Interaction of 1,1-Diamino-2,2-Dinitroethylene and Gallium - DFT Treatment

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Abstract

1,1-diamino-2,2-dinitroethylene, commonly known as FOX-7, is an insensitive pull-push type explosive of nitramine class. Aluminized FOX-7, depending on its Al content and multiplicity was found to be susceptible to decomposition. Gallium atom, a third group element below Al, also has an open shell electronic configuration. The present study considers FOX-7+nGa composites (n:1-3) and within the constraints of density functional theory at the level of UB3LYP/6-311++G(d,p) it has been found that gallium atom does not initiate any bond rupture. However, certain distortions in bond lengths and angles occur which might have some effect on the ballistic properties of FOX-7. Certain geometrical, quantum chemical and energy values of the composites as well as some spectral properties are presented.

1. Introduction

1,1-diamino-2,2-dinitroethylene (DADE, DADNE) is an insensitive high explosive known as FOX-7 [1]. It was synthesized in 1998 by members of the Swedish Defense Research Agency (FOI) [2, 3]. Many researchers have investigated its explosive potential thoroughly [4-18]. Nitration of 4,6-dihydroxy-2-methylpyrimidine and then hydrolysis constitutes an alternative route to FOX-7 [19].

It is a novel insensitive high-energy material possessing good thermal stability and low sensitivity. Moreover, it exhibits excellent application performance among the

$$H_2N \diagdown \qquad NO_2$$

FOX-7

$$H_2N \diagup \qquad NO_2$$
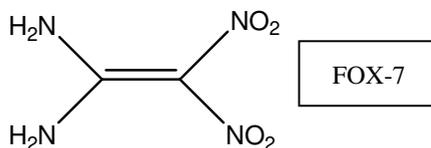
insensitive ammunitions and solid propellants. It has a simple molecular composition and structure, but it exhibits abundant chemical reactivity including coordination reactions, nucleophilic substitutions, acetylate reactions, oxidation and reduction reactions, electrophilic addition reactions etc., [20, 21]. FOX-7 is much less sensitive than RDX (in terms of impact, friction, and electrostatic discharge sensitivities) [22] although RDX or HMX possesses the same C/H/N/O ratio as FOX-7 has. FOX-7 possesses many polymorphic forms of such as α- and β-forms. Of which the α-form reversibly turns into β-form by heat treatment [23, 24]. At higher temperature, an irreversible conversion of β-polymorph occurs to yield γ-phase which decomposes at 504 K [23] and its decomposition has been extensively searched [25]. The effect of high pressure on the crystal structure of FOX-7 has also been studied [26]. In the last couple of decades several FOX-7 based propellant formulations have been developed in order to obtain propellant composites having a minimum or reduced smoke production [27].

Recently, some novel derivatives of FOX-7 and their properties as energetic materials have been reported [28, 29]. Some aluminized FOX-7 compositions were reported [30, 31]. Also some molecular orbital calculations were reported on aluminized FOX-7 [32-36].

In the present study, interaction of 1,1-diamino-2,2-dinitroethylene and gallium (FOX-7+nGa (n:1-3)) have been investigated at the molecular level within the restriction of density functional theory (DFT).

2. Method of Calculations

Geometry optimizations of all the presently considered structures leading to energy minima were initially achieved by using MM2 method followed by semi-empirical PM3 self-consistent fields molecular orbital (SCF MO) method [37, 38] at the restricted level [39, 40]. Subsequent optimizations were achieved at Hartree-Fock level using various basis sets. Then, the geometry optimizations were managed within the framework of density functional theory (DFT) using unrestricted B3LYP functional (UB3LYP) [41, 42] at the level of 6-311++G(d,p). The exchange term of B3LYP consists of hybrid

Hartree-Fock and local spin density (LSD) exchange functions with Becke's gradient correlation to LSD exchange [42, 43]. Note that the correlation term of B3LYP consists of the Vosko, Wilk, Nusair (VWN3) local correlation functional [44] and Lee, Yang, Parr (LYP) correlation correction functional [45]. Presently, the vibrational analyses have been also done at the same level of calculations which had been performed for the optimizations. The total electronic energies (E) are corrected for the zero point vibrational energy (ZPE) to yield E_c values. The normal mode analysis for each structure yielded no imaginary frequencies for the $3N$-6 vibrational degrees of freedom, where N is the number of atoms in the system. This indicates that the structure of each molecule corresponds to at least a local minimum on the potential energy surface. All these calculations were done by using the Spartan 06 package program [46].

3. Results and Discussion

Quite often some metals are employed with certain explosives to get better performance exhibiting ammunitions. Aluminum is one of them. However, a recent computational study has revealed that depending on the aluminum content and over all spin state of the composite, aluminized FOX-7, may undergo decomposition, while aluminum atom is oxidized and one of the N-O bonds of FOX-7 is broken [36].

In the present study, various gallium composites of FOX-7 are considered as analogous approach to its above mentioned aluminized composites. Gallium atom which is a third group element below the aluminum has $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^1$ electronic configuration in its ground state. The ionization potentials of the electrons of Ga atom in valency shell (n:4) are 30.6, 20.43 and 5.97 eV for 4s, 4s and 4p, respectively [47]. Whereas, the respective potentials for Al atom (3s, 3s and 3p) are 28.31, 18.74 and 5.96 eV [47]. The effective nuclear charge of Ga atom is greater than that of aluminum. Hence, the gallium atom shows smaller tendency to ionize than the aluminum atom [47]. All these information suggest that the presence of Ga should not cause any bond dissociation (maybe up to certain number of Ga atoms) of FOX-7 by transferring sufficient amount of electron density. However, it might cause deformation type perturbations on FOX-7 structure, thereby influencing its push-pull type behavior which might causes some changes on its ballistic properties. Below the influence of Ga on FOX-7 structure at the molecular level has been investigated. Note that depending on the number of Ga atoms, the composites have different multiplicities.

