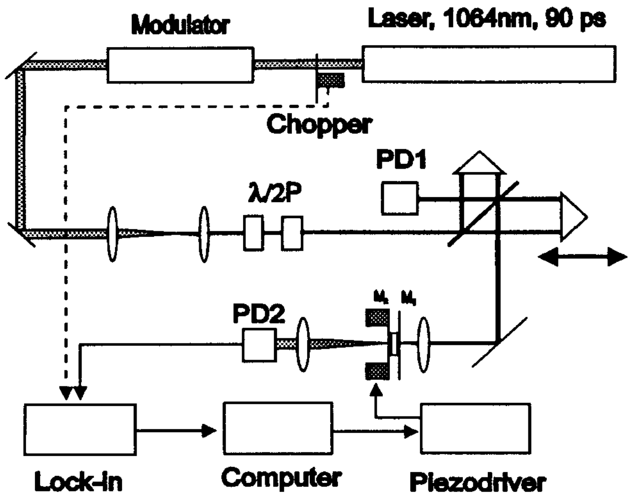

Fig.1 Experimental set-up for the Fabry-Perot measurement. P, polarizer; PD's, photodiodes; M's, mirrors.

Fig.2 presents the change in transmission through the cavity when the distance between the mirrors in the cavity is increased at a low input intensity. The result is the standard Airy function. However, since this is a nonlinear cavity, the transmission peak shifts as the intensity is increased through time overlap of input pulses. The direction of such a shift provides the sign of n_2. We observed a negative sign of n_2 for PTS. Measurement of the change in transmission as a function of time delay indicates changes larger than 50%. Increases as well as decreases in transmission depending on different bias points were measured, with a response time that was pulse-width-limited. The insets in Fig.2 show the change in transmission as a function of time delay. The ultrafast response time that was observed clearly shows that the nonlinearity did not involve any slow process such as thermal. These results demonstrate all-optical switching at a picosecond time scale. This is the first time such switching has been demonstrated for an organic material.

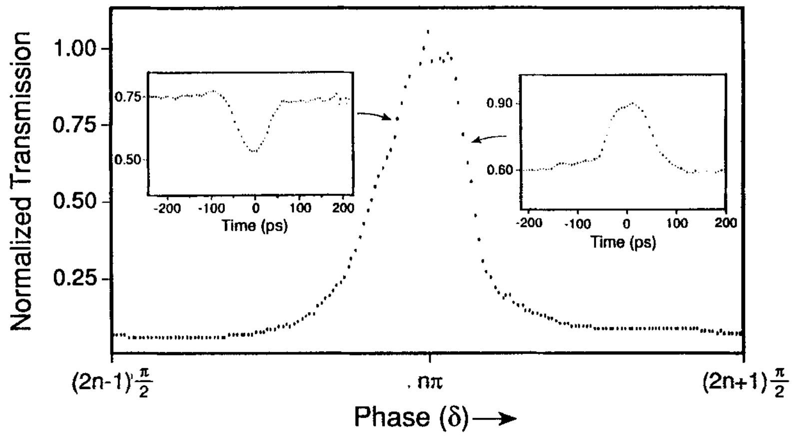

Fig.2 One of the Febry-Perot fringes. The insets show the changes in transmision as a function of time delay while the cavity is tuned to the indicated bias points.

Z-SCAN

The z-scan measurement can be used to measure nonlinear dispersive and absorptive properties[5]. We have performed z-scan measurements at various peak intensities while keeping the average power fixed and thus eliminating any significant thermal contribution. The measurement was performed at 1.06μm wavelength and more details of this measurement has been recently reported[6]. Figure 3 provides the z-scan data for nonlinear refractive index only and was obtained by separating the two photon contribution. The data in Fig.3 correspond to a peak intensity of 20 MW/cm^2. The line shape of the z-scan shows that the sign of n_2 is negative.

