

1.7 Nonlinear Optical Investigations of Organic Crystals

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Since, in spite of their moderate quality, the first DAST (4'-dimethylamino-N-methyl-4-stilbazolium tosylate) crystals grown in our group, turned out to be very promising for nonlinear optical applications, much time has been devoted both to growing new larger crystals of better quality and to synthesising other structure-like charge-transfer molecules.

The progress made in growing higher quality DAST crystals allowed a refinement of our results which led to the confirmation of the values obtained earlier : the salt has very large nonlinear optical susceptibilities, the effective coefficient for phase matching being more than one order of magnitude larger than the corresponding value for LiNbO_3 at 1542 nm. Thanks to the large size DAST crystal ($15 \times 15 \times 3 \text{ mm}^3$) grown recently we are able to investigate directly the electro-optic properties of the crystal.

From a large number of benzaldehyde phenylhydrazone derivatives synthesised during the last year, DANPH (4-dimethylaminobenzaldehyde-4-nitrophenylhydrazone) showed the most promising microscopic hyperpolarisabilities, measured successively by EFISH and HRS. Crystal structure determination revealed a very interesting molecular packing, the charge-transfer axis of the molecules forming an angle of about 50 degree with the polar axis, rendering the future crystal quite interesting for second harmonic generation.

With the successful growth of a monocrystal in a non-centrosymmetric form, the first experiments to investigate the optical properties of the crystal have been performed leading to the following values of the indices of refraction $n_1=2.07$, $n_2=2.06$, $n_3=1.44$ at 1064 nm, emphasising the large anisotropy of the crystal. Using the traditional Maker-Fringe method, the effective nonlinear susceptibility for phase matching has been measured giving a value comparable to DAST. Bulk crystals are in the process of being cut and polished to enable a refinement of the obtained values and a direct measurement of the second harmonic under phase-matching conditions.