transient 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 (ethylene glycol dimethacrylate or fully deuterated version) tuned by TFPMA (tetrafluoroproplmethacrylate), 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 (pentafluorophenylmethacrylate / tetrafluoroproplmethacrylate). 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 (~0.2 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.

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 and applications as three-wave mixing and nonlinear optical parametric configuration without resorting to electronic resonance enhancement is currently established as follows.

Due to their high effective d coefficients (\(d = 27 \text{ pm/V at } 1.064 \mu\text{m}\)) and their availability in both single crystalline and crystalline cored fiber formats with adequately oriented dielectric axis,\(^4\) extremely large cubic nonlinearities in quadratic molecular media induced by cascading second order processes

\[ n_{\text{off}}(1320 \text{ nm}) = -6 \times 10^{-4} \text{ (cm}^2/\text{GW)} \]

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 cubic hyperpolarizabilities 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 \(10^{-12} \text{ cm}^2/\text{W}\) (see Fig. 3), which are being currently further challenged by using more efficient structures such as NPP.

In conclusion, the by now solidly established virtues of cascading and the high quadratic nonlinearities of phase-matching 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.

**References**