

Stereocontrol of ultrafast electron dynamics in ABCU with attosecond laser pulses : a computational study⁺

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Supporting Information

The supporting information contains 3 Tables that give more details about the *ab initio* computations on ABCU.

Table S1 : Matrix elements of the dipole for the 19 states included in the computation of the electron dynamics. The GS and 18 excited states are computed at the average CAS(8,13)/6-31++G(d,p) level.¹

Table S2 : List of the main CSF's contributing to the GS and the 18 excited states in the average CAS(8,13)/6-31++G(d,p) computation.¹

Table S3 : Energies, permanent and transition dipole moments for the ten lowest states below the IP computed at the CIS(D)/ 6-31++G(d,p) level.²

⁺ Presented at the XIII Stereodynamics meeting, Santa Cruz, November 2010

Table S1. Matrices of the x, y and z components of the dipole operator computed in the basis of the 19 electronic states obtained from a CAS average (8,13)/6-31++G(d,p) computation at the equilibrium geometry of the GS computed at the same level.

x component (in a.u.)

	GS(A)	1 st A	1 st E	1 st E	2 nd A	2 nd E	2 nd E	3 rd E	3 rd E	3 rd E	3 rd A	4 th E	4 th E	4 th E	4 th A	5 th A	6 th E	6 th E	16 ES	17 ES	18 ES
GS	-0.01	0.00	0.65	-0.39	0.00	-0.06	-0.26	-0.13	0.13	0.00	0.15	-0.08	-0.02	-0.14	-0.04	0.24	-0.04	0.03	0.07		
1 st A	0.00	0.00	-3.64	1.94	-0.01	0.05	0.55	-0.67	0.23	0.01	-0.04	0.03	0.01	-0.02	0.00	0.03	-0.01	-0.09	0.05		
1 st E	0.65	3.64	0.66	0.22	-2.21	-1.23	3.45	-0.17	-0.42	1.61	-0.03	-0.01	-0.03	-0.10	0.02	0.02	0.07	0.01	0.01		
1 st E	-0.39	1.94	0.22	-0.66	1.24	3.44	1.23	-0.40	0.21	-0.96	-0.01	0.02	0.02	0.01	-0.10	0.07	0.03	0.01	0.02		