Figure 1 shows the optimized structures of FOX-7 and its gallium composites from
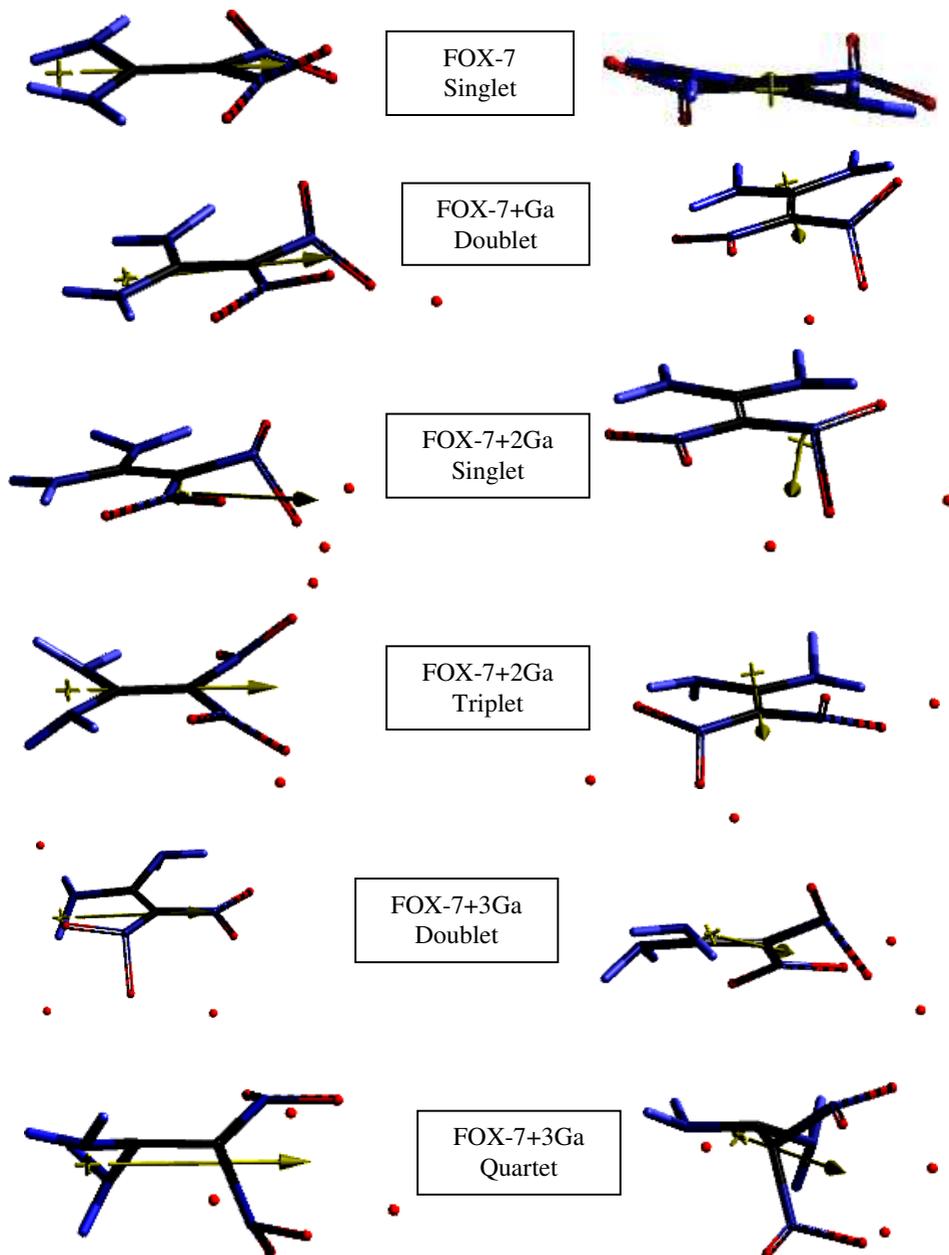
Figure 1. Optimized structures of FOX-7 and FOX-7+Ga composites from different angles of view.

different angles of view. As seen in the figure, FOX-7 molecule is nearly planar. This geometry, together with two dimensional network of hydrogen bonds, contributes the pull-push character of the molecule and stability thus providing insensitiveness to the molecule. The electron donating ability of the amino groups in FOX-7 satisfies greatly the electron demand of the nitro groups, thus reducing their oxidative power. In the FOX-7+Ga composites, this nearly planar geometry is distorted bit by bit and in FOX-7+3Ga quartet the nitro groups became nearly perpendicular to the plane engendered by the amino groups and carbon atoms. That means that the composite should have lost its pull-push type character.

