Electronic Excited States of Trimethylenemethane and Bis-verdazyl Dimer

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Abstract

❖ Molecular System that exhibit high and low spin electronic state are called as Qudits and found recent interest for quantum computing[1].

❖ Our interest is the better understand spin Hamiltonian in two model systems.

$$H(S) = H_{ZI} + H_{EX} + H_{ZFI} + H_{HFI} + H_{DD} + H_{NQI}$$

❖ We are reporting here Electronic Structure and Magnetic characteristics in organic biradical TMM and Bis-Verdazyl Dimer.

❖ TMM have triplet ground state and Bis-Verdazyl have singlet ground state.

❖ We have calculated Exchange interaction and zero field splitting (E & D) parameter.

Introduction

❖ TMM and Bis-verdazyl dimer has a non- Kekule Hydrocarbon.

Characteristics of Qubit and example[2]

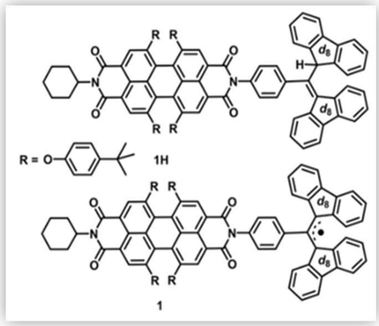
❖The fundamental units of quantum information are called qubits. Qudits are the electronic states with different spin-multiplicities.

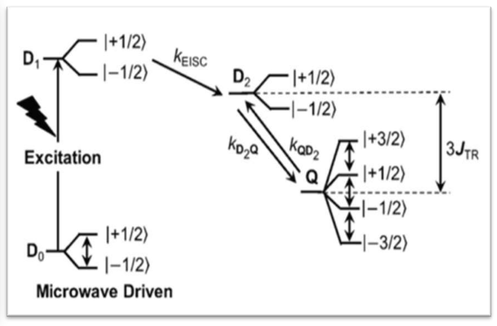
❖Qubit development based to the Divincenzo criteria , criteria given below

✓Well-defined qubits

✓Initialization to a pure state

✓Long coherence times





Objectives

❖To understand the electronic structure and spin dynamics of TMM molecule.

❖To calculate the excited state energies of TMM.

❖To understand Jahn Teller distortion in TMM.

❖To calculate Exchange interaction(J) in TMM molecule.

❖To calculate Zero Field Splitting Parameter (E and D) in TMM.

❖To Calculate Energy deference between singlet and triplet electronic state in BVD.

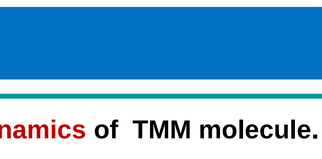
❖To Calculate Magnetic Nature with help of Exchange coupling (J) in BVD.

Set Up

❖Optimization: CASSCF/CCD/GAMESS

❖Excited states: SA6-XMCQDPT/CCD/GAMESS/6,8

❖ Zero Field Splitting Parameter: def2-TZVP def2/J def2/JK def2-TZVP/C/ORCA



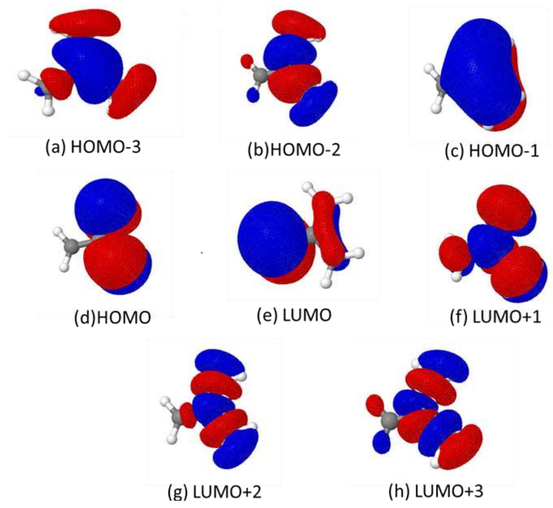
❖ Exchange interaction using Broken Symmetry approximation[3]

