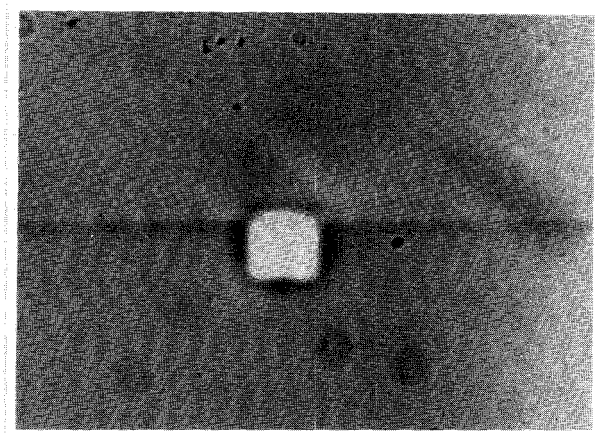
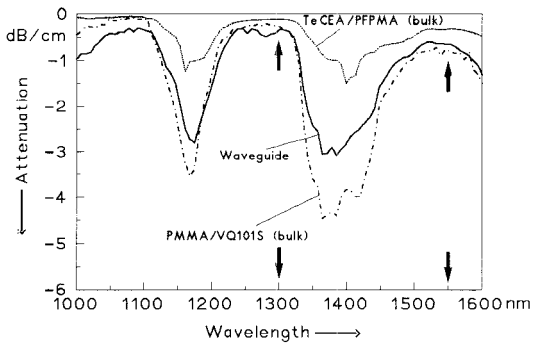


CWE4 Fig. 2. Polished endface of a buried polymeric straight waveguide, core material: PFPMA/TeCEA.



CWE4 Fig. 3. Attenuation of bulk PMMA, PFPMA/TeCEA, PFPMA/TeCEA/PMMA-integrated waveguide in dB/cm.

transparent in the desired wavelength ranges around 1300 and 1550 nm. It must be easily tunable with respect to the refractive index and not dissolving the substrate microstructures. Two complete new, UV-curable material systems have been developed to work with PMMA as substrate and cover material. The first material system is EGDMA or EGDMA-D14 (ethylenglycoledimethacrylate or fully deuterated version) tuned by TFPMA (tetrafluorinepropylmethacrylate), whereby TFPMA lowers the refractive index of EGDMA. The deuterated version shows low losses at 1300 nm (<0.1 dB/cm), however, unacceptable losses at 1550 nm (~1.5 dB/cm). The other material system is PFPMA/TeCEA (pentalfluorophenylmethacrylate / tetrafluorinepropylmethacrylate).[4,5] PFPMA lowers the refractive index of TeCEA. This polymer system shows low transmission loss at 1300 nm (~0.1 dB/cm) as well as at 1550 nm (~0.3 dB/cm) (Fig. 3).

The best achieved intrinsic waveguide losses for straight waveguides with PMMA as substrate material are 0.2 dB/cm at 1300 nm (~0.1 dB/cm bulk absorption, ~0.1 dB/cm scattering loss) and 0.7 dB/cm at 1550 nm (~0.3 dB/cm bulk absorption, ~0.3 dB/cm cladding absorption, ~0.1 dB/cm scattering loss). The spectral behaviour of the insertion loss of a straight PFPMA/TeCEA-waveguide is shown in Fig. 3 in comparison to the bulk substrate material PMMA and the bulk waveguide core material PFPMA/TeCEA. Due to weak waveguiding the optical field penetrates relatively deep into the cladding and, therefore, the influence of the PMMA substrate on the absorption spectrum becomes obvious, in particular at 1550 nm. The waveguides are designed to yield high overlap with standard telecommunication fibres. Coupling efficiencies better than 98% are achieved.