As seen in the figure some bond angles and dihedral angles concerning the amino as well as the nitro groups change as the number of Ga atoms vary. Figure 2 shows the bond lengths of FOX-7 and its gallium composites. The carbon-carbon bond lengths in the composites do not vary much (1-38-1.48 Å), but N-O bond which is 1.48 Å (in FOX-7 1.21-1.24 Å) in singlet FOX-7+2Ga and C-NO$_2$ bond which is 1.52 Å (in FOX-7 1.43 Å) in quartet FOX-7+3Ga notably elongate.
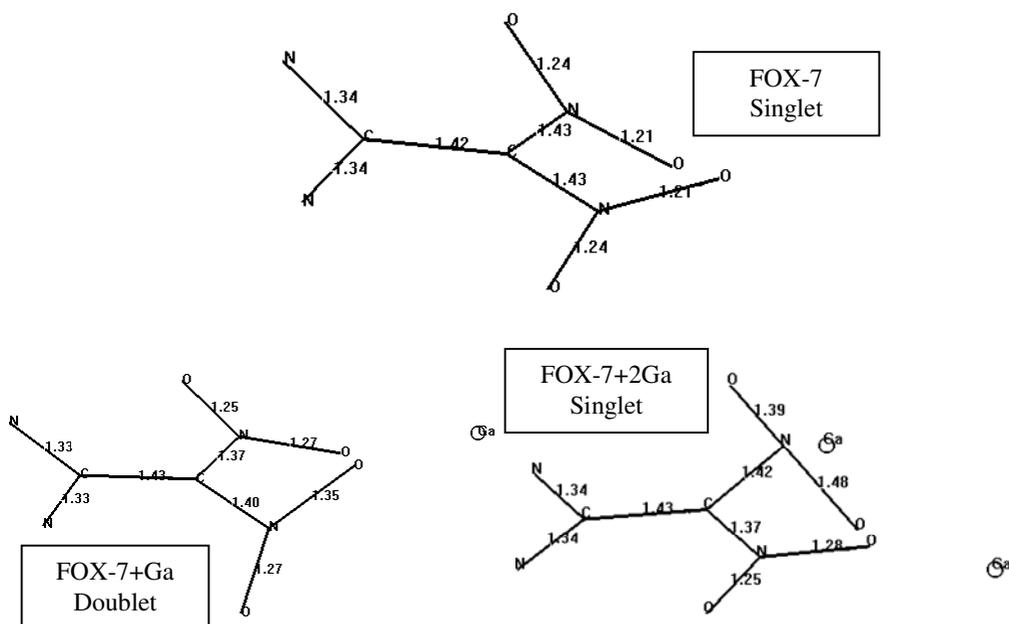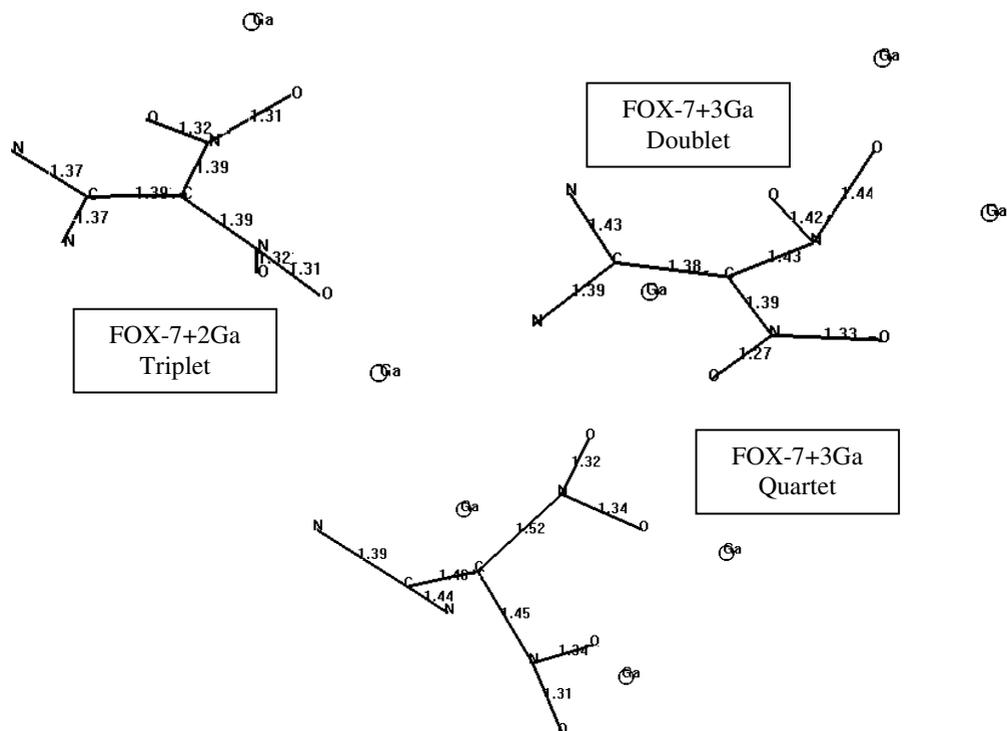
Figure 2. Bond lengths (Å) of FOX-7 and its gallium composites.

The presently calculated bond lengths of FOX-7 are in good accord with the literature data such that the reported length of the bond between carbon atoms in the molecule (1.456 Å) is intermediate between the length of a normal single bond (1.54 Å) and a normal double bond (1.34 Å). The lengths of the C–NO$_2$ bonds (1.42 and 1.39 Å) are close to the normal bond length of nitrogen-carbon with sp^2 hybridization (1.40 Å), but the C–NH$_2$ bonds (1.31 and 1.32 Å) are shorter by about 0.1 Å. The molecule has a generally planar structure [10, 48, 49].

Figure 3 shows the IR spectra of FOX-7 and its gallium composites. FOX-7 possesses two N-H stretchings at 3469-3676 cm^{-1}. Its NH$_2$ scissoring vibrations occur at 1644 cm^{-1}. The C=C and C-N stretchings happen in the region of 1568-1623 cm^{-1}. FOX-7+Ga composite has three N-H stretchings (asymmetric and symmetric) in the region of 3344-3667 cm^{-1}. Whereas FOX-7+2Ga singlet has four but FOX-7+2Ga triplet just two N-H stretchings. Depending on the number and multiplicity of the composites IR spectra of them below 1500 cm^{-1} exhibit some peculiarities.
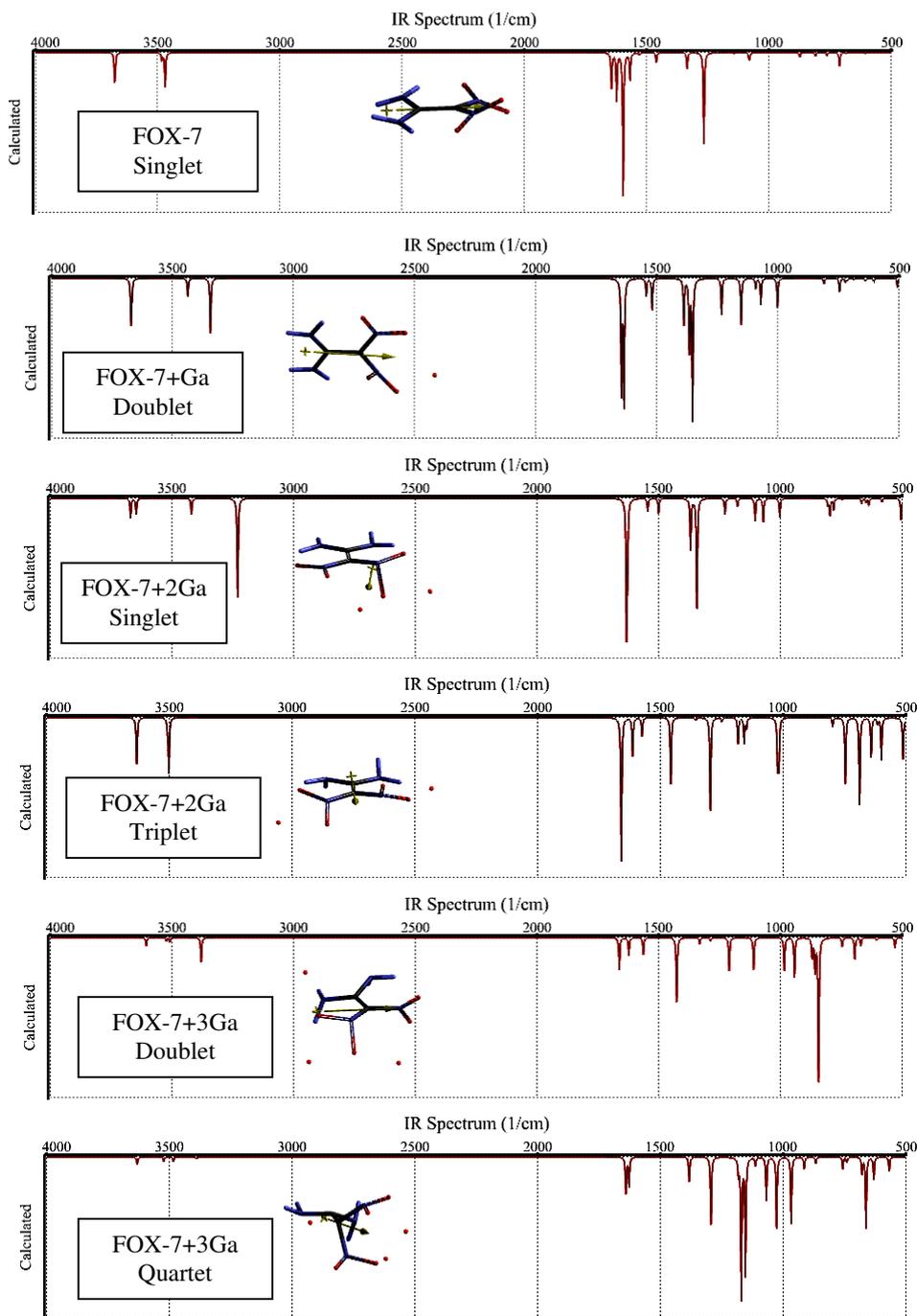
Figure 3. The calculated IR spectra of FOX-7 and its gallium composites.