2 nd A	0.00	-0.01	-2.21	1.24	0.03	1.07	0.46	3.38	-2.20	-0.02	-0.10	0.08	0.01	0.00	0.01	0.01	0.01	-0.02	0.01		
2 nd E	-0.06	0.05	-1.23	3.44	1.07	1.36	0.20	-0.01	-0.96	-0.48	0.00	0.01	0.00	0.03	-0.01	0.01	0.00	-0.01	0.00		
2 nd E	-0.26	0.55	3.45	1.23	0.46	0.20	-1.37	-0.98	0.01	-0.46	0.01	0.00	0.00	0.00	0.01	-0.01	0.00	0.00	0.00		
3 rd E	-0.13	-0.67	-0.17	-0.40	3.38	-0.01	-0.98	0.17	1.02	0.58	0.00	0.01	-0.05	0.10	0.00	-0.01	-0.05	-0.02	-0.01		
3 rd E	0.13	0.23	-0.42	0.21	-2.20	-0.96	0.01	1.02	-0.20	-0.11	0.01	-0.01	0.02	-0.04	0.09	-0.06	-0.03	-0.01	0.01		
3 rd A	0.00	0.01	1.61	-0.96	-0.02	-0.48	-0.46	0.58	-0.11	-0.04	-0.07	0.04	0.01	0.02	0.02	-0.04	0.01	0.00	0.00		
4 th E	0.15	-0.04	-0.03	-0.01	-0.10	0.00	0.01	0.00	0.01	-0.07	-0.72	-0.27	-0.34	-3.15	2.01	0.87	-0.50	0.12	0.12		
4 th E	-0.08	0.03	-0.01	0.02	0.08	0.01	0.00	0.01	-0.01	0.04	-0.27	0.69	0.15	0.58	-0.66	1.91	-3.20	0.15	-0.20		
4 th A	-0.02	0.01	-0.03	0.02	0.01	0.00	0.00	-0.05	0.02	0.01	-0.34	0.15	0.09	0.18	-0.09	0.02	-0.01	-2.32	1.54		
5 th A	-0.14	-0.02	-0.10	0.01	0.00	0.03	0.00	0.10	-0.04	0.02	3.15	0.58	0.18	0.30	0.14	-0.14	-0.07	-2.14	0.49		
6 th E	-0.04	0.00	0.02	-0.10	0.01	-0.01	0.01	0.00	0.09	0.02	2.01	-0.66	-0.09	0.14	-0.44	-0.01	0.04	1.13	-0.35		
6 th E	0.24	0.03	0.02	0.07	0.01	0.01	-0.01	-0.01	-0.06	-0.04	0.87	1.91	0.02	-0.14	-0.01	-0.41	0.12	0.29	1.11		
16 ES	-0.04	-0.01	0.07	0.03	0.01	0.00	0.00	-0.05	-0.03	0.01	-0.50	-3.20	-0.01	-0.07	0.04	0.12	0.46	-0.05	-1.23		
17 ES	0.03	-0.09	0.01	0.01	-0.02	-0.01	0.00	-0.02	-0.01	0.00	0.12	0.15	-2.32	-2.14	1.13	0.29	-0.05	0.69	0.26		
18 ES	0.07	0.05	0.01	0.02	0.01	0.00	0.00	-0.01	0.01	0.00	0.12	-0.20	1.54	0.49	-0.35	1.11	-1.23	0.26	-0.53		