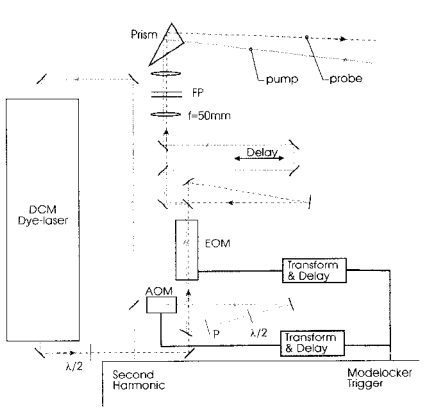
1. A. Neyer, T. Knoche, L. Müller, Fabrication of low-loss polymer waveguides using injection moulding technology, Electron. Lett. **29**, 399–400 (1993).
2. T. G. Ryan, S. Allen, *et al.* (ICI, UK), R. Cush, *et al.* (GEC Marconi, UK), "Polymeric waveguide devices," MOC/GRIN'93, Kawasaki, Japan, pp. 72–75.
3. L. Baraldi, M. T. Gale, J. Söchtig, Y. Oppliger, J. M. Stauffer, J. Meissner, "Fabrication of single-mode ridge waveguide structures by embossing into polycarbonate on glass," in *Proceedings of ECOC'92*, Berlin, Germany, pp. 561–564.
4. D. Bosc, B. Boutevin, Y. Pietrasanta, A. Rousseau, French Patent No. 87 16671, Nov. 20, 1987.
5. F. Hrabák, H. Pivcová, J. Polym. Sci.: Polym. Chem. Ed. **10**, 3125 (1972).

CWE5 **0930**

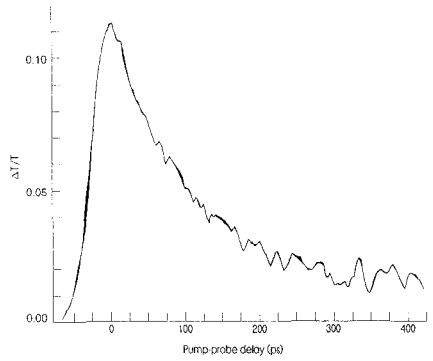
Characterisation of the intensity dependent refractive index of nonlinear polymers in solution

B. J. Offrein, H. A. G. M. van Wolferen, H. J. W. M. Hoekstra, A. Driessen, Th. J. A. Popma, *Mesa Research Institute, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands*

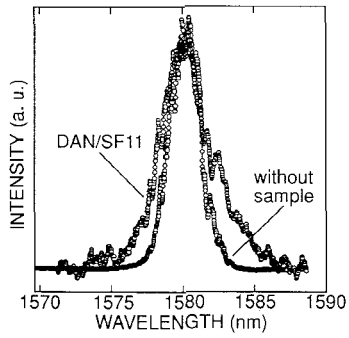
Polymers form, besides semiconductors, a class of interesting optically nonlinear (ONL) materials. The nonresonant nonlinearity of π-conjugated polymer systems can be relatively large and the nonlinear response time is often in the ps-regime, even in the resonant case. We use a Fabry–Perot to determine both the real and imaginary part of the intensity dependent refractive index of ONL polymers in solution. The etalon converts changes in the refractive index and absorption coefficient of the material inside the resonator (the polymer solution) into a transmission change.[1] It is useful to characterise the nonlinearity of the material in solution as it may be difficult to grow crystals or spin waveguides of newly developed nonlinear polymer systems. The Fabry–Perot is inserted in a pump–probe type setup, as shown in Fig. 1. The change of the optical parameters is induced by a 527-nm 35-ps pump pulse and detected with a 660-nm 5-ps probe pulse. The experimental setup consists of a cw-mode-locked Nd:YLF-laser, a SHG-unit, and a synchronously pumped DCM-dye-laser. In order to overcome thermal effects the peak power to average power ratio is increased by decreasing the repetition rate of both the pump and probe pulses from 76 MHz to about 20 kHz with the help of an electro-optic modulator. Additionally, the pump is modulated with an acousto-optic modulator to block every second pulse resulting in a pump repetition rate of about 10 kHz. The pump induced 10 kHz modulation in the probe is detected with a lock-in amplifier. Both pump and probe pulses are focused in the Fabry–Perot etalon containing the nonlinear material. For the nonlinear material we use the DANS side chain polymer,[2] as synthesised by Akzo, dissolved in cyclopentanone (10 mass% DANS). The 527 nm pump pulse excites the polymer reso-



