

1.5 Development of Electro-Optic Co-Crystals with Ideal Chromophoric Orientation and Large Second-Order Nonlinearities

(M.-S. Wong, F. Pan and Ch. Bosshard, in collaboration with V. Gramlich, Laboratory for Crystallography, ETH Zurich)

A series of novel merocyanine derivatives was designed and synthesized for the development of useful and efficient electro-optic crystals. Although a large proportion of them (5 out of the 8) are SHG active in the Kurtz and Perry powder test, the signals are relatively small, that is, of the order of the urea standard indicating a non-optimized orientation.

Therefore, the hydrogen bond induced self-assembly approach was employed to align the merocyanine dye within the co-crystal lattice (see also 1.4). With this approach, we have developed and demonstrated for the first time that highly nonlinear optically active as well as orientationally optimized crystalline materials for second-order nonlinear optics can be prepared by a proper design of hydrogen-bonded molecular aggregates. The two co-crystals, 4-{2-[1-(2-hydroxyethyl)-4-pyridylidene]-ethylidene}-cyclo-hexa-2,5-dien-1-one • methyl 2,4-dihydroxy-benzoate, **Mero-MDB** and 4-{2-[1-(2-hydroxy-ethyl)-4-pyridylidene]ethylidene}-cyclohexa-2,5-dien-1-one • 2,4-dihydroxypropiophenone, **Mero-DPP** (Fig.3) are potentially two of the best crystalline materials for electro-optic applications because of the perfect chromophoric orientation, the very large macroscopic nonlinear response, and the greatly improved crystal properties. As an example the melting point of the co-crystal **Mero-MDB** is 184.6 °C vs 151.6 °C for **Mero** and 118.9 °C for **MDB**. Furthermore, 4-{2-[1-(2-hydroxyethyl)-4-pyridylidene]-ethylidene}-cyclohexa-2,5-dien-1-one • 2,4-dihydroxyacetophenone, **Mero-DAP** co-crystal (Fig. 3) is a very interesting and attractive candidate for the application in optical parametric oscillators since it combines very large macroscopic susceptibilities with an ideal chromophoric orientation for frequency-conversion and good crystal properties.

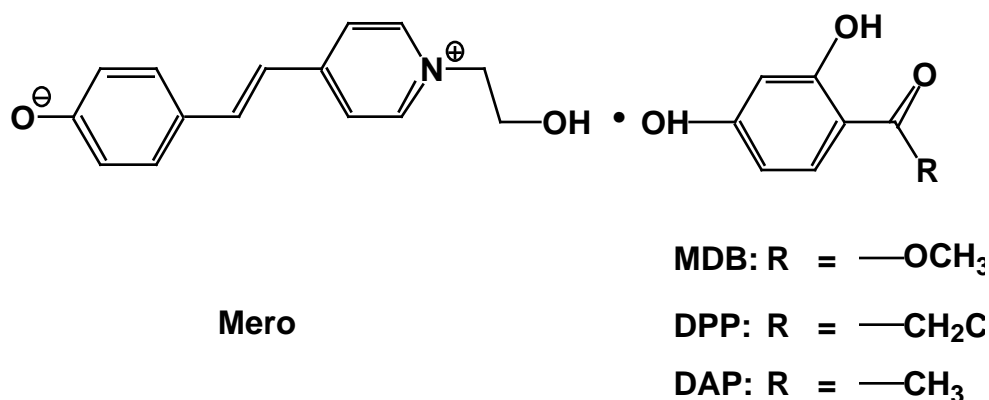


Fig. 3 Molecular structures of the chromophores forming the co-crystals.