1.9 Cascaded Second-Order Nonlinearities in Organic and Inorganic Single Crystals

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Large, nonresonant optical Kerr-like nonlinearities are a basic requirement for all-optical switching and related applications. Besides the nonlinearity which is proportional to $\chi^{(3)}$ there is another important contribution that occurs only in noncentrosymmetric materials. This contribution induces a nonlinear phase shift in nearly phase-matched second-harmonic generation interactions and other parametric processes. Recently the potential for all-optical switching was realized and large nonlinear phase shifts due to cascading (second-harmonic generation (SHG) and difference-frequency mixing (DFM)) were observed in several materials. Last year, we have shown both theoretically and experimentally that the combined processes of optical rectification (OR) and the linear electro-optic (EO) effect can also lead to an effective n_2 in noncentrosymmetric materials without the need for phase-matching.

We have now extended our theoretical understanding by taking into account local field effects and boundary conditions. This lead to an even better correspondence between theory and experiment. We have carried out calculations to evaluate the performance of noncentro–symmetric single crystals for both types of cascading (SHG and DFM, OR and EO). Our estimations clearly show that the potential of organics is superior as compared to inorganic materials. Currently measured values of cascaded n_2 are still lower than the ones of the best materials based on $\chi^{(3)}$. This could easily be changed with either better organic crystals or poled polymers. Special emphasis will have to be paid to nonlinear effects near the interesting telecommunication wavelengths at around 1.3 μm and 1.5 μm.