1.13 Functionalized Materials with All-Carbon Backbones for Nonlinear Optics

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In contrast to second-order non-linear optical effects, there is hardly any intuitive assessment on which type of molecules are suitable for third-order nonlinear optics. We performed third-order nonlinear optical measurements and deduced new structure-property relationships on a series of functionalized tetraethynylethenes (Fig. 5) that provide further insight into routes leading to the desired optical nonlinearities.

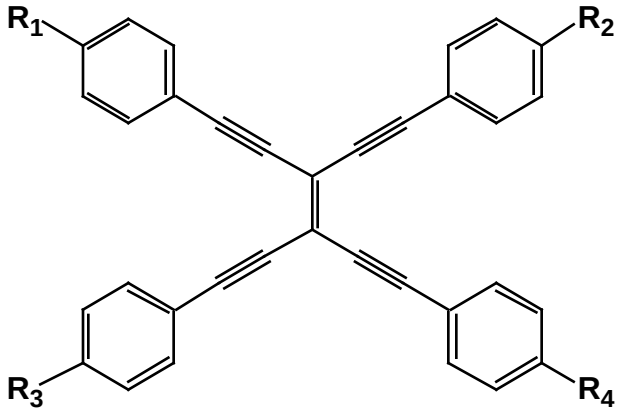

Fig. 5 All carbon backbone of the tetraethynylethenes. R_1-R_4 can be different donors as well as acceptors.

For the first time, the influence of donors and acceptors as well as of full two-dimensional conjugation on the third-order non linearity in one single class of materials is clearly demonstrated. We could show, that the nonlinear optical properties can be explained to a large extent by the possible conjugation paths and the strengths of donors and acceptors. The results obtained in this investigation should lead to materials with even further increased third-order nonlinearity. Moreover, a number of these compounds are of additional interest since they can be polymerised easily, thus facilitating the fabrication of thin films.

This class of materials is also interesting for second-order nonlinear optics. The direction of the dipole moment can strongly deviate from the vector part of the first - order hyper–polarizibility β in our two-dimensional molecules. In addition, off - diagonal tensor elements can be significantly larger than the diagonal components, as could be deduced from first EFISH measurements. Comparisons with computer simulations will give us a better understanding of the nonlinear optical behaviour of this new class of two-dimensional molecules.