$$J = -\frac{(E_{HS} - E_{BS})}{< S_{HS}^2 > - < S_{BS}^2 >}$$

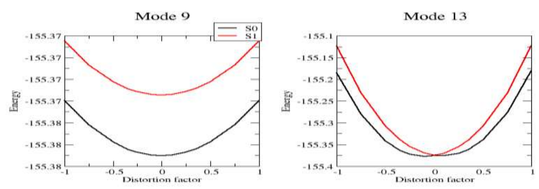
Results

Excited State Calculation of TMM (active space 8,8)

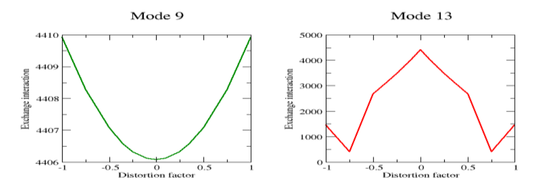
State	ΔE	*f*	μ
T_0	0.000	0.000	0.26
S_1	0.986	-	0.62
S_2	0.995	-	0.01
S_3	3.300	-	<0.01
T_1	4.445	0.003	0.57
T_2	4.476	0.002	<0.01



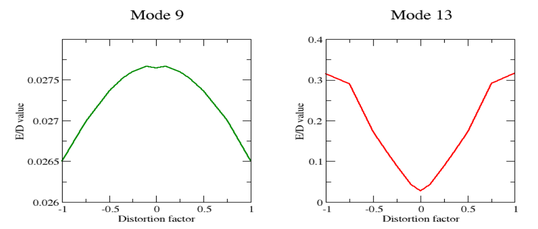
The Molecular orbital of the active Space (8,8) of TMM



Jahn Teller distortion representation of degenerate singlet electronic state modes 9 and 13.



The exchange interaction(cm-1) PES curve in mode 9 and mode 13 , and showing significant change in mode13 (Jahn teller active) with distortion factor. [4]



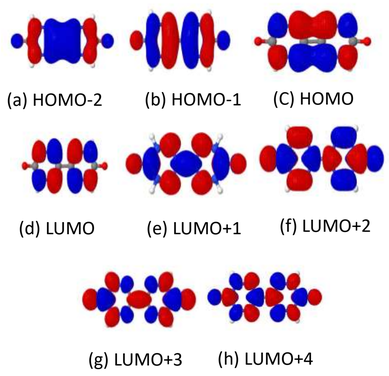
Calculate E/D values PES curve with distortion factor in modes[5]

	Experimental J value	Theoretical results
Gaussian package	16.1kcal/mol	11.27kcal/mol
ORCA package		12.5kcal/mol

Values	MRCI (cm^{-1})	
	D	E
Experimental	0.02	0
Theoretical	0.028	0.027

Excited State Calculation of Bis-Verdazyl Dimer(active space 6,8)

State	ΔE (ev)	Wavelength (nm)	*f*	μ
S_0	0.000	-	-	0.001
T_1	0.235	-	-	-
T_2	2.612	-	-	-
S_1	2.867	432.45	0.000	0.001
S_2	2.947	420.71	<0.01	0.002
T_3	2.948	-	-	-
S_3	3.072	403.59	0.011	0.001
S_4	3.163	391.98	0.000	0.002
T_4	3.836	-	-	-
S_5	4.115	301.29	>0.1	0.013
T_5	4.138	-	-	-
S_6	4.332	286.20	0.000	0.013
T_6	5.033	-	-	-



The Molecular orbital of the active Space (6,8) of Bis-Verdazyl diradical

Conclusions

❖ Value of exchange interaction varying drastically in jahn teller active modes.

❖ Value of E and D parameter match well with experimental vaule.

❖ Vertical excited state calculation result in BVD also done[6].

References

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