1.6 Induction of Non-Centrosymmetric Packing of Guest-Host Organic Ionic Complexes

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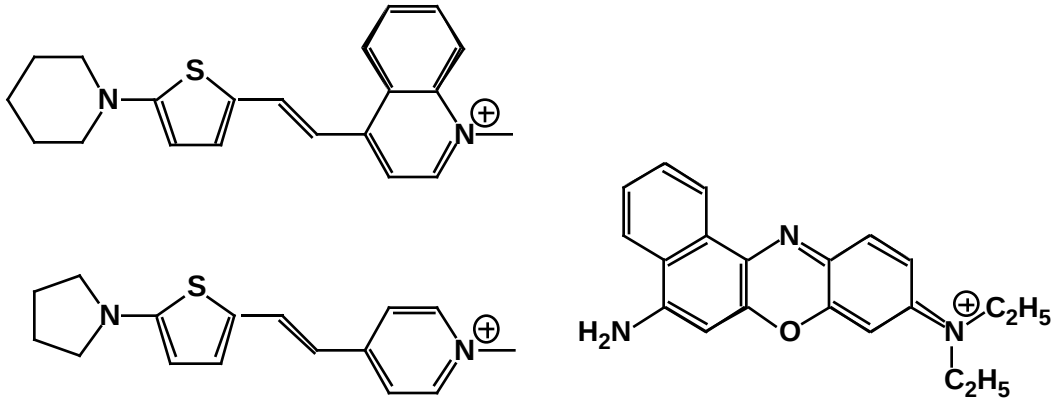
The organic ionic salt 4'-(dimethylamino)-*N*-methyl-4-stilbazolium tosylate, **DAST** exhibits very large second-order nonlinearities. Going along this line, a new series of highly hyperpolarizable stilbazolium cations, DAST analogues (Fig. 4a) , which contained either quinoline or thiophene rings, was designed and synthesized. Different types of counteranions were used to induce a guest-host ionic complex formation and the non-centrosymmetric packing. 47 % of them showed a positive SHG activity in the Kurtz and Perry powder test but the signals were generally small.



(a) (b)

Fig. 4 Molecular structure of highly hyperpolarizable stilbazolium cations (a) and nile blue cations (b).

In addition, nile blue cation (Fig. 4b) is a highly hyperpolarizable chromophore with a strong charge-transfer transition, λ_{ICT} at 633 nm but has never been incorporated into a crystal lattice for second-order nonlinear optics. As a result, the guest-host complex approach was employed to align the nonlinear optically active nile blue cations. Again, the probability of forming acentric crystals was found to be rather high (3 out of the 7) but the orientation was not optimized for second-order optical nonlinear effects.