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## Second-order nonlinearities of anionic 3-dicyanomethylen-5,5-dimethyle-1-[2-(4-hydroxyphenyl)ethenyl]-cyclohexene

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### Abstract

Due to the connecting one  $\pi$ -conjugated group with the two backside donor and acceptor groups, anionic 3-dicyanomethylen-5,5-dimethyle-1-[2-(4-hydroxyphenyl)ethenyl]-cyclohexene (**1**) might possess nonlinear optical (NLO) properties. To estimate the potential for second-order NLO behaviour; the electric dipole moments and dispersion-free dipole polarizabilities and first hyperpolarizabilities have been determined by density functional theory (DFT) at B3LYP/6-31++G(*d*, *p*) level. Quantum mechanical calculations using time-dependent Hartree-Fock (TDHF) procedure have been utilized to evaluate frequency-dependent second-order nonlinearities of **1**. The one-photon absorption (OPA) characterization of **1** has been theoretically obtained by means of configuration interaction (CI) with 6-31G basis set. Our theoretical results on the maximum OPA wavelength, second-order susceptibilities and corresponding microscopic NLO responses are accorded with the previous experimental observations of the investigated compound. The highest occupied molecular orbitals, the lowest unoccupied molecular orbitals and the HOMO-LUMO band gaps for **1** have been also examined by DFT/B3LYP method.

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**Keywords:** First hyperpolarizability; Dipole polarizability; NLO; one-photon absorption; time-dependent Hartree-Fock; density functional theory

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## 1. Introduction

Organic materials exhibiting second-order optical nonlinearity have been considered very attractive for numerous applications during the last 20 years, such as high speed electro-optic modulation, field detection, frequency conversion [1-2] and terahertz wave generation and detection [3]. Conventional organic nonlinear optical (NLO) chromophores are based on a long  $\pi$ -conjugated bridge that connects electron donor and electron acceptor groups and which, therefore, display a large dipole moment [1-2,4-6]. To achieve a macroscopic second-order nonlinearity, the polar chromophores are aligned by additional external electric field poling in polymers and by self-assembly based on specific intermolecular interactions in crystals. However, the tendency for highly polar chromophores towards antiparallel dipole-dipole aggregation often leads to reduced poling efficiency in a polymer system [7] and to centrosymmetric arrangement of molecules in the crystalline state [8]. Quantum mechanical calculations have helped to identify polyene and thiophene based chromophores with the highest nonlinearity [9]. Spraul *et al.* [10] have reported the synthesis of heterocyclic bridged triarylamine chromophores with high hyperpolarizability  $\beta$ . Ring locked phenyltetraene-bridged chromophores [11] and analogues in general exhibit high  $\beta$  values and electro-optic (EO) devices derived from them have shown commercial promise. Due to their high ground state dipole moment induced head to tail interactions, noncentrosymmetric alignment remains a challenge. The rigid isophorone bridge also causes crystallization and solubility problems. Usage of bulky protected donor groups, attachment of bulky groups to the acceptor are methods sought for minimizing these interactions. It has been also recognized that the addition of aromatic rings in the donor may impose a quinoidal bonding pattern and help to prevent unfavorable organization of the chromophores [12].

On the basis of the molecular structure connected one  $\pi$ -conjugated group with the two backside donor and acceptor groups, one could expect that the title compound (Fig. 1) may show second-order NLO behaviour. In this study, our research interest has been focused on the theoretical investigation of second-order NLO properties of anionic 3-dicyanomethylen-5,5-dimethyle-1-[2-(4-hydroxyphenyl)ethenyl]-cyclohexene (**1**). Therefore, we have evaluated second-order NLO responses using density functional theory (DFT) method on electric dipole moments, static dipole polarizabilities and first hyperpolarizabilities. We have also computed dynamic values of dipole polarizabilities, second-order hyperpolarizabilities and susceptibilities utilizing ab-initio time-dependent Hartree-Fock (TDHF) procedure.  $\chi^{(2)}$  (second-order susceptibility) and  $\beta$  values of **1** obtained theoretically are compared with its experimental findings and also measurement results of a few similar compounds in the literature. In addition to NLO properties, the HOMO-LUMO energies and gaps of outer molecular orbitals and also the electronic transition wavelengths of the lowest lying transitions, respectively, for **1** have been calculated by DFT/B3LYP and CI (configuration interaction) methods.