Table 1 tabulates some properties of the composites. The first column includes PM3//UB3LYP/6-311++G(d,p) values for the heat of formation values at the standard states in vacuum whereas the second column lists T1 results. The composite formation appears to be exothermic process in all the cases. T1 calculations involve more elaborate treatment. For FOX-7+2Ga case the triplet state has less exothermic heat of formation value in both cases of the calculations. Whereas for FOX-7+3Ga composite the quartet is more exothermic than the doublet by PM3//UB3LYP/6-311++G(d,p) level of calculations in contrast to T1 calculations which yield results that the quartet happens to be less exothermic than the doublet.

Table 1. Some properties of the composites.

Structure	Heat of formation*	Heat of formation**	Area (Å^2)	Volume (Å^3)	Ovality	Dipole moment (Debye)	MW (amu)
FOX-7+Ga Doublet	-107.552	-4754520.42	162.97	134.30	1.28	6.60	217.80
FOX-7+2Ga Singlet	-227.515	-9509154.14	186.58	158.74	1.32	6.04	287.52
FOX-7+2Ga Triplet	-193.662	-9509024.15	195.76	161.81	1.36	2.92	287.52
FOX-7+3Ga Doublet	-344.364	-14263573.76	212.69	184.95	1.35	7.96	357.25
FOX-7+3Ga Quartet	-582.205	-14263121.28	202.77	182.90	1.30	4.76	357.25

Heat of formation values in kJ/mol.*PM3// UB3LYP/6-311++G(d,p), **T1

As for the areas and volumes, as normally expected, they increase as the number of Ga atoms increase. However, the greater multiplicity state has greater values keeping the Ga content be the same but the quartet has smaller area and volume as compared to the doublet for FOX-7+3Ga in the case of FOX-7+3Ga composites. The ovality follows the same trend.

On the other hand, the order of dipole moments of the composites (FOX-7+nGa) is 3Ga(d) > Ga(d) > 2Ga(s) > 3Ga(q) > 2Ga(t) where the letter in parenthesis indicate the multiplicities.

