Supporting Information

for

Electronic Structure of Liquid Methanol and Ethanol from Polarization-Dependent Two-Photon Absorption Spectroscopy

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TABLE S1. Term labels, excitation energies (E_{ex}), 1PA oscillator strength (f_L), microscopic (β) and macroscopic (σ) 2PA cross sections for parallel polarization, and polarization ratio of methanol. The pump energy used for this calculation is 4.6 eV (266 nm). The level of theory used EOM-CCSD/d-aug-cc-PVDZ

States	E_{ex} (eV)	f_L ($\times 10^{-3}$)	β_{2PA} (atomic unit)	σ_{2PA} (GM)	Polarization ratio
1A"	6.73	4.0	20	0.67	1.3
2A"	7.90	30.0	24	1.08	1.3
2A'	8.31	26.6	4	0.22	2.4
3A"	8.33	6.4	2	0.08	1.3
3A'	8.75	1.2	20	1.13	1.8
4A"	8.95	0.2	26	1.56	1.3
5A"	9.14	0.8	8	0.51	1.3
4A'	9.28	1.0	18	1.16	2.1
6A"	9.30	0.9	14	0.89	1.3
7A"	9.38	0.1	17	1.07	1.3
5A'	9.44	0.5	197	12.92	12.9
8A"	9.49	5.6	0.1	0.005	1.3
6A'	9.63	22.0	27	1.82	6.9
7A'	9.68	44.3	9	0.05	1.3
9A"	9.68	2.3	1	0.60	1.6
10A"	9.98	2.1	4	0.30	1.3
8A'	10.15	36.3	12	0.89	11.4
11A"	10.17	2.4	0.05	0.003	1.0
12A"	10.20	10.4	2	0.19	1.3
13A"	10.42	0.5	3	0.23	1.2
9A'	10.47	10.7	7	0.58	5.3
14A"	10.50	0.7	0.3	0.03	1.2

TABLE S2. Term labels, excitation energies (E_{ex}), 1PA oscillator strength (f_L), microscopic (β) and macroscopic (σ) 2PA cross sections for parallel polarization, and polarization ratio of ethanol. The pump energy used for this calculation is 4.6 eV (266 nm). The level of theory used EOM-CCSD/d-aug-cc-PVDZ

States	E_{ex} (eV)	f_L ($\times 10^{-3}$)	β_{2PA} (atomic unit)	σ_{2PA} (GM)	Polarization ratio
1A"	**6.73**	**2.8**	**24**	**0.81**	**1.3**
2A"	**7.87**	**17.0**	**30**	**1.37**	**1.3**
2A'	**8.07**	**27.6**	**5**	**0.24**	**1.3**
3A"	**8.15**	**19.3**	**9**	**0.44**	**1.3**
3A'	**8.67**	**0.9**	**49**	**2.70**	**6.4**
4A"	**8.85**	**0.6**	**1**	**0.06**	**1.3**
5A"	**8.91**	**1.1**	**9**	**0.53**	**1.3**
6A"	**9.03**	**0.8**	**16**	**0.96**	**1.3**
7A"	**9.08**	**2.6**	**6**	**1.36**	**2.2**
4A'	**9.08**	**0.1**	**23**	**0.38**	**1.3**
5A'	**9.11**	**0.3**	**138**	**8.43**	**13.8**
8A"	**9.33**	**3.7**	**0.1**	**0.01**	**1.3**
6A'	**9.38**	**10.6**	**3**	**0.22**	**22.2**
9A"	**9.44**	**6.8**	**1**	**0.05**	**1.3**
7A'	**9.47**	**17.6**	**125**	**8.23**	**72.1**
10A"	**9.71**	**2.6**	**2**	**0.17**	**1.3**
11A"	**9.74**	**31.9**	**2**	**0.15**	**1.3**
8A'	**9.77**	**101.1**	**17**	**1.22**	**8.4**
12A"	**9.79**	**0.2**	**7**	**0.48**	**1.3**