y component (in a.u.)

	GS(A)	1 st A	1 st E	1 st E	2 nd A	2 nd E	2 nd E	3 rd E	3 rd E	3 rd E	3 rd A	4 th E	4 th E	4 th E	4 th A	5 th A	6 th E	6 th E	16 ES	17 ES	18 ES	18 ES
GS	0.00	0.00	0.38	0.65	0.00	0.26	-0.07	-0.13	-0.13	0.00	0.08	0.15	-0.01	-0.03	0.23	0.05	0.15	-0.04	-0.05			
1 st A	0.00	0.00	-1.94	-3.65	0.00	-0.53	0.06	-0.23	-0.67	0.00	-0.02	-0.04	0.00	-0.01	0.02	0.01	0.02	-0.05	-0.08			
1 st E	0.38	-1.94	0.21	-0.67	-1.26	3.45	1.23	-0.39	0.20	0.96	-0.01	0.02	-0.02	-0.06	-0.01	0.03	-0.11	0.01	0.00			
1 st E	0.65	-3.65	-0.67	-0.22	-2.18	1.23	-3.45	0.21	0.39	1.61	0.02	0.01	-0.04	-0.03	-0.07	-0.10	0.01	0.00	0.00			
2 nd A	0.00	0.00	-1.26	-2.18	0.02	-0.46	1.07	2.22	3.36	0.03	-0.07	-0.11	0.01	0.00	0.00	-0.01	0.00	-0.01	-0.01			
2 nd E	0.26	-0.53	3.45	1.23	-0.46	0.18	-1.37	-0.96	0.00	0.46	0.01	0.00	-0.01	0.00	0.00	-0.01	0.01	0.00	0.00			
2 nd E	-0.07	0.06	1.23	-3.45	1.07	-1.37	-0.19	0.00	0.98	-0.45	0.00	-0.01	0.00	0.02	0.00	0.02	0.01	0.01	0.01			
3 rd E	-0.13	-0.23	-0.39	0.21	2.22	-0.96	0.00	1.00	-0.20	0.09	0.00	-0.01	-0.02	0.07	0.01	0.00	0.10	-0.01	0.02			
3 rd E	-0.13	-0.67	0.20	0.39	3.36	0.00	0.98	-0.20	-1.03	0.59	-0.02	-0.01	-0.05	0.05	0.05	0.09	-0.02	0.02	0.00			
3 rd A	0.00	0.00	0.96	1.61	0.03	0.46	-0.45	0.09	0.59	0.00	-0.03	-0.08	0.01	0.00	-0.05	-0.01	-0.03	0.01	-0.02			
4 th E	0.08	-0.02	-0.01	0.02	-0.07	0.01	0.00	0.00	-0.02	-0.03	-0.28	0.64	-0.10	-1.92	-3.19	0.61	0.67	0.09	-0.15			
4 th E	0.15	-0.04	0.02	0.01	-0.11	0.00	-0.01	-0.01	-0.01	-0.08	0.64	0.28	-0.48	-0.94	-0.43	-3.18	-1.99	-0.24	-0.16			
4 th A	-0.01	0.00	-0.02	-0.04	0.01	-0.01	0.00	-0.02	-0.05	0.01	-0.10	-0.48	0.03	0.10	0.12	0.02	0.04	-1.35	-2.67			
5 th A	-0.03	-0.01	-0.06	-0.03	0.00	0.00	0.02	0.07	0.05	0.00	-1.92	-0.94	0.10	-0.03	-0.51	0.04	-0.01	-1.51	-0.70			
6 th E	0.23	0.02	-0.01	-0.07	0.00	0.00	0.00	0.01	0.05	-0.05	-3.19	-0.43	0.12	-0.51	-0.24	-0.07	0.01	-1.64	-0.41			
6 th E	0.05	0.01	0.03	-0.10	-0.01	-0.01	0.02	0.00	0.09	-0.01	0.61	-3.18	0.02	0.04	-0.07	-0.06	-0.39	0.27	-1.50			
16 ES	0.15	0.02	-0.11	0.01	0.00	0.01	0.01	0.10	-0.02	-0.03	0.67	-1.99	0.04	-0.01	0.01	-0.39	0.23	0.22	-0.78			
17 ES	-0.04	-0.05	0.01	0.00	-0.01	0.00	0.01	-0.01	0.02	0.01	0.09	-0.24	-1.35	-1.51	-1.64	0.27	0.22	0.42	-0.43			
18 ES	-0.05	-0.08	0.00	0.00	-0.01	0.00	0.00	0.02	0.00	-0.02	-0.15	-0.16	-2.67	-0.70	-0.41	-1.50	-0.78	-0.43	-0.29			

z component (in a.u.)