The measured differences in the peak to valley normalized transmissions (ΔT_{p-v}) in z-scans, after separating the two photon contribution, are shown in Fig.4. These data were recorded for different peak intensities while keeping the average power fixed. The results show that (ΔT_{p-v}) increases linearly with peak power for the same average power. The magnitude of n_2 that we have obtained from these data is 1.5 (±0.5)x10^{-5} cm^2/MW. The z-scan measurement with an open aperture led to a two photon absorption coefficient (α_2) of 65 (±12) cm/GW, consistent with a previous report[8].

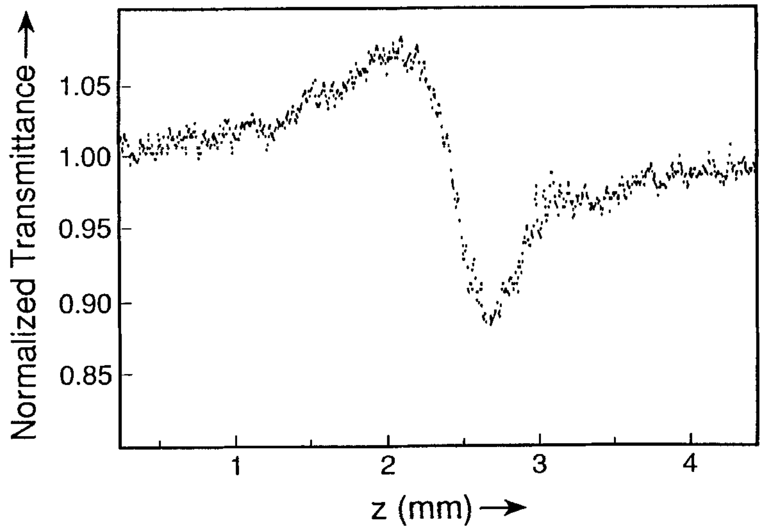

Fig.3 Z-scan data for PTS at a specific peak intensity.

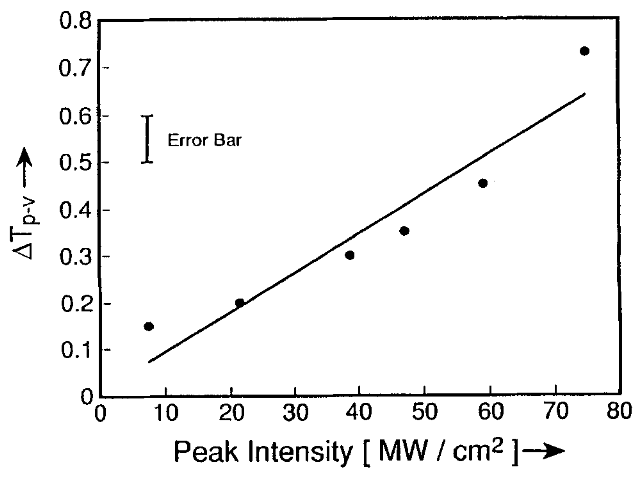

Fig.4 The difference between normalized peak and valley transmission (ΔT_{p-v}) as a function of peak intensity. The average power in the beam was kept constant as the peak power was increased.

The magnitudes of nonlinearities reported above are consistent with our previous reports on nonlinear interferometric measurements in single crystal waveguides[7]. The sign of the nonlinear refractive index is consistent with our Mach-Zehnder interferometric measurement which was not discussed in this report. The negative sign was also previously observed in several other off-resonant measurements on polydiacetylenes[10,11,12]. This sign can be explained using a three level model and also a Kramers-Kronig transformation utilizing the available photoinduced bleaching and absorption data[6]. Since twice the pump frequency used in this experiment lies above the first two photon transition, a negative sign in the real part of $\chi^{(3)}$ is expected. In addition the induced transmission at the exciton peak due to off-resonant pumping also contributes significantly to the magnitude and negative sign of n_2.