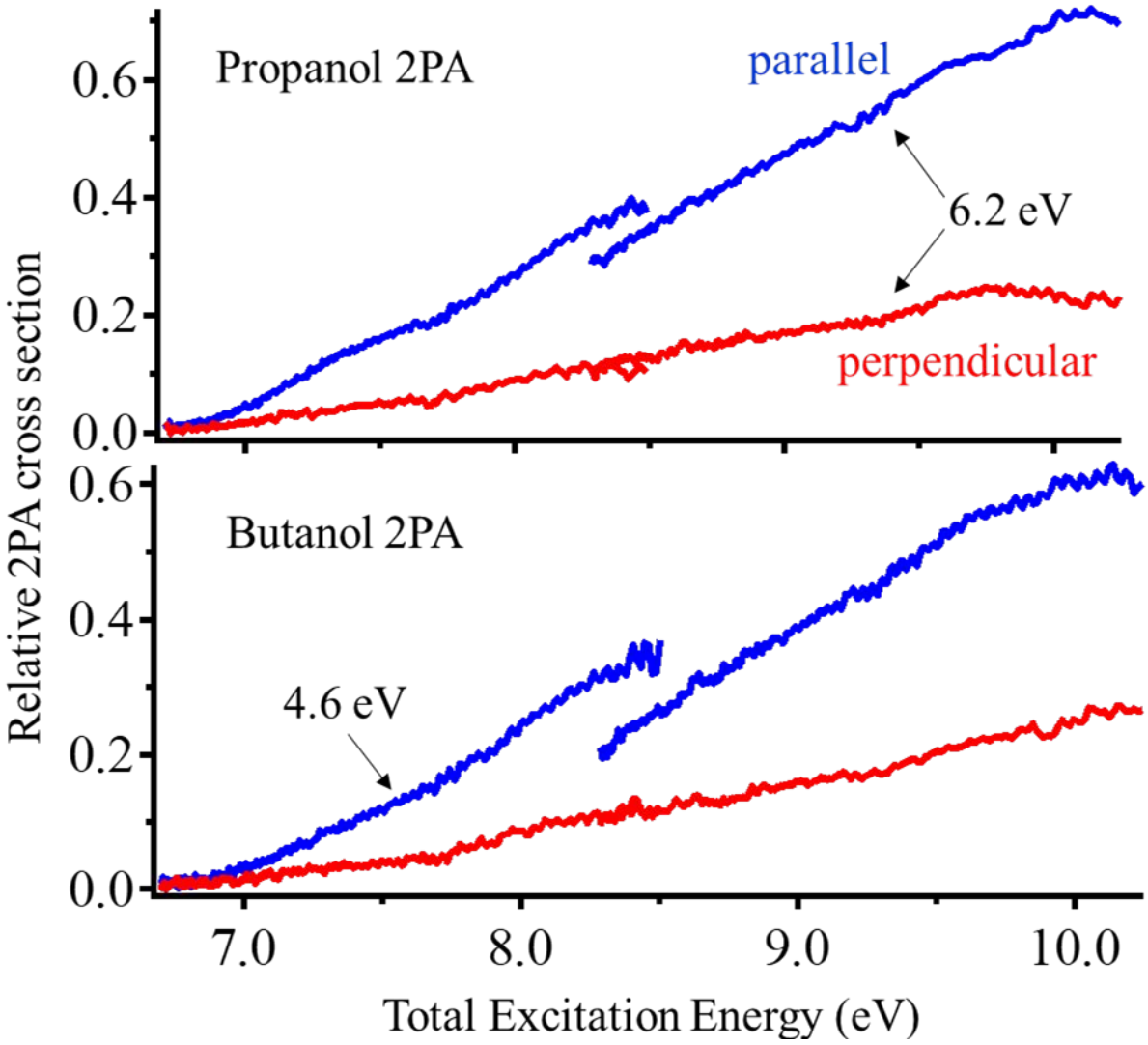

Fig. S1. Liquid phase 2PA spectra of propanol and butanol. Spectra recorded with both a 4.6 eV pump and a 6.2 eV pump are shown in the same figure; parallel (blue) and perpendicular polarization (red).