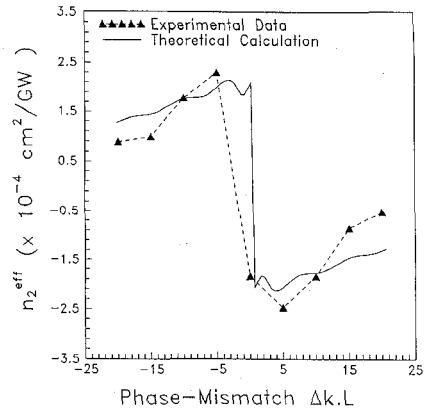
CWE5 Fig. 1. Pump–probe setup. FP: Fabry–Perot, AOM: acousto-optic modulator, EOM: electro-optic modulator, P: polariser.



CWE5 Fig. 2. Relative probe transmission change as a function of the probe pulse time delay.



CWE6 Fig. 1. Spectral broadening observed in a DAN fiber.



CWE6 Fig. 3. The sign and magnitude change of n_2 by tuning the phase mismatch, based on Z-scan measurements in a bulk DAN single crystal.

nantly. An absorption saturation was measured at the probe wavelength of 660 nm corresponding to $\mathrm{Im}(\chi^{(3)}(\omega_s;\omega_p, -\omega_p,\omega_s))$ is $-1.0 \pm 0.5 \times 10^{-9}$ esu. By varying the pump–probe pulse delay (Fig. 2), the relaxation time was determined from an exponential fit to be 100 ± 5ps (1/e-value). From experiments we deduce that the minimum detectable refractive index and absorption coefficient changes are of the order of 5×10^{-8} and 10^{-2} dB/cm respectively.

1. J. E. Ehrlich, D. T. Neilson, D. J. Goodwill, A. C. Walker, C. Johnston, W. Sibbett, J. Opt. Soc. Am. B. **10**, 492 (1993).
2. M. B. Marques, G. Assanto, G. I. Stegeman, G. R. Möhlmann, E. W. P. Erdhuisen, W. H. G. Horsthuis, Appl. Phys. Lett. **58**, 2613 (1991).

CWE6 **0945**

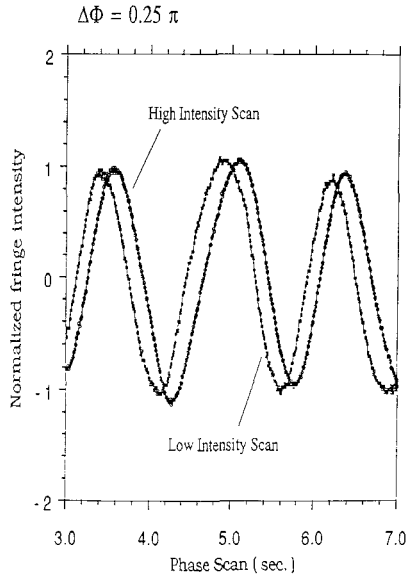
Very large cubic nonlinearities in quadratic molecular media induced by cascading second order processes

P. Vidakovic, J. Zyss, D. Kim,* W. Toruellas,* G. Stegeman,* W. Moerner,** R. Twieg,** G. Bjorklund,** *FRANCE TELECOM/CNET/PAB, 196, Avenue Henri Ravera, 92220 Bagneux, France*

Cascading has been recently proposed[1] and demonstrated in KTP-based structures as a means to achieve significant optically induced phase shifts because of coupled second-harmonic and down-conversion processes in a mismatched configuration. In view of record high quadratic nonlinear susceptibilities of organic materials,[2] we have extended this approach to molecular crystals, which had been previously developed toward such applications as three-wave mixing and optical parametric oscillation. Their potential toward outstanding performance in the realm of $\chi^{(3)}$ processes based on cascading[3] and eventually outperforming other type of materials and nonlinear effects toward all-optical signal processing is being currently established as follows.