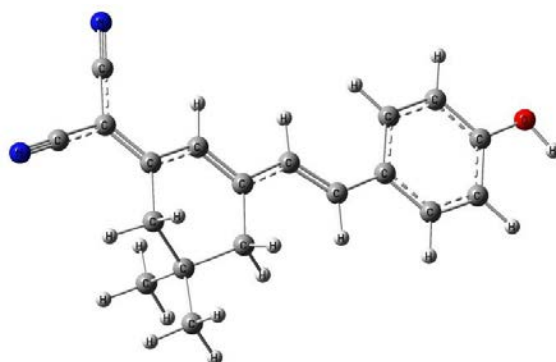


Fig. 1. Chemical structure of compound **1**.

## 2. Theoretical Calculations

The theoretical computations involve the determination of electric dipole moments, dispersion-free and frequency-dependent dipole polarizability and first hyperpolarizability tensor components. The molecular geometry of **1** has been firstly optimized. The geometry optimization has been followed by the calculations of electric dipole moments, static dipole polarizabilities and first static hyperpolarizabilities.  $\mu$ , dispersion-free  $\alpha$  (dipole polarizability) and  $\beta$  have been calculated using the finite field (FF) scheme [13]. The 6-31++G(*d, p*) polarized and diffused basis set was found adequate for obtaining reliable hyperpolarizability values. The hyperpolarizabilities of most molecules are sensitive to the description of the tails of the wave function and so high-order diffuse and polarization functions are required in the basis set to determine convergence of the property [14]. To avoid the problem of basis set completeness, various basis sets (including polarized and diffuse functions) have been implemented by Kenawi *et al.* [15], at the Hartree-Fock and density functional levels of theory, to calculate the hyperpolarizabilities of diclofenac sodium. The results obtained [15] show the importance of including the *p* and *d* polarized functions in the double split valence set for the best description of the static dipole hyperpolarizabilities of diclofenac sodium. Since hyperpolarizabilities are derivatives of the molecular energy with respect to the strength of the applied electric field, their theoretically calculated values may be sensitive to basis set features. Consequently, various basis sets including polarized and diffuse functions could be implemented to calculate the hyperpolarizabilities. One expects the basis set 6-31++G(*d, p*) to yield molecular property values of near-Hartree-Fock quality. 6-31++G(*d, p*) basis set has been also employed to compute the electric dipole moments, static dipole polarizabilities and first static hyperpolarizabilities. All geometry optimization,  $\mu$ , static  $\alpha$  and  $\beta$  calculations have been performed by GAUSSIAN03W [16] at DFT/ B3LYP level with 6-31++G(*d, p*) basis set. Systematic investigations have provided adequate evidence for the potential of DFT methods in electric property calculations. We rely on the widely used B3LYP which denotes the hybrid functional [17], a linear combination of the gradient functionals proposed by Becke [18] and by Lee, Yang and Parr [19] together with the Hartree-Fock local Exchange function [20]. B3LYP is stated as one of the sophisticated DFT methods, which are widely used in many fields of quantum chemistry, atomic and molecular physics. The averaged (isotropic) dipole polarizability  $\langle\alpha\rangle$  and the magnitude of  $\beta_{tot}$  (total first static hyperpolarizability) have been calculated using the following expressions, respectively [21-22]:

$$\langle\alpha\rangle = (\alpha_{xx} + \alpha_{yy} + \alpha_{zz})/3 \quad (1)$$

$$\beta_{tot} = [(\beta_{xxx} + \beta_{xyy} + \beta_{zzz})^2 + (\beta_{yyy} + \beta_{yzz} + \beta_{yxx})^2 + (\beta_{zzz} + \beta_{zxx} + \beta_{zyy})^2]^{1/2} \quad (2)$$

Knowledge of the frequency-dependent hyperpolarizabilities is required in order to make a direct comparison with experiment since all experiments involve at least one time-dependent field. This involves solution of the time-dependent Schrödinger equation. Thus, frequency-dependent hyperpolarizabilities are determined from analytic derivative calculations or using the sum-over-states formulation. Frequency-dependent hyperpolarizabilities have been frequently implemented at the self-consistent field level of theory (known as TDHF). With increasing molecular size the electronic hyperpolarizability becomes larger and its frequency-dependence often becomes stronger.  $\alpha(-\omega;\omega)$  and  $\beta(0;\omega,-\omega)$  at  $\omega = 0.04282$  atomic units (a.u.) (*i.e.* at  $\lambda = 1064$  nm wavelength) according to laser frequency used in second-order NLO measurements have been carried out using the TDHF method with 6-31++G(*d, p*) basis set implemented in the GAMESS [23] program. The frequency-dependent  $\beta(0;\omega,-\omega)$  computations at considered  $\omega$  frequency were carried out by optical rectification (OR) group of the TDHF procedure. This method appeared to be a good compromise between accuracy and calculation duration.

