

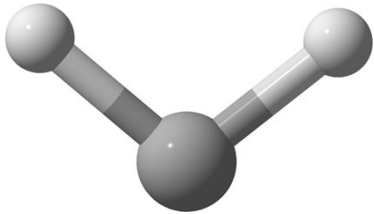
Strong-field ionization of polyatomic molecules: Ultrafast H atom migration and bond formation
in photo dissociation of CH₃OH

Rituparna Das, Deepak K. Pandey, Vinitha Nimma, Madhusudhan P, Pranav Bhardwaj, Pooja Chandravanshi, Muhammed Shameem K M, Dheeraj K. Singh, and Rajesh K Kushawaha

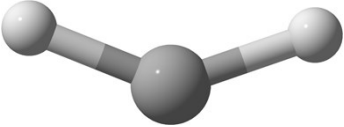
Supplementary Information

Figure S1. CYLview generated structures, total electronic energy, zero-point energy (ZPE) correction, sum of electronic energy and ZPE, HOMO and LUMO energy levels, and HOMO-LUMO energy gaps (E_{gap}) for various isomers along dissociation pathway of CH_3OH^+ calculated at the M06-2X/6-311++G(d,P) level of theory. Energy values are in Hartree.

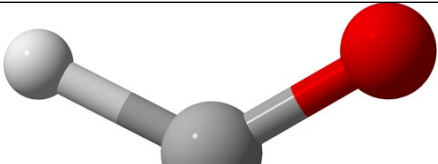
CH_2

	Electronic Energy (EE) = -39.118961
	Zero-point Energy Correction = 0.016863
	EE + Zero-point Energy = -39.102098
	$E_{HOMO} = -0.31591$
	$E_{LUMO} = -0.08391$
	$E_{gap} = 0.232$

CH_2^+

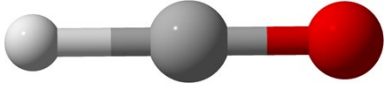
	Electronic Energy (EE) = -38.760619
	Zero-point Energy Correction = 0.016695
	EE + Zero-point Energy = -38.743924
	$E_{HOMO} = -0.72494$
	$E_{LUMO} = -0.45286$
	$E_{gap} = 0.27208$

CHO

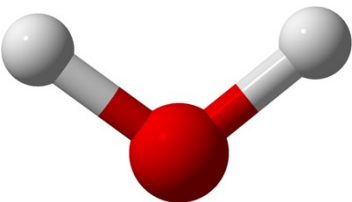
	Electronic Energy (EE) = -113.838141
	Zero-point Energy Correction = 0.013290
	EE + Zero-point Energy = -113.824850
	$E_{HOMO} = -0.28143$

	$E_{LUMO} = -0.01104$
	$E_{gap} = 0.27039$

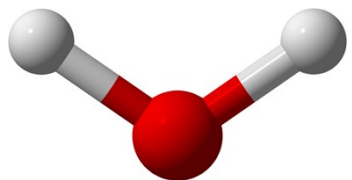
CHO⁺

	Electronic Energy (EE) = -113.536884
	Zero-point Energy Correction = 0.016733
	EE + Zero-point Energy = -113.520150
	$E_{HOMO} = -0.90407$
	$E_{LUMO} = -0.32001$
	$E_{gap} = 0.58406$


H₂O

	Electronic Energy (EE) = -76.420906
	Zero-point Energy Correction = 0.021617
	EE + Zero-point Energy = -76.399289
	$E_{HOMO} = -0.40086$
	$E_{LUMO} = -0.00722$
	$E_{gap} = 0.39364$

H₂O⁺

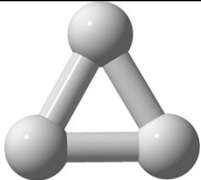
	Electronic Energy (EE) = -75.956109
	Zero-point Energy Correction = 0.018796
	EE + Zero-point Energy = -75.937314
	$E_{HOMO} = -0.94169$
	$E_{LUMO} = -0.26203$
	$E_{gap} = 0.67966$

H₃

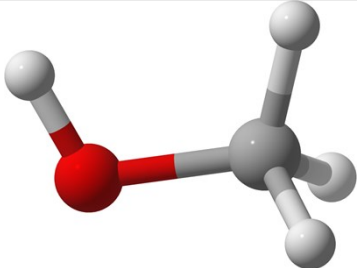
	Electronic Energy (EE) = -1.665920
	Zero-point Energy Correction = 0.011806
	EE + Zero-point Energy = -1.654114

	$E_{HOMO} = -0.06936$
	$E_{LUMO} = -0.03232$
	$E_{gap} = 0.03704$

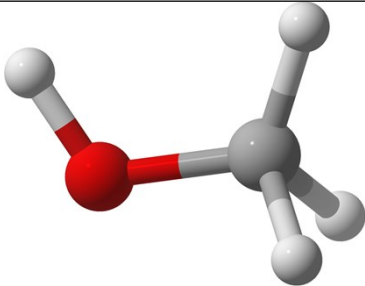
H₃⁺

	Electronic Energy (EE) = -1.343087
	Zero-point Energy Correction = 0.023260
	EE + Zero-point Energy = -1.319827
	$E_{HOMO} = -1.10821$
	$E_{LUMO} = -0.20041$
	$E_{gap} = 0.9078$