Due to their high effective d coefficients (d ≈ 27 pm/V at 1.064 μm) and their availability in both single crystalline and crystalline cored fiber formats with adequately oriented dielectric axis,[4] 4-(N,dimethylamino)-3-acetamidonitrobenzene (DAN) crystals[5] have been selected.

$$n_2^{eff}\ (1320\ \mathrm{nm}) = -8\times10^{-4}\ (\mathrm{cm}^2/\mathrm{GW})$$



CWE6 Fig. 2. The shift of fringes observed at high intensity in a DAN fiber.

Four types of experiments have been performed,[3] namely:

- Third-harmonic generation on 1-μm-thick PMMA films doped by DAN (at a concentration of 7×10^{20} cm^{-3}) at 1.904-μm and 6-ns pulse duration.
- Spectral broadening as large as 1.75 at 1.55 μm of 600-fs pulses in a 2.4-mm-long DAN/SF15 fiber (see Fig. 1).
- Z-scan in a 0.77-mm-thick DAN single crystal (at 1064 μm and 35 ps pulse duration) at various orientations corresponding to different angular departures away from phase-matching and therefore different dephasing conditions between the injected fundamental and the generated harmonic wave (Fig. 2).
- High precision pulse modulated Mach Zender interferometry at 1.32 μm with 90 ps pulses on a DAN/SF1 fiber adapted for an efficient Cerenkov type second harmonic generation.

These experiments provide consistent evidence of the following features:

- Confirmation of the cascading origin of the observed cubic phenomena, which cannot be accounted for by a mere oriented gas summation of molecular electronic cubic hyperpolarisabilities:γ such a scheme has been checked to lead to lower efficiencies by at least two orders of magnitude as compared to our experimental observations. Indeed, $\chi^{(3)}$(molecular) is only 4 times larger than $\chi^{(3)}$(silica glass), while the corresponding value for the crystal is 100 times that of the silica glass.
- Influence of the phase-matching conditions that allow to monitor the magnitude and the sign of n_2 (verified on the bulk DAN single crystal) fully confirmed theoretical predictions for. In the case of fibers, whenever second harmonic was absent in crystal cored fibers, the nonlinear phase shift was negligible.
- Extremely high n_2 nearly 10^{-12} cm^2/W (see Fig. 3), which are being currently further challenged by using more efficient structures such as NPP.[6]

In conclusion, the by now solidly established virtues of cascading and the high quadratic nonlinearities of phase-matchable organic crystalline structures may be uniquely combined so as to possibly rejuvenate the field of cubic nonlinear optics and related all-optical signal processing applications. The limits between quadratic and cubic nonlinear optics are somewhat blurred whereas high cubic nonlinearities can be achieved in a parametric configuration without resorting to electronic resonance enhancement.

*C.R.E.O.L., *University of Central Florida, Orlando, FL 32826*
**I. B. M. Almaden Research Center, 650 Harry Road, Almaden, CA 95120*

1. G. I. Stegeman, M. Sheik-Bahae, E. Van Stryland, G. Assanto, Optics Lett. **18**, 13 (1993).
2. *Nonlinear Optical Properties of Organic Molecules and Crystals*, D. S. Chemla, J. Zyss, Eds., (Academic Press, London, 1987).
3. D. Y. Kim, W. Toruellas, J. Kang, C. Bosshard, G. Stegeman, W. E. Moerner, R. Twig, G. Bjorklund, submitted to Optics Lett.