Many types of frequency-dependent first hyperpolarizability computations have been discussed in the literature in the form of  $\beta-V$  ( $\beta$  vector) which is the vector part of the first hyperpolarizability. In this work, the  $\beta-V$  value has been calculated using the following expressions:

$$\beta-V = (\beta_x^2 + \beta_y^2 + \beta_z^2)^{1/2} \quad (3)$$

where  $\beta_i (i = x, y, z)$  is given by:

$$\beta_i = (1/3) \sum_{j=x,y,z} (\beta_{ij} + \beta_{ji} + \beta_{jj}) \quad (4)$$

We have also computed the orientationally averaged (isotropic) value of second-order susceptibility  $\langle \chi^{(2)}(-\omega; \omega, 0) \rangle$  which represents the nonlinear interaction of second-order.  $\langle \chi^{(2)}(-\omega; \omega, 0) \rangle$  calculation at  $\omega = 0.04282$  a.u. was carried out by electro-optics Pockels effect (EOPE) group of the TDHF method with 6-31++G(d, p) basis set implemented in the GAMESS [23] program. To calculate all electric dipole moments, hyperpolarizabilities and susceptibilities, the origin of Cartesian coordinate system  $(x, y, z) = (0, 0, 0)$  has been chosen at the center of mass of **1**.

Besides, the transition wavelengths ( $\lambda_{max}$ ) of the lowest lying electronic transitions for **1** have been determined by CIDRT group (configuration interaction with all doubly occupied molecular orbitals from Hartree-Fock reference determinant) of the TDHF method using 6-31G basis set implemented in the GAMESS [23] program. All one-photon absorption (OPA) wavelengths, dynamic  $\alpha$ ,  $\beta$  and  $\langle \chi^{(2)} \rangle$  calculations have been performed on a PC with an Intel (R) core (TM) I7-2630QM operator, 5.8 GB RAM memory and 2 GHz frequency using Linux PC GAMESS version running under Linux Fedora release 11 (Leonidas) environment.

To understand the relationship of NLO properties with the molecular structure; HOMOs, LUMOs and HOMO-LUMO gaps have been also generated by GAUSSIAN03W [16] program at DFT/ B3LYP level with 6-31++G(d, p) basis set. The HOMO-LUMO band gap ( $E_g$ ) could be expressed as follows:

$$E_g = E_{LUMO} - E_{HOMO} \quad (5)$$

### 3. Computational results and discussion

In order to understand the dependence of the electronic absorption properties of **1** on its structure, the vertical transition energies from the ground state to each excited state have been computed, giving OPA, i.e., the UV-Vis spectrum. The calculated wavelengths ( $\lambda_{max}$ ) for the maximum of optical absorption are shown in Fig. 2.  $\lambda_{max}$  values centered at 492.05 nm and 588.11 nm are located in the visible region. The computed  $\lambda_{max}$  values are in good agreement with the experimental results recorded at 452 nm and 563 nm in  $\text{CHCl}_3$  and  $\text{C}_2\text{H}_5\text{OH}$  solvents, respectively, by Ref. [24] (Fig. 2). Suresh *et al.* [25] measured the  $\lambda_{max}$  values for two chromophores in a series of triarylamine-polyene chromophores as 550 nm and 585 nm. The theoretically (588.11 nm) and experimentally (563 nm) obtained  $\lambda_{max}$  values of **1** are quite consistent with the experimental results of a similar series of chromophores reported in Ref. [25]. The most widely used method for calculating properties of electronically excited states is CI. One starts from the Hartree-Fock wave function of the corresponding electronic ground state and performs a CI calculation. In this way, one can obtain a reasonably accurate description of the optical spectrum of the system. The calculated vertical transition energies in Fig. 2 are only slightly larger than the experimental values of **1**. The overestimated excitation energy may originate from the truncated CI method that does not correlate excited states as well as the ground states.