Figures 4 and 5 show the ESP and natural charges on the atoms of the composites. Note that the ESP charges are obtained by the program based on a numerical method that generates charges that reproduce the electrostatic potential field from the entire wavefunction [46]. In both types of charge calculations Ga atom(s) acquire some positive charge less than unity, causing some distortions in the pull-push contributors, namely the amino and nitro groups. The carbon-carbon bond lengths do not vary appreciably (see Figure 2) but the others are affected which are accompanied by changes in bond and dihedral angles. In some of the composites considered, the groups are notably out of plane, e.g., quartet state of FOX-7+3Ga.
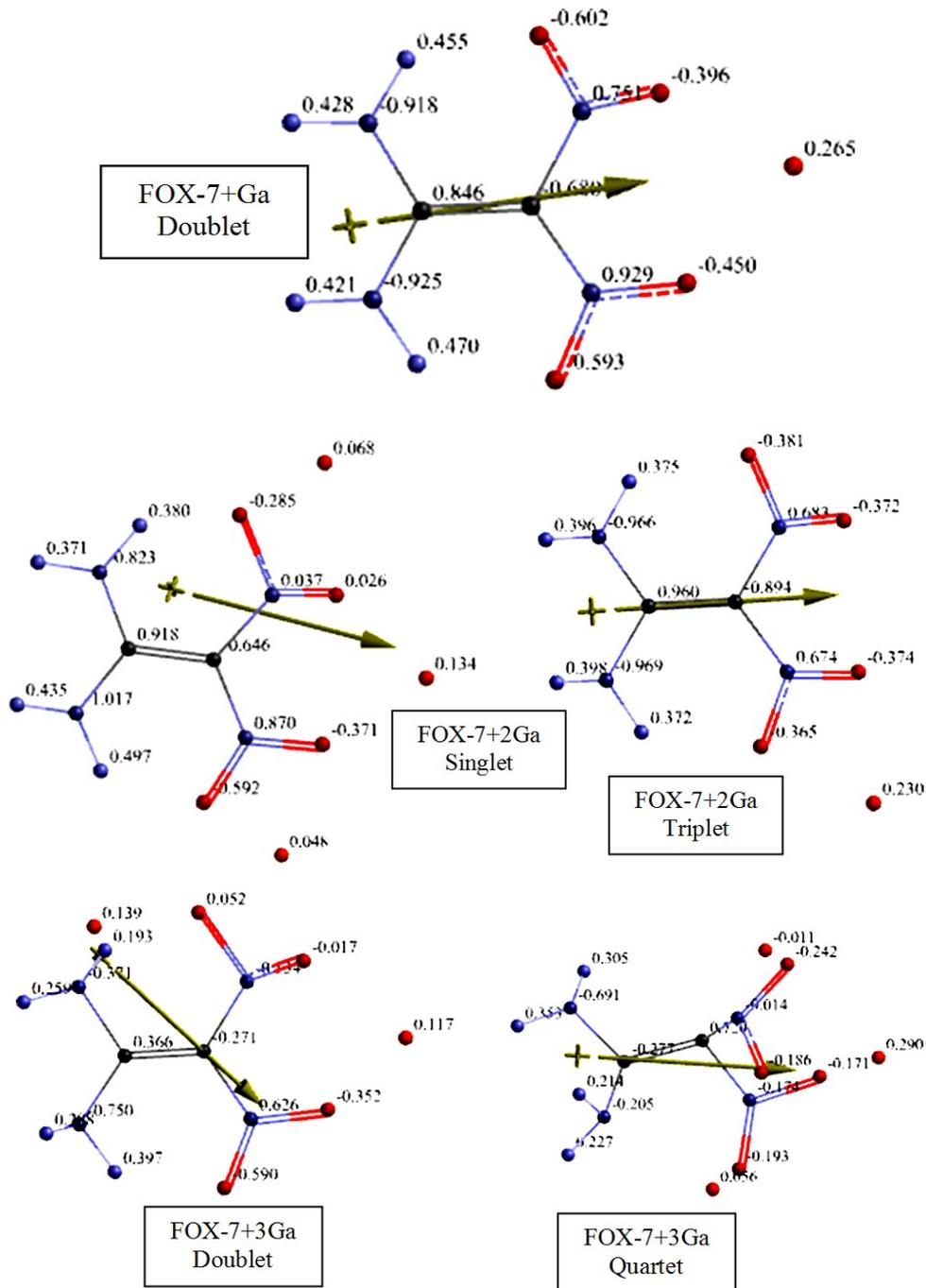
Figure 4. ESP charges on atoms of the composites.
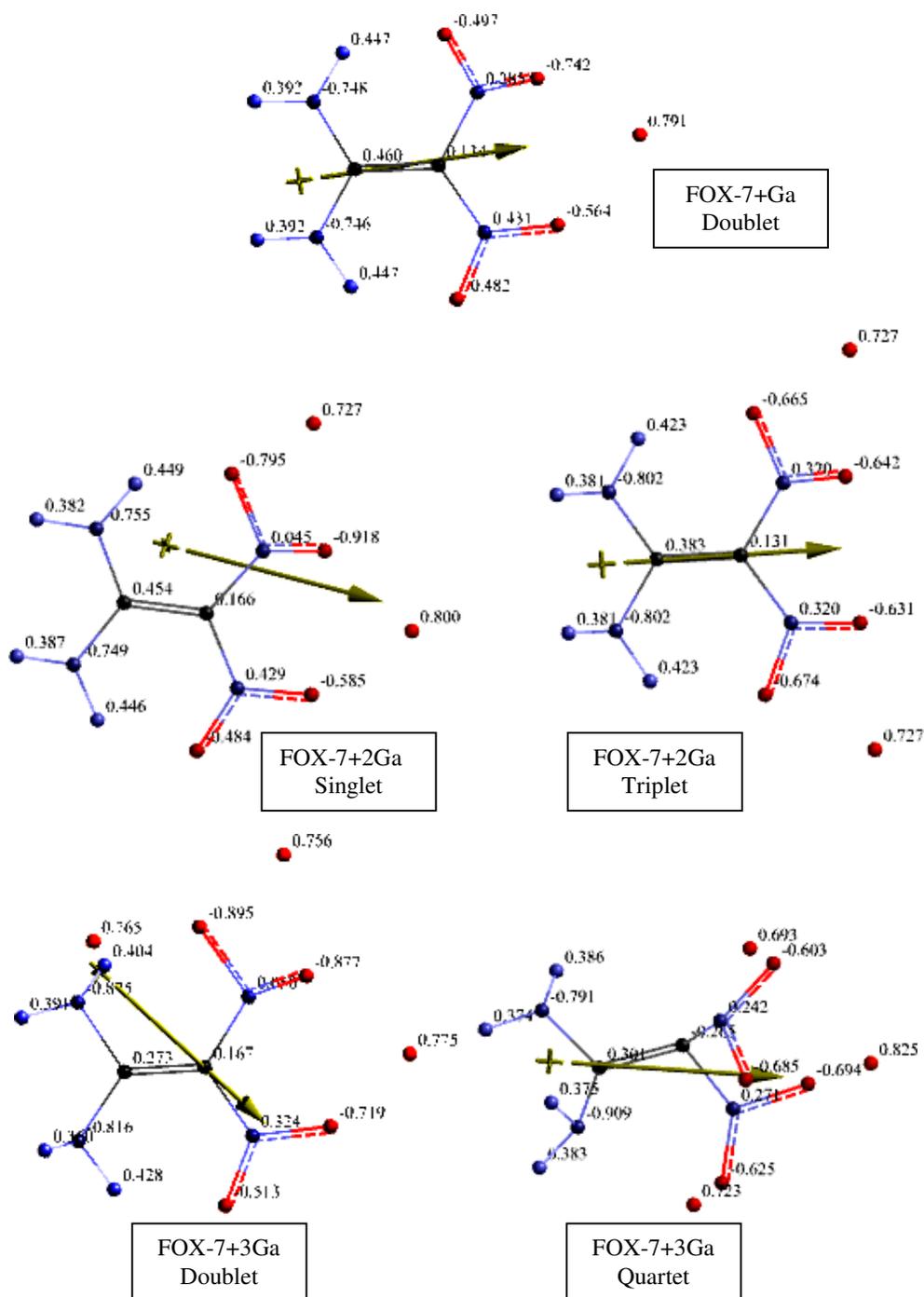
Figure 5. Natural charges on atoms of the composites.

Table 2 shows various energies of the composites considered where E, ZPE and E_c stand for the total electronic energy, zero-point vibrational energy and the corrected total electronic energy, respectively. As seen in the table, keeping the number of Ga atoms in each set, lower multiplicity having composite is more stable than the higher one based on E_c values.

Table 2. Various energies of the composites.

Structure	E	ZPE	E_c
FOX-7+Ga Doublet	-6625228.50	236.465330	-6624992.035
FOX-7+2Ga Singlet	-11679113.0	237.189441	-11678875.81
FOX-7+2Ga Triplet	-11679017.4	235.419464	-11678781.98
FOX-7+3Ga Doublet	-16732845.5	236.923245	-16732608.58
FOX-7+3Ga Quartet	-16732666.4	232.273075	-16732434.13

Energies in kJ/mol.

