Supporting Information

Study of double core hole excitations in molecules by X-ray double-quantum-coherence signals: a multi-configuration simulation

Weijie Hua,[ab] Kochise Bennett,[a] Yu Zhang,[a] Yi Luo,[bc] and Shaul Mukamel[*a]

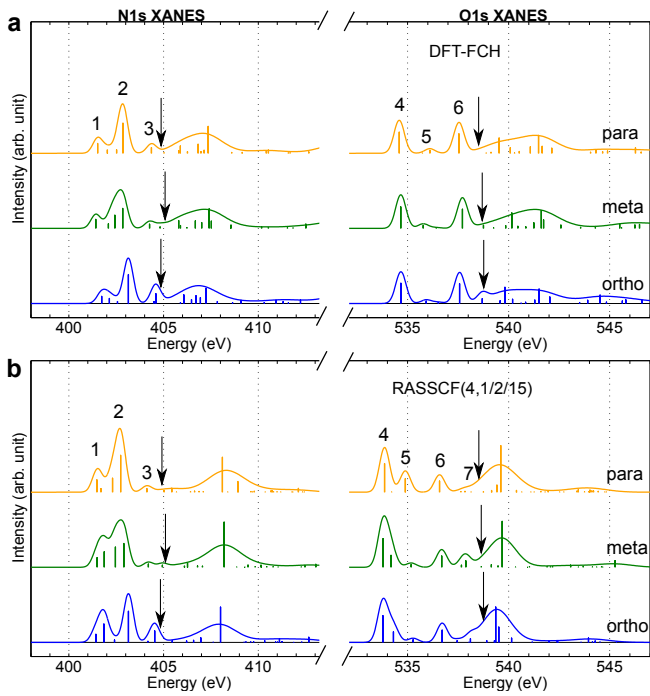**This PDF file contains**

[a] Department of Chemistry, University of California, Irvine, CA 92697-2025, United States. E-mail: smukamel@uci.edu

[b] Department of Theoretical Chemistry and Biology, School of Biotechnology, KTH Royal Institute of Technology, S-10691 Stockholm, Sweden.
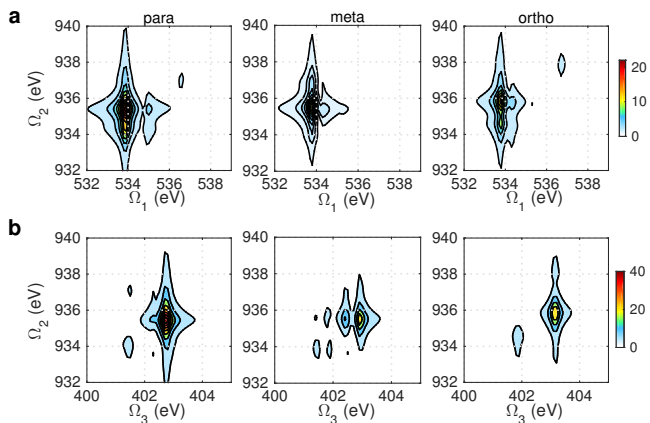
[c] Hefei National Laboratory for Physical Sciences at the Microscale, University of Science and Technology of China, Hefei, Anhui 230026, China.