The large nonlinearities of certain organic compounds arise from extended  $\pi$ -conjugated systems, as well as the presence of asymmetrical charge transfer processes. Charge transfer originates from the electron-donating and electron-accepting properties of an aromatic ring substituent. The magnitude of NLO response can be easily altered by changing the donor, acceptor or the  $\pi$ -backbone. It has been established that the NLO properties increase with increasing strength of donor and/or acceptor fragments [26]. The design strategy used by many with success involves the donor and acceptor groups at the terminal positions of a  $\pi$  bridge to create highly polarized molecules which could exhibit large molecular nonlinearity. The effects of certain donors and acceptors and influence of their positions in a certain structure should be investigated and after the inexpensive theoretical approach the targeted synthesis can be carried out. The DFT theory was chosen to compute the static dipole polarizabilities and first hyperpolarizabilities of **1** due to its reliability already discussed [27]. Practically, after optimizing geometries,  $\alpha_{ij}$  and  $\beta_{ijk}$  components were determined using FF theory. As the static dipole polarizability and first hyperpolarizability

values depend on the DFT functional used, we carried out the computation of dispersion-free  $\alpha$  and  $\beta$  at the DFT level using B3LYP method [19]. The  $\beta$  value of the constituent molecules is essential for a structure-based NLO behaviour understanding. Molecules containing conjugated  $\pi$ -electron systems with charge asymmetry usually exhibit large values of  $\beta$  [28]. The higher dipole moment values are associated, in general, with larger projection of  $\beta_{tot}$  quantities. The connection between the electric dipole moments of an organic molecule having donor-acceptor substituents and first hyperpolarizability is widely recognized in the literature [29-30]. Several research groups have tried to identify the molecules with potentially optimal nonlinearities through the Two-Level model. For example, Marder *et al.* [9] used a four-site Hückel Model to examine how each of the Two-Level parameters varies with the electron-donating and electron-accepting abilities of appended substituents. One of the conclusions obtained from their work was that non-zero  $\mu$  value might permit to find non-zero  $\beta$  value. In this study, the static dipole polarizabilities and first static hyperpolarizabilities, respectively, are computed by the numerical first and second derivatives of the electric dipole moments according to the applied field strength in FF approach. Electric dipole moments, selected values of the static dipole polarizabilities and first hyperpolarizabilities computed for **1** are shown in Tables 1-3. There is a strong relationship between the calculated  $\mu$  and  $\beta_{tot}$  values. Therefore, rather high  $\mu$  value (10.877 Debye) of **1** in Table 1 may be responsible for enhancing its  $\beta_{tot}$  value ( $102.070 (\times 10^{-30})$  esu) in Table 3. In addition, the involvements of donor (methyl) and acceptor (cyano) groups attached to  $\pi$ -conjugated system result in obvious enhancement of static  $\langle\alpha\rangle$  and  $\beta_{tot}$  (Tables 2-3).  $\mu$  and  $\beta_{tot}$  values, respectively, in a series of similar chromophores to **1** were computed by Kwon *et al.* [31] as 9.62 Debye and  $79.23 (\times 10^{-30})$  esu for a chromophore with hydrogen *N*-substituent and also as 9.93 Debye and  $80.60 (\times 10^{-30})$  esu for a chromophore with methyl *N*-substituent. Our calculation results for **1** on  $\mu$  and  $\beta_{tot}$  (Tables 1,3) are in reasonable accordance with the values of similar chromophores reported by Ref. [31].

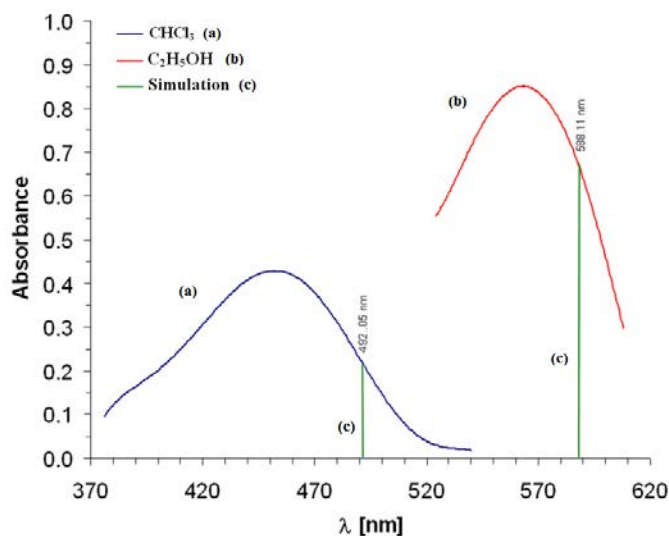


Fig. 2. UV-Vis absorption spectra measured in  $\text{CHCl}_3$  and  $\text{C}_2\text{H}_5\text{OH}$  solvents and theoretical simulations of the compound **1**.

