Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Size dependence of optical nonlinearity for H-capped carbon chains, H-(C≡C)$_n$-H (n = 3-15): Analysis of its nature and prediction for long chains

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Table S1 Optimized Cartesian coordinates (in Å) of the carbon chains H-(C≡C)$_n$-H (n = 10-14) at the ωB97XD/def2-TZVP level

H-(C≡C)$_{10}$-H (Charge = 0, Spin multiplicity = 1)			
Atom	x	y	z
C	0.00000000	0.00000000	-10.94314807
C	0.00000000	0.00000000	-12.14485568
C	0.00000000	0.00000000	-9.58024794
C	0.00000000	0.00000000	-8.37065151
C	0.00000000	0.00000000	-7.01645174
C	0.00000000	0.00000000	-5.80447148
C	0.00000000	0.00000000	-4.45251089
C	0.00000000	0.00000000	-3.23980008
C	0.00000000	0.00000000	-1.88853814
C	0.00000000	0.00000000	-0.67561848
C	0.00000000	0.00000000	0.67549497
C	0.00000000	0.00000000	1.88839776
C	0.00000000	0.00000000	3.23972262
C	0.00000000	0.00000000	4.45238032
C	0.00000000	0.00000000	5.80447915
C	0.00000000	0.00000000	7.01637416
C	0.00000000	0.00000000	8.37077364
C	0.00000000	0.00000000	9.58032176
C	0.00000000	0.00000000	10.94330074
C	0.00000000	0.00000000	12.14501692
H	0.00000000	0.00000000	13.20906425
H	0.00000000	0.00000000	-13.20887227

H-(C≡C)$_{11}$-H (Charge = 0, Spin multiplicity = 1)			
Atom	x	y	z

C	0.00000000	0.00000000	-9.65249682
C	0.00000000	0.00000000	-10.86209055
C	0.00000000	0.00000000	-12.22497740
C	0.00000000	0.00000000	-13.42667887
C	0.00000000	0.00000000	-8.29831626
C	0.00000000	0.00000000	-7.08633605
C	0.00000000	0.00000000	-5.73440825
C	0.00000000	0.00000000	-4.52168882
C	0.00000000	0.00000000	-3.17049121
C	0.00000000	0.00000000	-1.95753941
C	0.00000000	0.00000000	-0.60656497
C	0.00000000	0.00000000	0.60643347
C	0.00000000	0.00000000	1.95742985
C	0.00000000	0.00000000	3.17035609
C	0.00000000	0.00000000	4.52162688
C	0.00000000	0.00000000	5.73428924
C	0.00000000	0.00000000	7.08635886
C	0.00000000	0.00000000	8.29825239
C	0.00000000	0.00000000	9.65263298
C	0.00000000	0.00000000	10.86217760
C	0.00000000	0.00000000	12.22514356
C	0.00000000	0.00000000	13.42685350
H	0.00000000	0.00000000	14.49090850
H	0.00000000	0.00000000	-14.49070324

H-(C≡C)$_{12}$-H (Charge = 0, Spin multiplicity = 1)			
Atom	*x*	*y*	*z*
C	0.00000000	0.00000000	-8.36819084
C	0.00000000	0.00000000	-9.58016718

C	0.00000000	0.00000000	-10.93433954
C	0.00000000	0.00000000	-12.14393004
C	0.00000000	0.00000000	-13.50680948
C	0.00000000	0.00000000	-14.70850577
C	0.00000000	0.00000000	-7.01627812
C	0.00000000	0.00000000	-5.80355949
C	0.00000000	0.00000000	-4.45239165
C	0.00000000	0.00000000	-3.23943170
C	0.00000000	0.00000000	-1.88851899
C	0.00000000	0.00000000	-0.67548881
C	0.00000000	0.00000000	0.67537100
C	0.00000000	0.00000000	1.88839256
C	0.00000000	0.00000000	3.23933718
C	0.00000000	0.00000000	4.45226792
C	0.00000000	0.00000000	5.80351166
C	0.00000000	0.00000000	7.01617270
C	0.00000000	0.00000000	8.36822544
C	0.00000000	0.00000000	9.58011633
C	0.00000000	0.00000000	10.93448459
C	0.00000000	0.00000000	12.14402614
C	0.00000000	0.00000000	13.50698289
C	0.00000000	0.00000000	14.70868791
H	0.00000000	0.00000000	15.77274874
H	0.00000000	0.00000000	-15.77253680