	GS(A)	1 st A	1 st E	1 st E	2 nd A	2 nd E	2 nd E	3 rd E	3 rd E	3 rd E	3 rd A	4 th E	4 th E	4 th E	4 th A	5 th A	6 th E	6 th E	16ES	17ES	18ES
GS	-0.44	0.06	0.00	0.00	-0.28	0.00	0.00	0.00	0.00	0.07	-0.05	0.00	-0.35	-0.13	0.00	-0.08	-0.01	0.01	0.00		
1 st A	0.06	-4.13	0.01	0.00	2.62	0.00	0.01	0.00	0.00	-0.76	-0.02	0.00	-0.13	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	
1 st E	0.00	0.01	-1.51	0.00	-0.01	-0.84	0.12	-3.50	0.14	0.00	-0.02	0.00	0.00	-0.02	-0.01	0.01	-0.01	-0.21	0.01		
1 st E	0.00	0.00	0.00	-1.53	0.01	-0.11	-0.84	-0.14	-3.50	-0.03	0.00	-0.01	0.00	-0.01	-0.04	0.01	0.01	0.00	-0.27		
2 nd A	-0.28	2.62	-0.01	0.01	3.84	0.01	0.01	0.03	-0.04	4.35	-0.08	0.00	-0.57	0.11	0.00	0.06	0.00	-0.01	0.00		
2 nd E	0.00	0.00	-0.84	-0.11	0.01	0.34	0.00	-0.04	-0.48	0.00	0.00	-0.01	0.00	0.01	0.02	-0.01	0.01	0.03	-0.02		
2 nd E	0.00	0.01	0.12	-0.84	0.01	0.00	0.35	0.48	-0.03	0.04	0.00	0.00	0.00	0.01	0.00	-0.02	-0.01	0.01	0.04		
3 rd E	0.00	0.00	-3.50	-0.14	0.03	-0.04	0.48	3.93	0.00	-0.05	0.02	0.00	0.00	0.02	0.01	0.00	0.01	0.21	0.01		
3 rd E	0.00	0.00	0.14	-3.50	-0.04	-0.48	-0.03	0.00	3.99	-0.02	0.00	0.01	0.01	0.00	0.05	-0.01	0.00	-0.01	0.28		
3 rd A	0.07	-0.76	0.00	-0.03	4.35	0.00	0.04	-0.05	-0.02	5.70	-0.06	0.00	-0.45	0.16	0.00	0.08	0.00	0.01	0.00		
4 th E	-0.05	-0.02	-0.02	0.00	-0.08	0.00	0.00	0.02	0.00	-0.06	3.63	0.00	-0.01	-0.83	0.16	1.15	0.08	-2.10	0.01		
4 th E	0.00	0.00	0.00	-0.01	0.00	-0.01	0.00	0.00	0.01	0.00	0.00	3.59	0.00	0.08	1.02	-0.16	0.82	-0.02	-2.00		
4 th A	-0.35	-0.13	0.00	0.00	-0.57	0.00	0.00	0.00	0.01	-0.45	-0.01	0.00	3.62	-1.00	0.00	-0.71	-0.05	0.24	0.00		
5 th A	-0.13	0.00	-0.02	-0.01	0.11	0.01	0.01	0.02	0.00	0.16	-0.83	0.08	-1.00	0.23	-0.07	0.00	0.12	-0.30	-0.04		
6 th E	0.00	0.00	-0.01	-0.04	0.00	0.02	0.00	0.01	0.05	0.00	0.16	1.02	0.00	-0.07	0.33	-0.04	0.13	-0.03	0.07		
6 th E	-0.08	0.00	0.01	0.01	0.06	-0.01	-0.02	0.00	-0.01	0.08	1.15	-0.16	-0.71	0.00	-0.04	0.28	0.05	0.44	0.08		
16ES	-0.01	0.00	-0.01	0.01	0.00	0.01	-0.01	0.01	0.00	0.00	0.08	0.82	-0.05	0.12	0.13	0.05	0.16	-0.02	0.19		
17ES	0.01	-0.01	-0.21	0.00	-0.01	0.03	0.01	0.21	-0.01	0.01	-2.10	-0.02	0.24	-0.30	-0.03	0.44	-0.02	-2.18	0.00		
18ES	0.00	0.00	0.01	-0.27	0.00	-0.02	0.04	0.01	0.28	0.00	0.01	-2.00	0.00	-0.04	0.07	0.08	0.19	0.00	-1.56		

Table S2 : Coefficients of the CSF's larger than |0.05| for the lowest 10 states (below the IP) of the 19 state CAS average (8,13)/6-31++G(d,p) computation at the geometry of the neutral GS. The largest coefficients are written in bold. The convention for the occupation of the 13 active MO's in the first column is the following : 2 means doubly occupied, / means spin α and \ means spin β .