Table 1. The ab-initio calculated electric dipole moment  $\mu$  (Debye) and dipole moment components for **1**.

| $\mu_x$ | $\mu_y$ | $\mu_z$ | $\mu$  |
|---------|---------|---------|--------|
| -8.871  | -6.292  | -0.118  | 10.877 |

Table 2. Some selected components of the static  $\alpha(0;0)$  and  $\langle\alpha\rangle(0;0)$  ( $\times 10^{-24}$  esu) value for compounds **1**.

| $\alpha_{xx}$ | $\alpha_{yy}$ | $\alpha_{zz}$ | $\langle\alpha\rangle$ |
|---------------|---------------|---------------|------------------------|
| 86.270        | 38.296        | 20.825        | 48.464                 |

Table 3. Some selected components of the static  $\beta(0;0,0)$  and  $\beta_{tot}(0;0,0)$  ( $\times 10^{-30}$  esu) value for the compound **1**.

| $\beta_{xxx}$ | $\beta_{xxy}$ | $\beta_{xyy}$ | $\beta_{yyy}$ | $\beta_{xxz}$ | $\beta_{yyz}$ | $\beta_{zzz}$ | $\beta_{yzz}$ | $\beta_{zzx}$ | $\beta_{tot}$ |
|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|
| 105.624       | -4.879        | -3.420        | -0.441        | -1.958        | 0.148         | -0.327        | -0.574        | -0.355        | 102.070       |

In spite of the rapid development of highly advanced experimental techniques on NLO, the theoretical understanding of NLO properties shows that the responses of the molecule to the external application of an electric field are also of much importance. Hyperpolarizability and susceptibility tensors can describe the response of molecules to an external electric field. To compare our theoretical results with that of experimental values in the literature, we have investigated the dispersion behavior of dipole polarizabilities, second-order hyperpolarizabilities and susceptibilities calculated by the TDHF method at  $\omega = 0.04282$  a.u. (see Tables 4-6). TDHF method provided a reasonable compromise between the demands on high efficiency on the level of theory employed and on the accuracy of calculations on one hand and the computational capabilities on the other hand. Kolev *et al.* [24] employed the Kurtz powder technique [32] to measure the second-order susceptibilities and effective hyperpolarizabilities of **1** (with a pulsed Q-switch Nd:YAG laser operating at 1064 nm wavelength and providing 10 ns duration laser pulses). As a result, it was discovered an increase for the effective second-order susceptibilities of **1** from  $14.8 (\times 10^{-9}$  esu) to  $37.2 (\times 10^{-9}$  esu) and for the dynamic second-order hyperpolarizabilities from  $10.5 (\times 10^{-30}$  esu) to  $22.4 (\times 10^{-30}$  esu) during an increase in the electrostatic field strengths up to 1.8 kV/cm and a decrease in the crystallite sizes from 300 nm to 10 nm [24]. In this work, the dynamic second-order hyperpolarizabilities and susceptibilities, respectively, of **1** have been calculated to be  $21.886 (\times 10^{-30}$  esu) and  $31.605 (\times 10^{-9}$  esu), quite consistent magnitudes as compared to the cases of the measurement results reported by Kolev *et al.* [24] ( $\beta = 10.5 (\times 10^{-30}$  esu) –  $22.4 (\times 10^{-30}$  esu) and  $\chi^{(2)} = 14.8 (\times 10^{-9}$  esu) –  $37.2 (\times 10^{-9}$  esu)) (see Tables 5-6).  $\beta$  value for a chromophore in a series of triarylamine polyene chromophores was evaluated by Suresh *et al.* [25] using Hyper Rayleigh Scattering at 1604 nm as  $\beta = 700 (\times 10^{-30}$  esu). The calculated value of  $\beta - V$  at 1064 nm for **1** in Table 5 is about 31 times lower than that of the similar chromophore at different wavelengths reported in Ref. [25]. The second-order hyperpolarizability for a isophrone-type analogue in a series of triene chromophores was measured using electric-field-induced second harmonic (EFISH) experiment by Ermer *et al.* [33] ( $\beta = 114 (\times 10^{-30}$  esu) at  $\lambda = 1907$  nm). Our computation on dynamic  $\beta - V$  for **1** (Table 5) is about factor of 5 lower than the experimental finding obtained by Ermer *et al.* [33] for a rather similar compound. Zhou *et al.* [34] measured the second-order susceptibility value for a chromophore in a series of phenyltetraene-based chromophores to be  $\chi^{(2)} = 200.5 (\times 10^{-9}$  esu) at 1310 nm. The computed result on  $\chi^{(2)}$  of **1** (Table 6) has been obtained to be about 6 times lower than that of the experimental data for a similar chromophore measured by Ref. [34] at different wavelength of incident laser beam.