H-(C≡C)$_{13}$-H (Charge = 0, Spin multiplicity = 1)			
Atom	*x*	*y*	*z*
C	0.00000000	0.00000000	-7.08541936
C	0.00000000	0.00000000	-8.29813163

C	0.00000000	0.00000000	-9.65004382
C	0.00000000	0.00000000	-10.86201181
C	0.00000000	0.00000000	-12.21618940
C	0.00000000	0.00000000	-13.42577567
C	0.00000000	0.00000000	-14.78865495
C	0.00000000	0.00000000	-15.99034642
C	0.00000000	0.00000000	-5.73426236
C	0.00000000	0.00000000	-4.52130443
C	0.00000000	0.00000000	-3.17041864
C	0.00000000	0.00000000	-1.95738109
C	0.00000000	0.00000000	-0.60658071
C	0.00000000	0.00000000	0.60647202
C	0.00000000	0.00000000	1.95728214
C	0.00000000	0.00000000	3.17030777
C	0.00000000	0.00000000	4.52122728
C	0.00000000	0.00000000	5.73415641
C	0.00000000	0.00000000	7.08538512
C	0.00000000	0.00000000	8.29804347
C	0.00000000	0.00000000	9.65008557
C	0.00000000	0.00000000	10.86197387
C	0.00000000	0.00000000	12.21633361
C	0.00000000	0.00000000	13.42587262
C	0.00000000	0.00000000	14.78882260
C	0.00000000	0.00000000	15.99052380
H	0.00000000	0.00000000	17.05458913
H	0.00000000	0.00000000	-17.05438487

H-(C≡C)$_{14}$-H (Charge = 0, Spin multiplicity = 1)			
Atom	*x*	*y*	*z*

C	0.00000000	0.00000000	-5.80316287
C	0.00000000	0.00000000	-7.01611132
C	0.00000000	0.00000000	-8.36727621
C	0.00000000	0.00000000	-9.57997308
C	0.00000000	0.00000000	-10.93190855
C	0.00000000	0.00000000	-12.14385675
C	0.00000000	0.00000000	-13.49806874
C	0.00000000	0.00000000	-14.70764633
C	0.00000000	0.00000000	-16.07053826
C	0.00000000	0.00000000	-17.27222449
C	0.00000000	0.00000000	-4.45228360
C	0.00000000	0.00000000	-3.23924959
C	0.00000000	0.00000000	-1.88847353
C	0.00000000	0.00000000	-0.67541430
C	0.00000000	0.00000000	0.67533829
C	0.00000000	0.00000000	1.88839453
C	0.00000000	0.00000000	3.23918120
C	0.00000000	0.00000000	4.45220483
C	0.00000000	0.00000000	5.80311080
C	0.00000000	0.00000000	7.01603697
C	0.00000000	0.00000000	8.36725647
C	0.00000000	0.00000000	9.57991217
C	0.00000000	0.00000000	10.93194703
C	0.00000000	0.00000000	12.14383320
C	0.00000000	0.00000000	13.49818670
C	0.00000000	0.00000000	14.70772364
C	0.00000000	0.00000000	16.07066838
C	0.00000000	0.00000000	17.27236668
H	0.00000000	0.00000000	18.33643545

H	0.00000000	0.00000000	-18.33627463

Table S2 Calculated polarizability of the carbon chains H-(C≡C)$_n$-H (n = 3-15) in the zero-frequency limit (λ = ∞ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the ωB97XD/LPol-ds level

