1.8 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. Such effects were first observed in high-power pulsed second-harmonic generation experiments. Recently the potential for all-optical switching was realized and large nonlinear phase shifts due to cascading were observed for the first time in potassium titanyl phosphate (KTP) waveguides. Unexpectedly large nonlinear refractive index changes observed in fibers of 4-(N,N-dimethylamino)-3-acetamidonitrobenzene (DAN) could also be explained by cascaded nonlinearities.

We have now demonstrated that there exist other combined processes of second-order nonlinear optical effects that give rise to a large effective nonlinear refractive index n_2 without the need for phase-matching. In particular, we have shown both theoretically and experimentally that the combined processes of optical rectification and the linear electro-optic effect lead to an effective n_2 in noncentrosymmetric materials.

Our investigations showed that this new contribution to n_2 is of comparable magnitude to the well-known one which is due to second-harmonic generation and difference frequency mixing. An increase in n_2 of more than a factor of three was measured for $KNbO_3$ when compared with $KTaO_3$. A further advantage is that there is no loss of the incident beam since we have no depletion of the fundamental as it occurs in the case of large nonlinear phase shift generated in nearly phase-matched second-harmonic generation interactions. It is also interesting to note that this contribution is always positive. For a general propagation direction in a noncentrosymmetric material both cascaded effects are important for induced nonlinear phase-shifts. It is, however, possible to find special propagation directions where one contribution is dominant as we have shown for $KNbO_3$.