Table 4. Some selected components of the frequency-dependent  $\alpha(-\omega;\omega)$  and  $\langle\alpha\rangle(-\omega;\omega)$  ( $\times 10^{-24}$  esu) value at  $\omega = 0.04282$  a.u. ( $\lambda = 1064$  nm) for the compounds **1**.

| $\alpha_{xx}$ | $\alpha_{yy}$ | $\alpha_{zz}$ | $\langle\alpha\rangle$ |
|---------------|---------------|---------------|------------------------|
| 52.194        | 23.216        | 6.734         | 27.381                 |

Table 5. Some selected components of the frequency-dependent  $\beta(0; \omega, -\omega)$  and  $\beta - V$  ( $\times 10^{-30}$  esu) value at  $\omega = 0.04282$  a.u. ( $\lambda = 1064$  nm) for the compound **1**.

| $\beta_{xxx}$ | $\beta_{yyy}$ | $\beta_{zzz}$ | $\beta_x$ | $\beta_y$ | $\beta_z$ | $\beta - V$                  |
|---------------|---------------|---------------|-----------|-----------|-----------|------------------------------|
| 39.379        | 0.727         | -0.137        | 21.805    | 1.780     | -0.597    | 21.886<br>(10.5 - 22.4) [24] |

Table 6. Measured effective  $\chi_{\text{exp}}^{(2)}$  (during an increase in the electrostatic field strengths up to 1.8 kV/cm and a decrease in the crystallite sizes from 300 nm to 10 nm) and computed  $\langle \chi^{(2)}(-\omega; \omega, \omega) \rangle$  values at  $\omega = 0.04282$  a.u. ( $\lambda = 1064$  nm) for the compound **1**.

| $\chi_{\text{exp}}^{(2)} (\times 10^{-9} \text{ esu})$ | $\langle \chi^{(2)} \rangle (\times 10^{-9} \text{ esu})$ |
|--------------------------------------------------------|-----------------------------------------------------------|
| (14.8 - 37.2) [24]                                     | 31.605                                                    |

In order to understand the electronic structure of **1**, the calculated HOMO and LUMO orbitals are shown in Fig. 3. The HOMO and LUMO energies and HOMO-LUMO energy gaps of **1** determined by DFT method at B3LYP/6-31++G(d, p) level are listed in Table 7. The DFT theory was chosen to compute the energies of frontier molecular orbitals of our system due to its reliability already discussed [27]. However, the calculations performed by this method are always with underestimated HOMO-LUMO gap. In Fig. 3, HOMO-1 refers to the second highest occupied molecular orbital. It is known that the molecular electronic spectrum is usually caused by the electron transition from the HOMO to the LUMO. The second and third-order NLO responses can be dictated by intramolecular charge-transfer (ICT) excitations involving the HOMO and LUMO orbitals in such a way that larger values of second and third-order hyperpolarizabilities should correspond to lower HOMO-LUMO gaps. Therefore, it could be expected that the investigated compound with rather low HOMO-LUMO gap might have second-order optical nonlinearity with non-zero responses (see Tables 5-7). The HOMO orbital distribution is localized mainly at the hydroxyl-phenyl electron donor group. The LUMO orbital distribution is localized at the dicyanomethylidene electron acceptor group (see Fig.3). The localizations indicate the ICT characteristics of **1**, such that the electron density moves from the electron donor group to the electron acceptor group. The HOMO-1 orbital with a greater energy is more delocalized than the LUMO+1, leading to higher value of band gap (0.21808 a.u.).