Figure 6 shows some of the molecular orbital energy levels of the composites. Note that some of the composites have certain multiplicities which lead to open shell structures and then the unrestricted calculations yield α- and β-type orbitals (in the figure a- and b-types). In those cases α-HOMO energy levels are higher than the respective β-type orbital energies. A similar situation happens for the LUMO energy levels with the exception of FOX-7+ Ga case. The HOMO and LUMO energy levels are collected in Table 3. It also includes Δε values which are the interfrontier energy gaps, namely ε_{LUMO}-ε_{HOMO}. The order of HOMO energy levels of FOX-7+nGa composites is 2Ga(s)<Ga(d) <3Ga(q)<2Ga(t)<3Ga(d). Whereas the LUMO energy levels follow the order of Ga(d)<2Ga(s)<3Ga(d)<2Ga(t)<3Ga(q). Consequently, the order of Δε values happens as 3Ga(q)>2Ga(s)>2Ga(t)>Ga(d)>3Ga(d). Note that Δε values in the table are constituted in between the smallest possible difference of the HOMO and LUMO energies corresponding to α- and/or β-type orbitals.
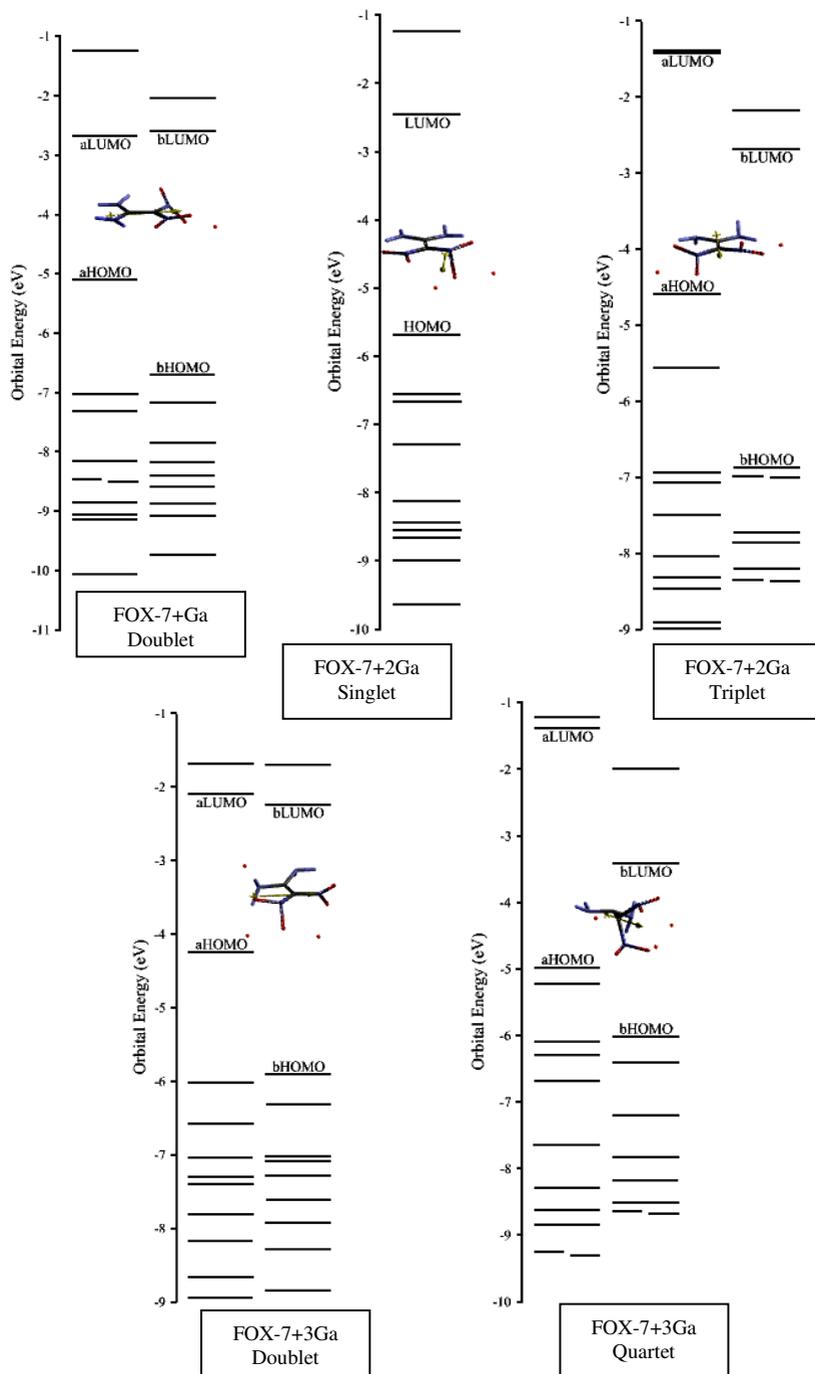
Figure 6. Some of the molecular orbital energy levels of the composites.

Table 3. Some molecular orbital energies of the composites.

Structure	HOMO	LUMO	Δε
FOX-7+Ga Doublet	-492.561583	-258.310027	234.2516
FOX-7+2Ga Singlet	-547.982115	-237.037986	310.9441
FOX-7+2Ga Triplet	-443.015922	-136.783885	306.232
FOX-7+3Ga Doublet	-409.494317	-202.589026	206.9053
FOX-7+3Ga Quartet	-480.136021	-133.599811	346.5362

Energies in kJ/mol.

