

Table S1: Changes Chemical Shifts upon Ionization of 4-Substituted Benzyltriflones **5a-e** Me₂SO solution.^a

Compound	δH_{α}	$\delta H_{2,6}$	$\delta H_{3,5}$	δH_4	δC_{α}	δC_1	$\delta C_{2,6}$	$\delta C_{3,5}$	δC_4	SO_2CF_3
<u>5a</u>	5.25	7.47	7.47	7.47	54.80	124.60	131.69	128.98	129.61	119.59
<u>C-5a</u>	3.38	6.91	6.91	6.46	55.02	142.18	120.31	127.74	116.85	124.33
<u>5b</u>	5.44	7.72	7.85	-	54.02	129.52	132.37	125.67	130.02	119.36
<u>C-5b</u>	3.61	7.14	7.03	-	57.65	146.93	119.07	124.52	115.89	123.77
<u>5c</u>	5.46	7.69	7.96	-	54.31	130.41	132.42	132.70	112.43	119.46
<u>C-5c</u>	3.69	6.95	7.17	-	60.20	147.91	119.15	131.30	95.58	123.22
<u>5d</u>	5.53	7.78	8.33	-	53.74	132.33	132.53	132.53	148.14	119.21
<u>C-5d</u>	4.07	6.95	7.75	-	65.72	151.29	117.90	124.33	131.45	122.62
<u>5e</u>	5.56	7.90	8.22	-	53.82	135.20	135.57	131.16	130.48	119.27
<u>C-5e</u>	4.06	7.07	7.36	-	65.15	152.78	118.80	130.39	110.06	122.62

(a) δ in ppm relative to internal SiMe₄, J in Hz; Solvent [¹H₂] Me₂SO; (b) Ref. 20.

Table S₂: Changes Chemical Shifts upon Ionization of substituted Benzyltriflones **9d-f** in Me₂SO solution.^a

Compound	δH_{α}	$\delta H_{2,6}$	$\delta H_{3,5}$	δC_{α}	δC_1	$\delta C_{2,6}$	$\delta C_{3,5}$	δC_4	SO_2CF_3
<u>9d</u>	5.79	5.79	5.79	47.70	118.70	150.60	127.53	132.00	118.80
C-9d	4.98	4.98	4.98	59.39	133.60	143.72	125.88	113.10	121.04
<u>9e^b</u>	5.83	5.83	5.83	48.96	121.03	151.37	126.00	149.27	119.90
C-9e ^b	5.82	5.82	5.82	73.46	134.80	139.78	125.26	130.70	120.18
<u>9f</u>	5.84	5.84	5.84	48.13	123.17	151.37	131.68	133.32	119.04
C-9f	5.87	5.87	5.87	76.85	135.50	140.49	130.79	106.22	120.07

(a) δ in ppm relative to internal SiMe₄, J in Hz; Solvent [¹H₂] Me₂SO; (b) Ref. 20.

Table S3: Changes Chemical Shifts upon Ionization of substituted Benzyltriflones **9a-c** in Me₂SO solution.^a

Compound	δH_{α}	δH_3	δH_5	δH_6	δC_{α}	δC_1	δC_2	δC_3	δC_4	δC_5	δC_6	SO ₂ CF ₃
<u>9a</u>	5.80	8.54	8.31	8.06	51.97	119.95	148.87	125.90	131.54	134.40	134.69	118.80
<u>C-9a</u>	5.52	8.10	7.34	7.95	66.22	139.83	136.69	125.80	115.41	131.98	122.48	121.04
<u>9b</u> ^b	5.87	8.89	8.72	8.08	51.99	124.64	148.79	121.05	148.42	128.39	136.05	119.90
<u>C-9b</u> ^b	5.78	8.70	7.84	7.84	72.25	143.83	143.91	121.39	134.12	123.89	125.01	120.18
<u>9c</u>	5.91	8.44	8.70	8.26	52.03	129.72	149.53	127.79	132.16	135.46	137.06	119.04
<u>C-9c</u>	5.74	7.61	7.94	7.46	73.15	135.20	144.96	131.61	100.27	122.63	129.54	120.07

(a) δ in ppm relative to internal SiMe₄, J in Hz; Solvent [¹H₂]Me₂SO; (b) Ref. 20.