	Incident wavelength (λ, in nm)	$\alpha_{iso}(\lambda)$ (in 10^{-23}esu)	$\alpha_{xx}(\lambda) = \alpha_{yy}(\lambda)$ (in 10^{-23}esu)	$\alpha_{zz}(\lambda)$ (in 10^{-23}esu)
H-(C≡C)$_3$-H	∞	1.242	0.616	2.494
	1907	1.250	0.618	2.514
	1460	1.255	0.619	2.527
	1340	1.257	0.619	2.534
	1064	1.266	0.621	2.557
H-(C≡C)$_4$-H	∞	1.903	0.779	4.153
	1907	1.919	0.780	4.195
	1460	1.930	0.782	4.225
	1340	1.935	0.782	4.239
	1064	1.953	0.784	4.292
H-(C≡C)$_5$-H	∞	2.697	0.941	6.208
	1907	2.724	0.943	6.286
	1460	2.744	0.944	6.343
	1340	2.753	0.945	6.369
	1064	2.788	0.947	6.468
H-(C≡C)$_6$-H	∞	3.609	1.103	8.620
	1907	3.653	1.105	8.750
	1460	3.686	1.107	8.844
	1340	3.701	1.108	8.887
	1064	3.758	1.110	9.052
H-(C≡C)$_7$-H	∞	4.624	1.265	11.340
	1907	4.691	1.268	11.540
	1460	4.740	1.269	11.680
	1340	4.763	1.270	11.750
	1064	4.849	1.273	12.000
H-(C≡C)$_8$-H	∞	5.728	1.427	14.330
	1907	5.823	1.430	14.610

	1460	5.892	1.432	14.810
	1340	5.924	1.433	14.910
	1064	6.047	1.436	15.270
	∞	6.900	1.589	17.520
	1907	7.027	1.592	17.900
H-(C≡C)$_9$-H	1460	7.120	1.594	18.170
	1340	7.164	1.595	18.300
	1064	7.330	1.599	18.790
	∞	8.143	1.751	20.930
	1907	8.307	1.754	21.410
H-(C≡C)$_{10}$-H	1460	8.428	1.757	21.770
	1340	8.484	1.758	21.940
	1064	8.701	1.762	22.580
	∞	9.429	1.913	24.460
	1907	9.635	1.917	25.070
H-(C≡C)$_{11}$-H	1460	9.786	1.919	25.520
	1340	9.857	1.920	25.730
	1064	10.130	1.925	26.540
	∞	10.760	2.075	28.120
	1907	11.010	2.079	28.860
H-(C≡C)$_{12}$-H	1460	11.190	2.082	29.410
	1340	11.280	2.083	29.670
	1064	11.610	2.088	30.660
	∞	12.120	2.237	31.880
	1907	12.410	2.241	32.760
H-(C≡C)$_{13}$-H	1460	12.640	2.244	33.420
	1340	12.740	2.246	33.720
	1064	13.140	2.251	34.920
	∞	13.500	2.398	35.710
	1907	13.850	2.403	36.740
H-(C≡C)$_{14}$-H	1460	14.110	2.406	37.510
	1340	14.230	2.408	37.870
	1064	14.700	2.414	39.280

	∞	14.910	2.560	39.620
	1907	15.310	2.565	40.810
H-(C≡C)$_{15}$-H	1460	15.610	2.569	41.700
	1340	15.750	2.571	42.110
	1064	16.300	2.577	43.740

Table S3 Calculated second hyperpolarizability of the carbon chains H-(C≡C)$_n$-H (n = 3-15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the ωB97XD/LPol-ds level

	Incident wavelength (λ, in nm)	$\gamma_{\|\|}(\lambda)$ (in 10^{-34}esu)	$\gamma_{xxxx}(\lambda) = \gamma_{yyyy}(\lambda)$ (in 10^{-34}esu)	$\gamma_{zzzz}(\lambda)$ (in 10^{-34}esu)
H-(C≡C)$_3$-H	∞	0.154	0.045	0.473
	1907	0.164	0.047	0.508
	1460	0.172	0.048	0.534
	1340	0.176	0.049	0.547
	1064	0.191	0.052	0.598
H-(C≡C)$_4$-H	∞	0.366	0.052	1.438
	1907	0.397	0.054	1.569
	1460	0.421	0.056	1.672
	1340	0.433	0.056	1.721
	1064	0.481	0.059	1.926
H-(C≡C)$_5$-H	∞	0.822	0.058	3.625
	1907	0.909	0.060	4.028
	1460	0.979	0.062	4.353
	1340	1.013	0.063	4.512
	1064	1.157	0.066	5.183
H-(C≡C)$_6$-H	∞	1.694	0.066	7.890
	1907	1.911	0.069	8.935
	1460	2.089	0.070	9.796
	1340	2.178	0.071	10.220
	1064	2.561	0.075	12.080
H-(C≡C)$_7$-H	∞	3.182	0.070	15.250
	1907	3.660	0.072	17.590
	1460	4.063	0.074	19.560
	1340	4.266	0.075	20.560
	1064	5.167	0.079	24.980
H-(C≡C)$_8$-H	∞	5.495	0.080	26.710
	1907	6.436	0.083	31.360

