Table 1: Vertical excitation energies and dominant contributions of the S_0 and S_1 states of fulvene optimized with SA2-CASSCF(6,6)/6-31G and MRCI(CAS(6,6))/6-31G. For MRCI, the Pople correction is also given (MRCI/+Pople).

State	ΔE (eV)	Configuration	%
		SA2-CASSCF(6,6) – S_0 optimization	
S_0	0.000	$(1b2)^2(2b2)^2(3b2)^0(4b2)^0$ $(1a2)^2(2a2)^0$	75.7
S_1	4.080	$(1b2)^2(2b2)^2(3b2)^1(4b2)^0$ $(1a2)^1(2a2)^0$	72.0
		$(1b2)^2(2b2)^1(3b2)^2(4b2)^0$ $(1a2)^1(2a2)^0$	14.8
		SA2-CASSCF(6,6) – S_1 optimization	
S_0	1.403	$(1b2)^2(2b2)^2(3b2)^0(4b2)^0$ $(1a2)^2(2a2)^0$	65.4
		$(1b2)^2(2b2)^1(3b2)^1(4b2)^0$ $(1a2)^2(2a2)^0$	13.8
S_1	2.630	$(1b2)^2(2b2)^2(3b2)^1(4b2)^0$ $(1a2)^1(2a2)^0$	70.8
		$(1b2)^2(2b2)^1(3b2)^2(4b2)^0$ $(1a2)^1(2a2)^0$	16.1
		SA2-CASSCF(6,6) – MXS optimization	
S_0	2.932	$(19a)^2(20a)^2(21a)^1(22a)^1(23a)^0(24a)^0$	71.7
		$(19a)^2(20a)^1(21a)^1(22a)^2(23a)^0(24a)^0$	15.5
S_1	2.932	$(19a)^2(20a)^2(21a)^2(22a)^0(23a)^0(24a)^0$	58.6
		$(19a)^2(20a)^1(21a)^2(22a)^1(23a)^0(24a)^0$	16.9
		MRCI – S_0 optimization	
S_0	0.000/0.000	$(1b2)^2(2b2)^2(3b2)^0(4b2)^0$ $(1a2)^2(2a2)^0$	69.0
S_1	3.907/3.779	$(1b2)^2(2b2)^2(3b2)^1(4b2)^0$ $(1a2)^1(2a2)^0$	70.1
		MRCI – S_1 optimization	
S_0	1.268/1.194	$(1b2)^2(2b2)^2(3b2)^0(4b2)^0$ $(1a2)^2(2a2)^0$	61.8
		$(1b2)^2(2b2)^1(3b2)^1(4b2)^0$ $(1a2)^2(2a2)^0$	10.5
S_1	2.638/2.600	$(1b2)^2(2b2)^2(3b2)^1(4b2)^0$ $(1a2)^1(2a2)^0$	67.3
		$(1b2)^2(2b2)^1(3b2)^2(4b2)^0$ $(1a2)^1(2a2)^0$	10.8

MRCI – MXS optimization
S_0
S_1

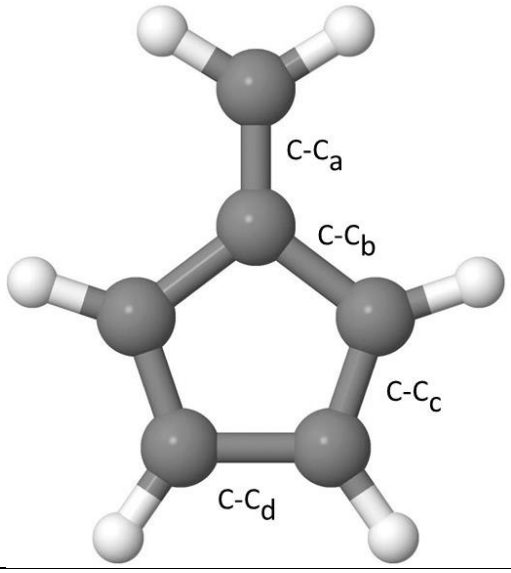
Table 2: Total energies in Hartree of fulvene

	S_0	S_1
SA2-CASSCF(6,6)-S_0 opt	-230.64459	-230.49466
SA2-CASSCF(6,6)-S_1 opt	-230.59303	-230.54794
SA2-CASSCF(6,6)-MXS-planar	-230.53683	-230.53683
MRCI-S_0 opt	-231.07035	-230.92677
MRCI+Q-S_0 opt	-231.14743	-231.00857
MRCI-S_1 opt	-231.02375	-230.97340
MRCI+Q-S_1 opt	-231.10356	-231.05189
MRCI-MXS-planar		

Table 3: Oscillator strength of the S_0 to S_1 transition of fulvene optimized with SA2-CASSCF(6,6)/6-31G* and MRCI(CAS(6,6))/6-31G.

Method	*f*
SA2-CASSCF(6,6) – S_0 optimization	0.00
SA2-CASSCF(6,6) – S_1 optimization	0.00
MRCI – S_0 optimization	0.01
MRCI – S_1 optimization	0.00

Table 4: C-C bond distances of the optimized S_0, S_1, and crossing seam structures using the SA2-CASSCF(6,6)/6-31G and MRCI(CAS(6,6))/6-31G methods.



	C-C_a	C-C_b	C-C_c	C-C_d
		SA2-CASSCF(6,6)		
S_0	1.352	1.479	1.360	1.482
S_1	1.497	1.402	1.475	1.363
MXS	1.583	1.373	1.538	1.321
		MRCI		
S_0	1.358	1.489	1.369	1.493
S_1	1.495	1.413	1.482	1.375
MXS				

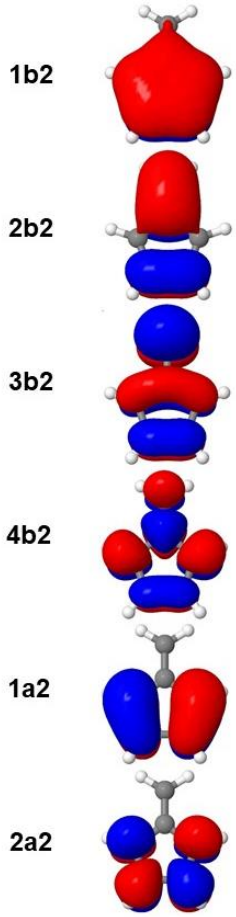


Figure 1: Optimized active orbitals for the S_0 optimized with SA2-CASSCF(6,6)/6-31G.

Directories on CCR:

	SA2-CASSCF(6,6)/6-31G
S_0 opt	/user/ub2037/fulvene/S0-CAS
S_1 opt	/user/ub2037/fulvene/S1-CAS
MXS	/user/ub2037/fulvene/MXS-CAS/mxs_opt
S_0 opt	/user/ub2037/fulvene/S0-CI
S_1 opt	/user/ub2037/fulvene/S1-CI
MXS	