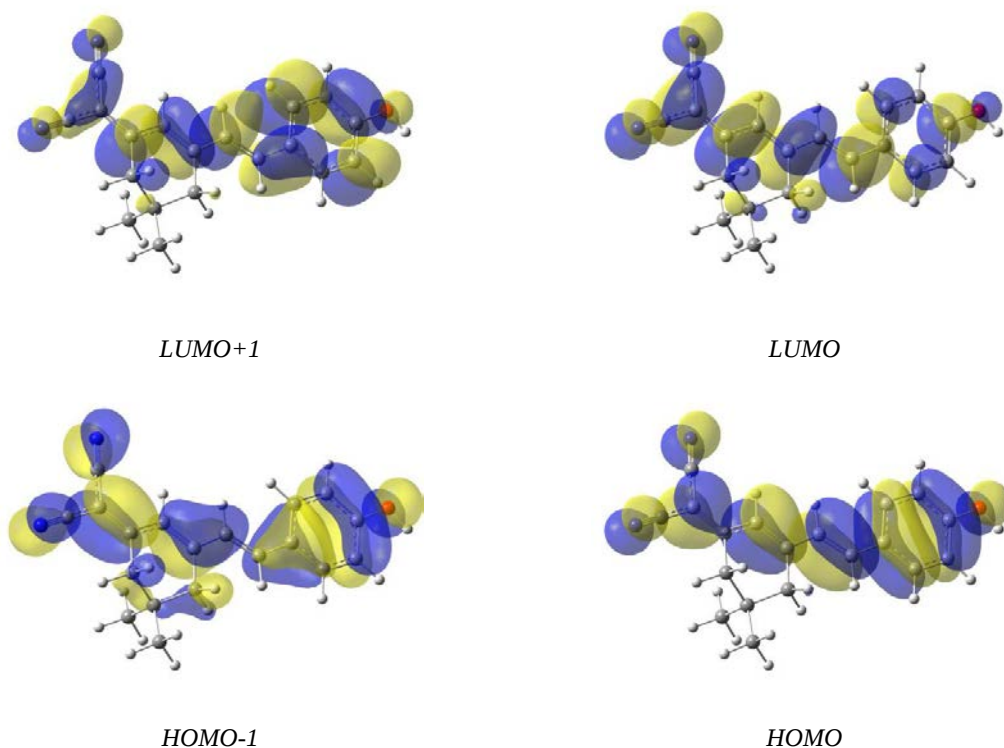


Fig.3. The first frontier and second frontier molecular orbitals of compounds **1**.

Table 7. Calculated HOMO-LUMO energy (a.u.) and HOMO-LUMO band gap ( $E_g$ ) values for **1**.

|                           |          |
|---------------------------|----------|
| HOMO                      | -0.22295 |
| LUMO                      | -0.11324 |
| $E_g$ [HOMO–LUMO]         | 0.10971  |
| HOMO–1                    | -0.26518 |
| LUMO +1                   | -0.04710 |
| $E_g$ [(HOMO–1)–(LUMO+1)] | 0.21808  |

#### 4. Conclusions

The OPA characterization of **1** has been obtained theoretically using CI method. According to the calculation results on the OPA characterization, the values of electronic transition wavelengths are located in the visible region, and are consistent with the experimental data previously reported. We have investigated the second-order NLO behaviour of **1** utilizing DFT and ab-initio TDHF procedure on electric dipole moments, static and dynamic dipole polarizabilities, second-order hyperpolarizabilities and also susceptibilities. Not only are the dipole moments and dipole polarizabilities encouragingly large, but also second-order hyperpolarizability and susceptibility values of **1** are higher than one would expect from this kind of compound. The computed non-zero hyperpolarizabilities and susceptibilities suggest that the title compound might have second-order NLO phenomena. The theoretical results on second-order hyperpolarizabilities and susceptibilities are compared with corresponding experimental values of **1** and with the values of similar structures reported in the literature. To provide a deeper understanding of the structure-property relationships of **1**; HOMO, LUMO and HOMO-LUMO band gaps for first and second frontier orbitals have been also evaluated by means of DFT method. Compound **1** shows small HOMO-LUMO gap, and thus produces the microscopic second-order hyperpolarizabilities with non-zero values.

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