The time-dependent UV-VIS spectra (TDDFT) of the composites are shown in Figure 7. As seen in the figure, FOX-7+2Ga singlet and FOX-7+3Ga doublet absorptions should occur in the visible region whereas the others occur in the UV-VIS range of the spectrum.
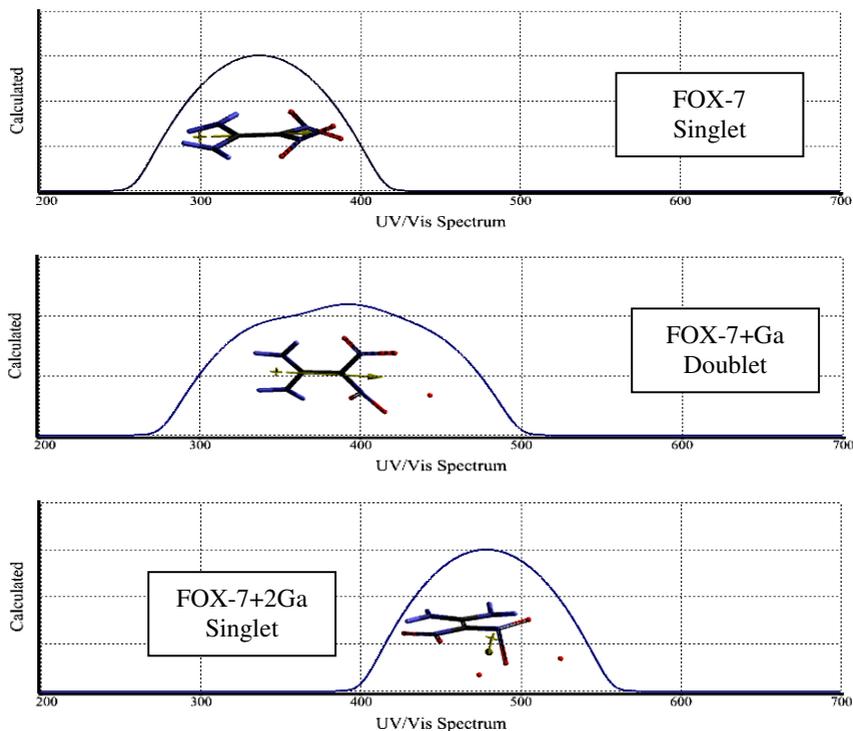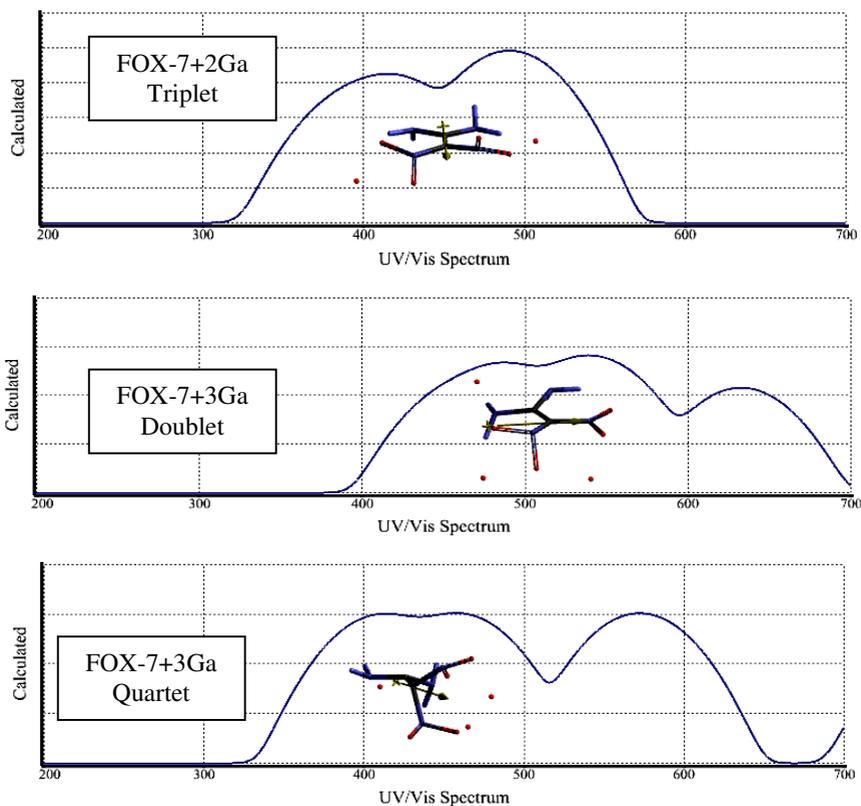
Figure 7. UV-VIS spectra of FOX-7 and its gallium composites.

Figure 8 shows the effect of gallium atoms on the HOMO and LUMO patterns of the composites.

FOX-7+2Ga
Singlet

| HOMO | LUMO |

FOX-7+2Ga
Triplet

| αHOMO | βHOMO | αLUMO | βLUMO |

FOX-7+3Ga
Doublet

| αHOMO | βHOMO | αLUMO | βLUMO |

FOX-7+3Ga
Quartet

| αHOMO | βHOMO | αLUMO | βLUMO |
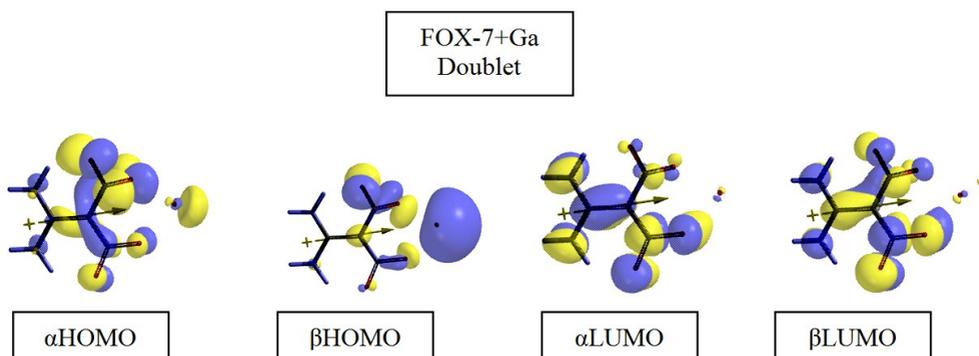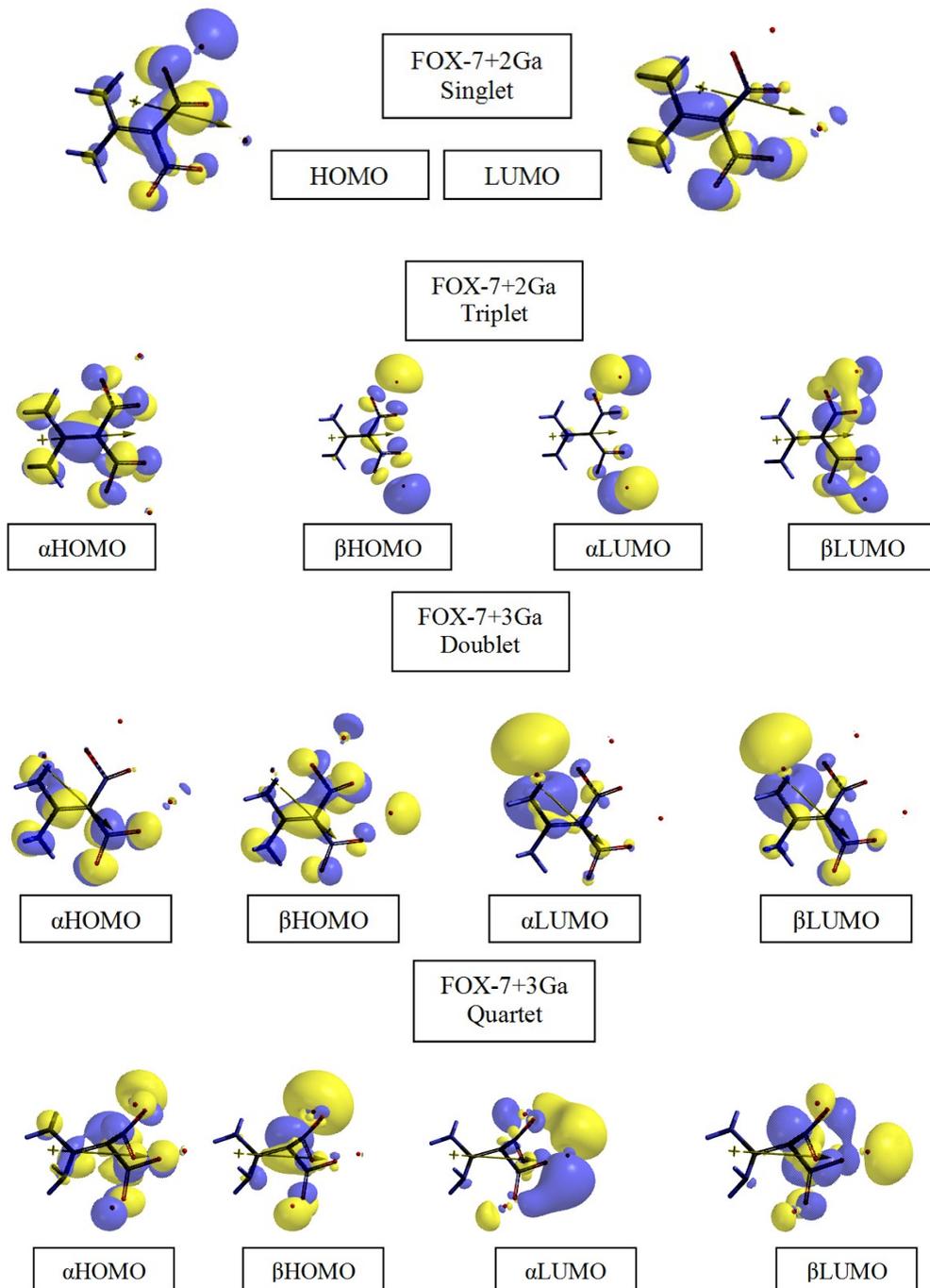
Figure 8. The HOMO and LUMO patterns of the composites.

