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# OPTICAL FREE INDUCTION DECAY BY EXTRACAVITY LASER-FREQUENCY SWITCHING

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In this letter we demonstrate the possibility of performing optical free induction decay by *extra*-cavity laser-frequency switching. The method is used to study optical dephasing of pentacene in p-terphenyl and the results are found to be consistent with those previously obtained by the intra-cavity switching technique. The prospects of extra-cavity frequency switching as a tool for studying optical dephasing on a subnanosecond time-scale and in the uv spectral range are emphasized.

Recently Brewer and Genack [1] introduced a very useful technique for performing optical coherent transient experiments with a tunable cw dye-laser. This technique employs, as originally proposed by Yariv [2], switching of a laser frequency by applying a voltage across an electro-optic crystal (EOC) placed *inside* the cavity of a dye-laser. The insertion of this EOC inside the laser cavity however increases substantially the laser-threshold which may prohibit usage of the technique when less efficient dyes than Rhodamine-6G are to be pumped. In this letter we show that optical coherent transient experiments may also be done by *extra*-cavity laser-frequency switching. We demonstrate the technique by study of the optical free induction decay (OFID) of pentacene in p-terphenyl.

In the extra-cavity frequency switching technique we employ the fact that a laser beam traversing an EOC crystal is frequency shifted when the EOC crystal is exposed to a voltage ramp [3]. A voltage ramp ($\mathrm{d}V/\mathrm{d}t$) across the EOC gives rise to a proportional negative change in refractive index by the Pockels effect and thus changes the optical pathlength ($\rho$). By the Doppler effect this induces a shift ($\Delta\upsilon$) of the frequency of the laser light traversing the EOC with an amount of:

$$\Delta\upsilon = -\frac{1}{\lambda}\frac{\mathrm{d}\rho}{dt} \sim +\frac{\mathrm{d}V}{dt}. \qquad (1)$$

Note that the extra-cavity induced shift is proportional to the voltage *ramp*, while the intra-cavity induced shift (averaged over times longer than the round-trip time) is proportional to the voltage *amplitude*. Given the order of magnitude of the Pockels coefficients and dimensions of commercially available EOC's [3], one can estimate the shift to be on the order of 1–1.5 GHz with a ramp of 1kV/ns. The ultimately obtainable shift is determined by the EOC transit time of about 50 ps.

As we were interested in high frequency shifts we needed a pulse-generator capable of delivering pulses of high amplitude and fast rise time into the capacitive load formed by the EOC and its connecting cable. The pulse-generator should also have a low output impedance in order to minimize the dissipation of the ensuing high currents. As such a pulse-generator was not at our disposal we had to build one. Utilizing a 20-stage cascaded circuit of normal npn BC 547 transistors (or its precursor BC 107), of which the first one was fired by a 1-10 V positive going trigger pulse, we succeeded in building a simple low cost all-transistorized high voltage pulser (HVP) of small dimensions (fig. 1). Keeping the connecting cable to the EOC (Inrad model 621-042, capacitance 80 pf) as short as possible (~5 cm), the output capability of the HVP was a 2–3 kV (adjustable) negative pulse with 15 ns risetime and a very favourable repetition rate ($\lesssim$ 5 kHz), which ensures signal averaging to be easily done. (During the experiments we used a repetition rate of 1 kHz to guarantee safe operation). Because of its small dimensions the HVP could easily be shielded from the detection electronics. Applying this high voltage ramp (–3 kV/15 ns) across

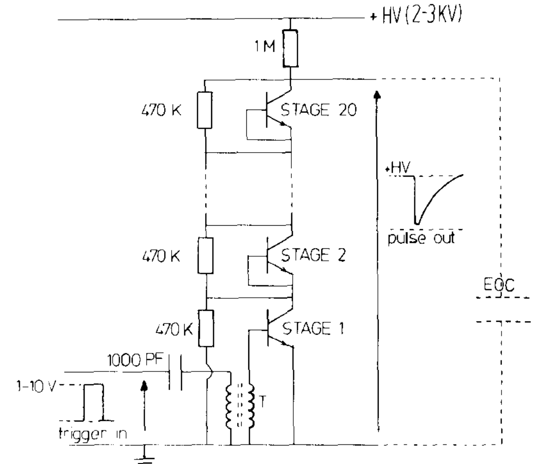


Fig. 1. Schematic of HVP: 20-stage cascade of npn transistors BC 547 or BC 107. H.V.: high voltage. T is a (1:1) transformer, consisting of a ferrite core with four windings of thin copper wire for both primary and secondary coil. Trigger input: amplitude + 1-10 V, rep. rate $\lesssim$ 5kHz. Pulse output: neg. amplitude 2–3 kV (adjustable), risetime 10–15 ns (depending on load capacitance, 20–80 pF), falltime 100 $\mu$s, duration ~ 1$\mu$s.

our EOC (half-wavelength voltage 300 V) a shift of about 350 MHz to lower frequency after one passage of the laser beam through the crystal is expected. Such a shift was indeed observed by studying the single mode output of a Spectra-Physics model 850 Rh6G dye laser, pumped by a Spectra-Physics model 165 $Ar^+$ laser, on a confocal Fabry-Pérot interferometer (Tropel model 240).