Table S1 RASSCF energy levels (in eV) of p-aminophenol selected for XDQC signal simulations. e^N and e^O are N1s and O1s SCH excited states, f^{NN}, f^{OO}, f^{NO} are DCH excited states. Shifts had been applied according to the text.
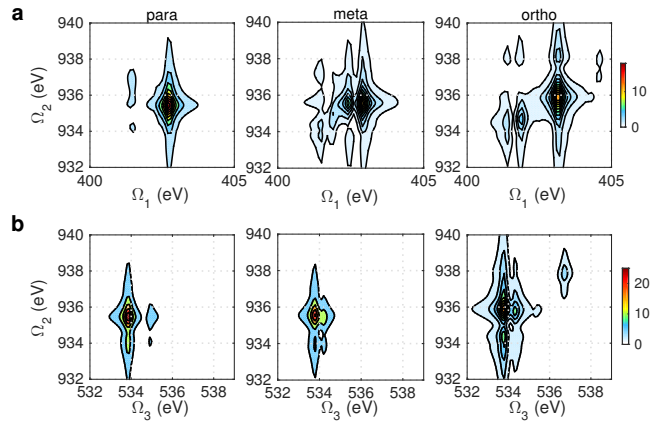
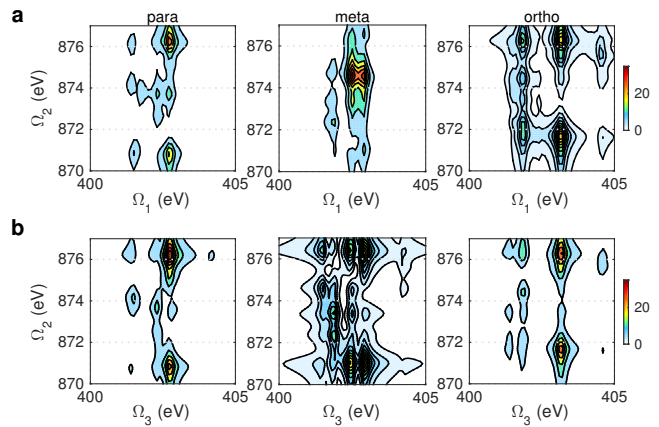| e_i^N	Energy	f_i^{NN}	Energy	f_i^{NO}	Energy
1	401.4822	1	870.8938	1	933.0650
2	401.6993	2	872.6560	2	933.3127
3	402.3080	3	872.7349	3	933.5179
4	402.7381	4	873.6410	4	933.5962
5	404.1175	5	873.6956	5	933.7148
		6	874.2115	6	933.8504
e_i^O	Energy	7	875.1804	7	934.0713
1	533.8622	8	875.9051	8	934.1381
2	534.2132	9	876.0974	9	934.3231
3	534.8883	10	876.1897	10	934.3720
4	536.5819			11	934.5522
5	537.6403	f_i^{OO}	Energy	12	934.7414
6	537.8379	1	1148.2804	13	934.8674
7	538.1405	2	1148.5340	14	935.0193
		3	1148.8677	15	935.3065
		4	1150.7172	16	935.3953
		5	1152.1360	17	935.4408
		6	1152.3517	18	935.5753
		7	1152.9617	19	935.9372
		8	1153.0162	20	935.9951
		9	1153.4704	21	936.0142
		10	1154.3092	22	936.2642
				23	936.6959
				24	936.7117
				25	936.8616
				26	936.9273
				27	936.9564
				28	937.0654
				29	937.4285
				30	937.8068

Figure S1 N1s and O1s XANES spectra of p-, m-, and o-aminophenols calculated at the (a) DFT-FCH and (b) RASSCF(4, 1/2/15) levels using a different convolution scheme than the main text, which better resolve the spectra above the ionic potentials (IP). Gaussian line shape was used with hwhm = 0.3 eV below the IP, linearly increasing hwhm in the next 1 eV, and hwhm = 1.0 eV in the region beyond. Arrows mark the calculated IP by the DFT-FCH method. Major peaks below the IP are labeled.
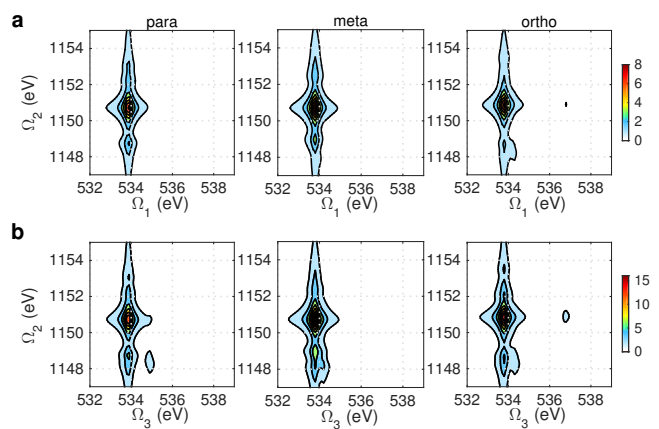

Figure S2 ON-A XDQC signals (absolute values) of p-, m-, and o-aminophenols (left to right) (see Table 1 of the text for notations). (a) $S_{III}(t_3 = 6.1\text{ fs}, \Omega_2, \Omega_1)$, (b) $S_{III}(\Omega_3, \Omega_2, t_1 = 0\text{ fs})$.

Figure S3 NO-A XDQC signals (absolute values) of p-, m-, and o-aminophenols (left to right). (a) $S_{III}(t_3 = 6.1\text{ fs}, \Omega_2, \Omega_1)$, (b) $S_{III}(\Omega_3, \Omega_2, t_1 = 0\text{ fs})$.

Figure S4 NN-A XDQC signals (absolute values) of p-, m-, and o-aminophenols (left to right). (a) $S_{III}(t_3 = 6.1\text{ fs}, \Omega_2, \Omega_1)$, (b) $S_{III}(\Omega_3, \Omega_2, t_1 = 0\text{ fs})$.

Figure S5 OO-A XDQC signals (absolute values) of p-, m-, and o-aminophenols (left to right). (a) $S_{III}(t_3 = 6.1\text{ fs}, \Omega_2, \Omega_1)$, (b) $S_{III}(\Omega_3, \Omega_2, t_1 = 0\text{ fs})$.