Figure 9 shows the electrostatic potential maps of the composites. Note that in the figure red and blue regions stand for electronegative and electropositive potential regions, respectively. As seen in the figure, Ga atom(s) perturbs electrostatic potential field around FOX-7 although no bond ruptures occur.
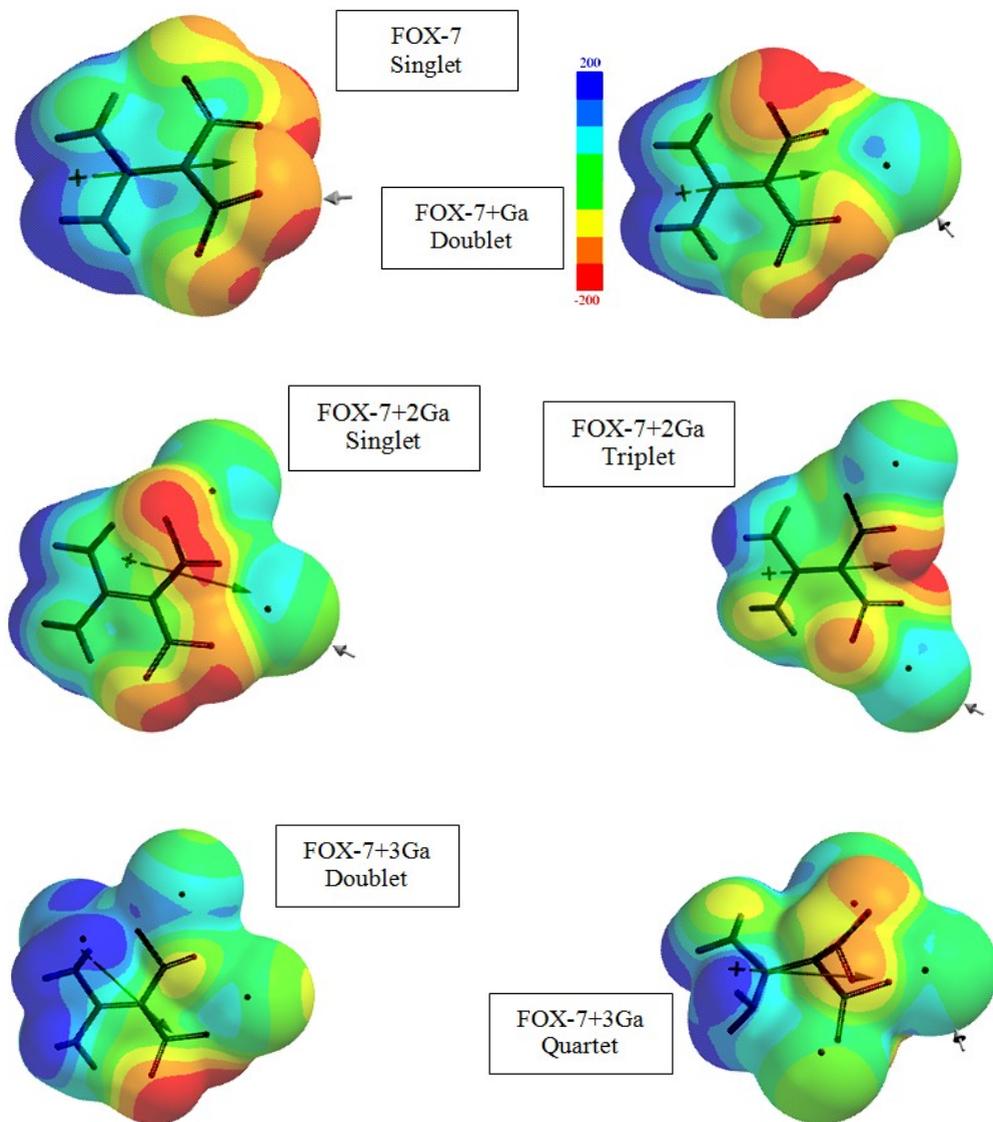
Figure 9. Electrostatic potential maps of FOX-7 and its gallium composites.

4. Conclusion

Within the restrictions of the density functional theory at the applied level, it has been found that FOX-7+nGa (n:1-3) composites are exothermic and electronically stable no matter the multiplicities of the composites are. However, certain bond length and angle distortions occur which affect certain geometrical, quantum chemical and energetic properties of FOX-7. All those perturbations probably affect ballistic properties of the explosive.

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