The OFID experiment on pentacene in p-terphenyl was performed with the experimental arrangement shown in fig. 2. The laser beam was sent three time through the EOC in order to increase the shift to about 1 GHz. Consequently this procedure made the frequency shift in three stages, but, as the reflecting mirrors were placed very close to the EOC (within 2.5 cm), the 1 GHz shift is effectively attained within about 500 ps. Next the beam passed through a pinhole (1.5 mm) and the sample, which was contained in a temperature variable liquid helium cryostat (Oxford Instruments model MD4A). The outcoming signal in the forward direction was focused with a 10 cm lens on a BPY 77 silicon photo-diode (risetime 200 ps). A Philips model PM 3400 sampling oscilloscope in conjunction with a Northern Scientific model 560 CAT was used to monitor and average the transient, that finally was plotted on a XY recorder.

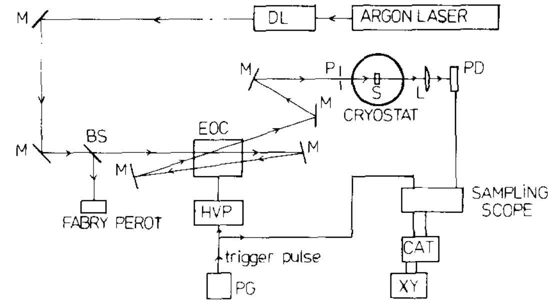


Fig. 2. Experimental set-up for OFID: D.L.: dye laser, M: mirror, B.S.: beamsplitter, P: pinhole, S: sample, L: lens, PD: photodiode, PG: Pulsegenerator. In the figure the size of the angles between incoming and reflected beams within the EOC have been exagerated for clarity.

To test the method we performed the OFID experiment on the $O_1$ (5921.6Å) and $O_2$ (5920.1Å) sites of pentacene in a p-terphenyl host crystal at 1.5 K, as these were recently investigated in our laboratory [4] by the intracavity switching method.

We indeed observed a 1 GHz OFID beat signal at the highest attainable ramp voltage. In fig. 3 we present the OFID signal of the $O_2$ site that was obtained at a ramp voltage of 2.5 kV and a laserpower of 2.8 mW/mm$^2$. Also shown is the semilogarithmic plot of the beat amplitude as a function of time, together with the straight line obtained by a least square fitting to the experimental points. As is shown in the figure

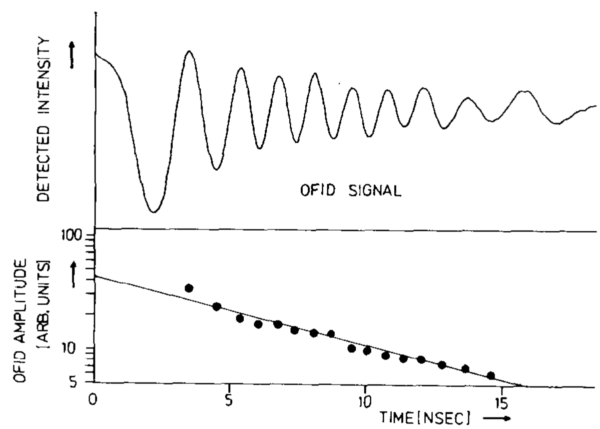


Fig. 3. Upper part: OFID signal of the $O_2$ site of pentacene in p-terphenyl at 1.5 K; laserpower: 2.8 mW/mm$^2$ Lower part: Semi-logarithmic plot of experimental beat amplitudes against time. The drawn line is a least square fitting to the experimental points.

the beat period walks off in the course of time, indicating ramp non-linearities. This of course does not influence the results. Irregularities in the peak amplitudes, caused by some residual electrical disturbances, however, may affect the results. We note that the OFID decay time of 7.5 ± 1.0 ns calculated from fig. 3 is, within the experimental errors, in accordance with previously obtained results by the intra-cavity technique [4]. This shows that the extra-cavity switching technique is a reliable method to study optical coherent transients. With the progress made in high power electrical switching techniques, for example by means of light-controlled silicon switches [5], this method should be useful in performing OFID experiments in the subnano-second regime. In this way one would be able to compare narrowband with broadband coherent excitation techniques (psec photon echoes [6]) on this time scale. The additional advantage of having the EOC placed outside the optical cavity seems also very promising for OFID experiments in the uv spectral range. Even the hitherto most powerful dye laser [7] is not likely to be capable of handling a power-consuming EOC inside the optical cavity together with a frequency doubling crystal. With the EOC placed outside the cavity the full uv power is available.

*Note added in proof.* After this paper was written de Voe and Brewer reported subnanosecond frequency switching using a traveling wave electro-optic phase modulator (Phys. Rev. Lett. 40 (1978) 862).

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