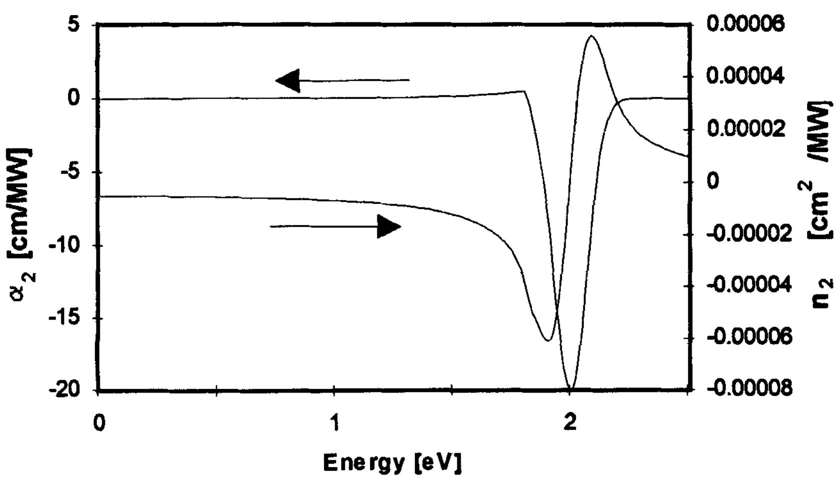

Fig.5 Photoinduced absorption and bleaching data for off-resonant pumping and the calcualted nonlinear refractive index. The magnitude of bleaching is estimated using ref.9.

The Kramers-Kronig relation is utilized to relate the nonlinear refractive index to the nonlinear absorption coefficient through equation 1. Using the available data on photoinduced absorption[8] and bleaching[9] of the exciton, for off-resonant pumping, the nonlinear refractive index has been calculated. The magnitude of the induced transmission (bleaching of exciton) has been estimated using ref.9. The results are presented in Fig. 5. The calculated sign and magnitude of n_2 are consistent with the experimental results.

$$n_2(E) = \frac{ch}{2\pi^2}\int_0^\infty \frac{\alpha_2(E')}{(E')^2 - E^2}\,dE' \qquad (1)$$

SUMMARY

We have performed picosecond time resolved measurements of a nonlinear Fabry-Perot cavity containing a PTS crystal. Large changes in transmission (50% modulation) was observed at low input intensities, with a pulse-width-limited response time. The sign of n_2 is negative which is consistent with our z-scan measurements. This is the first time that all-optical switching with time resolution has been demonstrated for a organic material.

REFERENCES

1. D. S. Chemla and J. Zyss (Eds.) Nonlinear Optical Properties of organic molecules and crystals (Academic, Orlando, 1987).
2. J. Messier, F. Kajzar, P. Prasad, and D. Ulrich (Eds) Nonlinear optical Effects in Organic Polymers (Kluwer, Boston, 1988).
3. H. M. Gibbs Optical Bistability (Academic, Orlando, 1985).
4. J. Jewell et al Appl. Phys. Lett. 46 918 (1985).
5. M. Sheik-Bahae, A. A. Said, T-H Wei, D. J. Hagan and E. W. Van Stryland, IEEE J. Quant. Electron., 26 760 (1990).
6. R. Quintero-Torres and M. Thakur, SPIE Proc., 2025, July (1993).
7. M. Thakur and D. M. Krol, Appl. Phys. Lett., 56 1213 (1990).
8. M. Liquime and J. P. Hermann, Chem. Phys., 26 431 (1977).
9. B. I. Greene, J. F. Muller, J. Orenstein, D. H. Rapkine, S. Schmitt-Rink, and M. Thakur, Phys. Rev. Lett. 61, 325 (1988).
10. F. Kajzar, L. Ledoux and J. Zyss, Phys. Rev., A, 36 2210(1987).
11. G.M. Carter, Y.J. Chen and S.K. Tripathy, Appl. Phys. Lett., 43 891 (1983).
12. M. Sinclair, D. McBranch, D. Moses and A.J. Heeger, Appl. Phys. Lett., 53 2374 (1988).