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Non-linear optical properties of selected organic systems: high accuracy Density Functional Theory calculations

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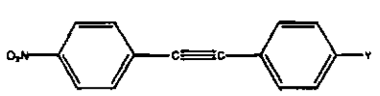
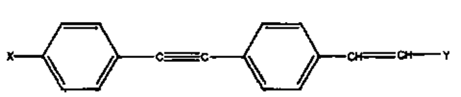
Electro optic materials have been extensively researched over the past few decades due to their potential applications in photonic technologies including all-optical switching and data processing. Our objective is to calculate the nonlinear optical properties of a range of three novel organic molecular systems. The approach is based on the concept of charge transfer (CT) between donor and acceptor through the spacer. In this research work, the first hyperpolarizability values (β) are calculated using the Density Functional Theory (DFT) method B3LYP, with the 6-31g(d) basis set of Gaussian98W software. The designing of systems with high charge transfer is key to this part, as intra molecular charge transfer between donor and acceptor will lead to a very large value of β. The first hyperpolarizability values (β) for the molecular system-1, a_1-a_3 and b_1–b_6 of system-2 have increased due to the substitution on nitrogen of NH_2 group (Figure 1). However the substitution on nitrogen may affect the planarity deviation of the molecules. The addition of a double bond to the spacer, which will enhance the conjugation, leads to larger hyperpolarizability values (β). But the consecutive double bond of the spacer will lead to the distortion of the molecule and it will disturb the charge transfer between the donor and the acceptor (Table 1).

System 1



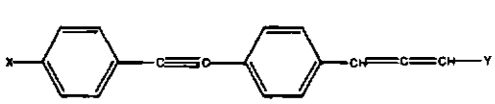
a_1 Y = NH_2 a_2 Y = $NH(CH_3)$
a_3 Y = $N(CH_3)_2$ a_4 Y = OCH_3

System 2



b_1 - Y = NH_2 b_2 - Y = $NH(CH_3)$ b_3 - Y = $N(CH_3)_2$ b_4 - Y = $N(C_2H_5)_2$ b_5 - Y = $N(C_3H_7)_2$ b_6 - Y = $N(C_4H_9)_2$ b_7 - Y = $NH(CH_3)$ b_8 - Y = $N(CH_3)_2$ (b_1.b_6)- X = NO_2 b_7-X = CN b_8 - X = CHO

System 3



c_1 - Y = NH_2 c_2 - Y = $NH(CH_3)$ c_3 - Y = $N(CH_3)_2$ c_1 - X = CN (c_2.c_3) - X = NO_2

Fig. 1: Designed organic systems

Table 1: Hyperpolarizability values of systems

Molecule	$\beta_{tot} \times 10^{-30}$ **(esu)**	**Molecule**	$\beta_{tot} \times 10^{-30}$ **(esu)**	**Molecule**	$\beta_{tot} \times 10^{-30}$ **(esu)**	**Molecule**	$\beta_{tot} \times 10^{-30}$ **(esu)**
a_1	161.534	b_1	210.241	b_5	324.229	c_1	56.917
a_2	190.699	b_2	256.303	b_6	331.355	c_2	138.357
a_3	271.249	b_3	289.407	b_7	138.122	c_3	143.622
a_4	114.084	b_4	307.763	b_8	193.791		

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