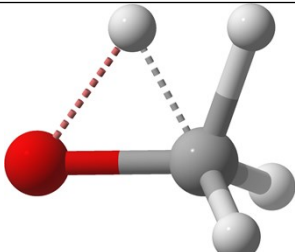
CH₃OH

	Electronic Energy (EE) = -115.704311 Hartree
	Zero-point Energy Correction = 0.051863
	EE + Zero-point Energy = -115.652448
	$E_{HOMO} = -0.35464$
	$E_{LUMO} = -0.0032$
	$E_{gap} = 0.35144$

CH₃OH⁺

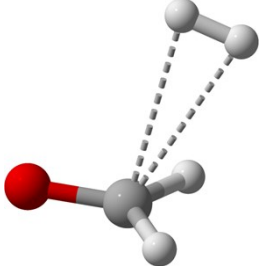
	Electronic Energy (EE) = -115.305553
	Zero-point Energy Correction = 0.047293
	EE + Zero-point Energy = -115.258260
	$E_{HOMO} = -0.34074$
	$E_{LUMO} = -0.01281$
	$E_{gap} = 0.32793$

TS1

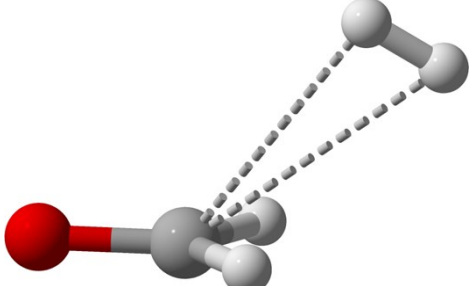
	Electronic Energy (EE) = -115.197528
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	Zero-point Energy Correction = 0.042678
	EE + Zero-point Energy = -115.154850
	$E_{HOMO} = -0.70651$
	$E_{LUMO} = -0.25731$
	$E_{gap} = 0.4492$

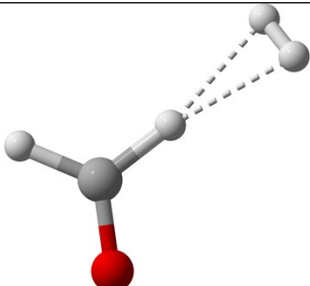
S1

	Electronic Energy (EE) = -115.256228
	Zero-point Energy Correction = 0.037284
	EE + Zero-point Energy = -115.218944
	$E_{HOMO} = -0.69907$
	$E_{LUMO} = -0.32273$
	$E_{gap} = 0.37634$

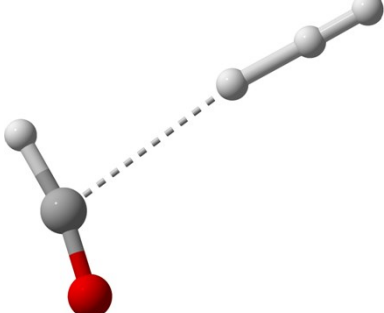
TS2

	Electronic Energy (EE) = -115.254612
	Zero-point Energy Correction = 0.035791
	EE + Zero-point Energy = -115.218821
	$E_{HOMO} = -0.67889$
	$E_{LUMO} = -0.329$
	$E_{gap} = 0.34989$

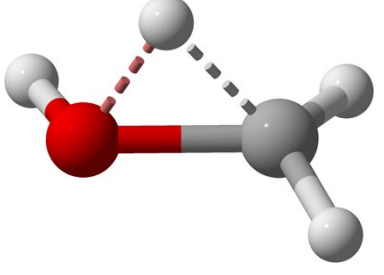
S2

	Electronic Energy (EE) = -115.257014
	Zero-point Energy Correction = 0.036751
	EE + Zero-point Energy = -115.220263
	$E_{HOMO} = -0.69221$
	$E_{LUMO} = -0.32079$
	$E_{gap} = 0.37142$

TS3

	Electronic Energy (EE) = -115.199333
	Zero-point Energy Correction = 0.029736
	EE + Zero-point Energy = -115.169597
	$E_{HOMO} = -0.50376$
	$E_{LUMO} = -0.30467$
	$E_{gap} = 0.19909$

TS4

	Electronic Energy (EE) = -115.259050
	Zero-point Energy Correction = 0.045320
	EE + Zero-point Energy = -115.213730
	$E_{HOMO} = -0.59678$
	$E_{LUMO} = -0.23125$
	$E_{gap} = 0.36553$

CH₂OH₂⁺

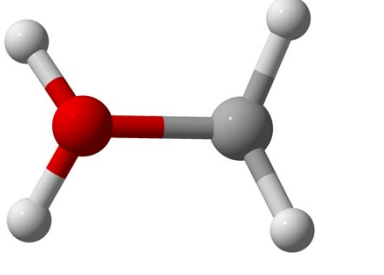
	Electronic Energy (EE) = -115.317304
	Zero-point Energy Correction = 0.049812
	EE + Zero-point Energy = -115.267492
	$E_{HOMO} = -0.54954$
	$E_{LUMO} = -0.2216$
	$E_{gap} = 0.32794$

Figure 3 (a): The table below shows H^+ , H_2^+ , H_3^+ , and total counts for CH_3OH (800 nm, 29 fs).