CSF	Ground state	1 st A	1 st E	1 st E	2 nd A	2 nd E	2 nd E	3 rd E	3 rd E	3 rd A)
2222000000000	0.997	0.029	0.000	0.000	0.047	0.000	-0.001	0.001	0.000	0.036
/222\000000000	-0.016	0.002	0.000	0.000	0.032	0.000	0.000	0.000	0.001	-0.047
22/2\000000000	-0.001	0.000	-0.003	0.000	0.002	-0.001	-0.002	-0.006	-0.001	-0.003
2/22\000000000	0.000	0.000	0.000	-0.004	0.000	-0.002	0.001	0.001	-0.007	0.000
222/0000\0000	0.002	-0.003	0.102	0.079	-0.008	-0.013	0.950	-0.181	-0.213	-0.022
222/00000\000	-0.001	-0.002	-0.080	0.107	0.006	0.948	0.012	0.216	-0.187	0.007
2/2200\000000	-0.004	-0.001	0.001	0.000	0.000	0.006	0.002	0.002	0.000	-0.002
22/20\0000000	0.004	0.001	0.001	0.001	0.000	0.006	0.002	0.003	0.001	0.002
2/220\0000000	-0.013	-0.004	0.000	-0.001	-0.008	-0.002	0.006	0.000	-0.003	-0.005
22/200\000000	-0.013	-0.004	0.001	0.001	-0.008	0.002	-0.005	0.002	0.002	-0.005
222/\000000000	-0.020	0.188	-0.002	0.001	0.782	0.002	-0.004	-0.001	0.007	-0.589
22/2000\00000	0.000	0.000	-0.007	0.001	0.000	-0.001	-0.005	-0.002	0.000	0.001
222/00\0000000	0.002	-0.023	0.724	0.032	-0.009	-0.100	0.043	0.678	-0.012	-0.024
222/000\00000	-0.058	0.724	0.033	-0.001	0.292	-0.007	0.015	0.012	-0.010	0.619
/2220\0000000	0.000	0.000	0.000	0.004	0.000	0.001	0.002	0.000	0.021	0.000
222/0\0000000	0.000	0.002	-0.032	0.720	0.000	0.046	0.100	0.013	0.682	0.010
222/00000000\	0.000	-0.001	-0.017	-0.678	0.001	0.187	0.225	0.075	0.668	0.006
222/00000000\0	-0.001	-0.059	0.672	-0.016	0.040	0.227	-0.178	-0.671	0.072	0.035
222/0000000\00	0.025	0.658	0.049	-0.002	-0.543	0.027	-0.026	-0.050	0.012	-0.511
2/22000\00000	0.000	0.000	-0.001	-0.007	0.000	-0.005	0.001	0.000	-0.002	0.000
/22200\000000	0.000	0.000	0.004	0.000	0.000	-0.002	0.001	0.021	-0.001	-0.001

/222000\00000	0.015	-0.005	0.000	0.000	0.001	0.000	0.000	0.001	0.000	0.013
2/220000\0000	-0.003	0.000	0.001	0.005	0.000	0.001	0.001	0.001	0.002	0.000
22/200000\000	0.003	0.000	0.001	0.004	0.000	0.002	0.001	0.001	0.001	0.000
2/220000\000	0.008	-0.001	0.005	-0.001	-0.002	-0.001	0.001	0.002	-0.001	-0.002
22/20000\0000	0.008	-0.001	-0.004	0.001	-0.002	0.001	-0.001	-0.002	0.000	-0.002
/2220000\0000	-0.001	0.000	-0.001	-0.001	0.000	0.000	-0.001	-0.001	-0.002	0.000

Table S3

	Excitation energy (eV)	Symmetry	Permanent dipole (a.u.)			transition dipole (a.u.)		
			x	y	z	x	y	z
GS			0.00	0.00	-0.17			
ES 1	3.76	A	0.00	0.00	-3.95	0.00	0.00	0.36
ES 2	4.13	E	-0.56	-0.66	-1.71	-0.14	0.49	0.00
ES 3	4.13	E	0.56	0.66	-1.72	0.49	0.14	0.00
ES 4	4.26	A	0.00	0.00	2.24	0.00	0.00	-0.11
ES 5	4.74	E	0.34	-1.25	-0.96	-0.15	0.21	0.00
ES 6	4.74	E	-0.34	1.24	-0.96	0.21	0.15	0.00
ES 7	4.79	A	0.00	0.00	3.26	0.00	0.00	0.05
ES 8	4.89	E	0.39	-1.02	1.40	0.04	0.04	0.00
ES 9	4.89	E	-0.41	1.02	1.40	-0.04	0.04	0.00

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