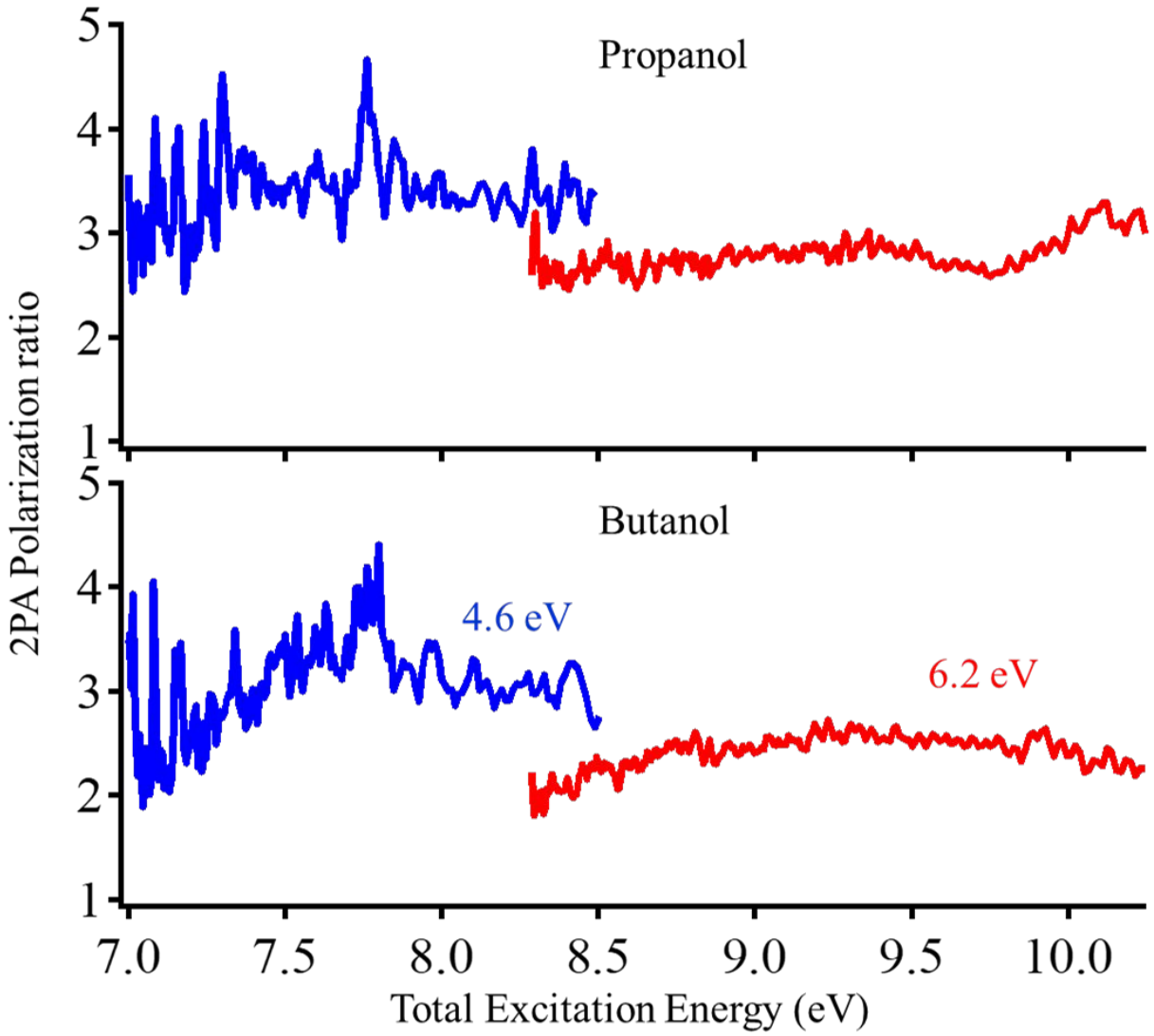

Fig. S2. The experimental polarization ratio (ρ) for propanol and butanol plotted against the total excitation energy with (blue) 4.6 eV and (red) 6.2 eV pump.