Linear polarization

Laser intensity ($\times 10^{13}$ W/cm²)	H^+	H_2^+	H_3^+	Total counts
1.464	3626	742	281	57580
2.074	48972	12303	4177	494767
2.684	126167	38038	14159	615655
3.294	117697	38844	13531	429667
3.904	297870	110973	38968	1.06562E6
4.514	637358	248615	86673	2.16302E6
5.124	1.39961E6	559553	192061	4.50857E6
5.734	436404	174967	59956	1.29985E6
6.344	914395	365239	123704	2.74564E6
6.954	1.7041E6	709146	246124	4.7542E6
7.564	146423	57942	20240	477268
8.174	166130	70747	24706	485537
8.784	763532	321847	112155	1.94869E6
9.394	267345	116523	41773	690857
10.004	996926	453612	167727	2.4285E6
10.614	756418	345990	129554	1.82521E6
11.224	234085	107725	41127	580155
11.834	147536	69298	26885	361262
12.444	238141	116818	46919	559954

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Circular polarization

Laser intensity (X10 ¹³ W/cm ²)	H ⁺	H ₂ ⁺	H ₃ ⁺	Total counts
1.464	23521	1056	749	832632
2.074	95333	24514	10295	944037
2.684	90093	20460	8322	886558
3.294	149205	40055	14896	804020
3.904	460022	127666	44194	1.82188E6
4.514	690632	199429	64504	2.48267E6
5.124	695254	211579	66464	2.23595E6
5.734	659170	219357	68682	1.99104E6
6.344	529268	192135	56079	1.54919E6
6.954	340189	123950	38092	933879
7.564	3.66982E6	1.44678E6	459210	9.39042E6
8.174	2.81924E6	1.06966E6	326906	7.20384E6
8.784	3.49724E6	1.40248E6	444396	8.74879E6
9.394	695640	299123	96616	1.71949E6
10.004	1.27317E6	560081	180619	3.13663E6
10.614	534921	240183	77584	1.31568E6
11.224	575806	271274	88072	1.39402E6
11.834	1.02341E6	476726	156137	2.47214E6
12.444	832949	420555	143977	2.01126E6

Figure 3 (c): The table below shows H⁺, D⁺, HD⁺, D₂⁺, D₂H⁺, D₃⁺, and total counts for CD₃OH (800 nm, 29 fs).

Laser intensity (X10¹³ W/cm²)	H⁺	D⁺	HD⁺	D₂⁺	D₂H⁺	D₃⁺	Total counts
1.464	33102	25929	0	0	0	0	700649
2.074	97749	106037	1485	16601	6431	0	1.4428E6
2.684	152703	159505	6081	27174	20554	0	1.58002E6
3.294	130946	139714	4932	23883	11515	0	1.11108E6
3.904	349975	345460	12835	62359	39067	3849	2.37841E6
4.514	205006	206923	6580	36901	22945	2480	1.2883E6
5.124	214181	228394	8100	38815	28349	4762	1.2436E6
5.734	269618	305686	10726	50394	44822	8358	1.40975E6
6.344	131601	159710	5255	25284	27848	6791	590096
6.954	82900	107939	3601	16887	22113	5524	324828
7.564	107185	132914	4085	19199	25054	6361	452045
8.174	127818	162478	4956	22170	34852	9520	497072
8.784	115120	150464	4389	20229	36186	8660	414016
9.394	94407	125190	3715	16892	30060	7043	352426
10.004	158336	209898	6216	27708	47476	10427	508307
10.614	97223	132454	3843	16384	26973	4798	298237
11.224	130313	176933	5253	21706	32403	5313	390320

11.834	141074	190763	5395	22312	29512	4134	406850
12.444	147527	198637	5622	22332	28228	3546	418427

Figure 3 (c): The table below shows H_2^+ , H_3^+ , and total counts for CH_3OH (1300 nm, 29 fs).

Laser intensity ($\times 10^{13}$ W/cm²)	H_2^+	H_3^+	Total counts
2.074	598	288	80770
3.294	13731	4044	873513
4.514	57185	14921	2.42786E6
5.734	205304	54030	3.12504E6
6.954	58285	15043	2.96569E6
8.174	32356	8626	3.3797E6
9.394	16202	4162	1.29868E6
10.614	22977	6627	1.44781E6
11.834	13256	3526	548997