	1460	7.246	0.085	35.360
	1340	7.661	0.086	37.410
	1064	9.548	0.090	46.750
$H\text{-}(C{\equiv}C)_9\text{-}H$	∞	8.798	0.087	43.140
	1907	10.480	0.090	51.470
	1460	11.950	0.093	58.790
	1340	12.720	0.094	62.590
	1064	16.290	0.098	80.320
$H\text{-}(C{\equiv}C)_{10}\text{-}H$	∞	13.260	0.094	65.350
	1907	16.030	0.098	79.160
	1460	18.520	0.100	91.520
	1340	19.830	0.101	98.030
	1064	26.070	0.106	129.100
$H\text{-}(C{\equiv}C)_{11}\text{-}H$	∞	18.930	0.106	93.590
	1907	23.210	0.110	114.900
	1460	27.110	0.112	134.300
	1340	29.190	0.114	144.700
	1064	39.320	0.119	195.200
$H\text{-}(C{\equiv}C)_{12}\text{-}H$	∞	25.830	0.109	128.000
	1907	32.070	0.113	159.200
	1460	37.860	0.115	188.000
	1340	40.970	0.117	203.500
	1064	56.450	0.122	280.700
$H\text{-}(C{\equiv}C)_{13}\text{-}H$	∞	33.960	0.116	168.600
	1907	42.650	0.120	211.900
	1460	50.810	0.123	252.600
	1340	55.240	0.124	274.800
	1064	77.720	0.129	386.900
$H\text{-}(C{\equiv}C)_{14}\text{-}H$	∞	43.290	0.123	215.100
	1907	54.910	0.127	273.100
	1460	65.970	0.130	328.300
	1340	72.030	0.132	358.600
	1064	103.300	0.137	514.800

	∞	53.670	0.129	267.000
	1907	68.700	0.133	342.000
H-(C≡C)$_{15}$-H	1460	83.170	0.136	414.200
	1340	91.180	0.138	454.200
	1064	133.200	0.144	663.800

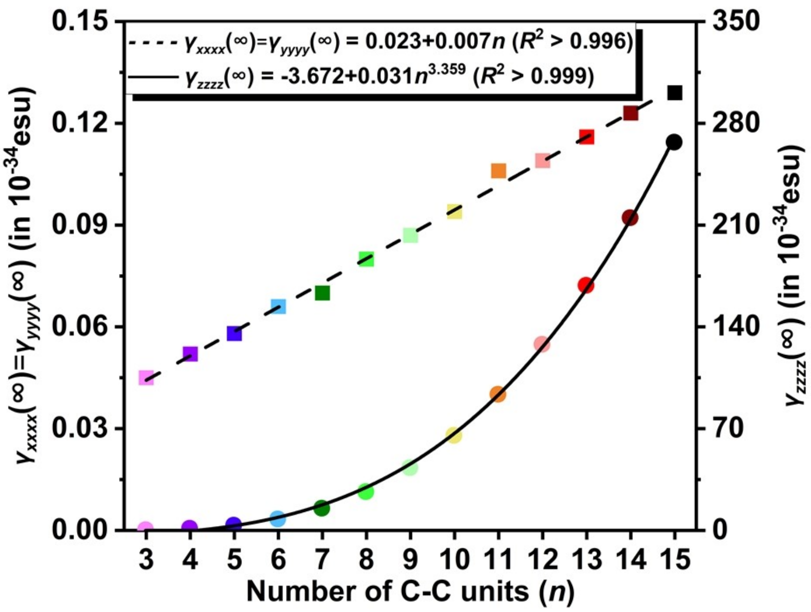

Fig. S1 Static second hyperpolarizability tensors as a function of the number of C-C units (n) in the carbon chains at the ωB97XD/LPol-ds level.

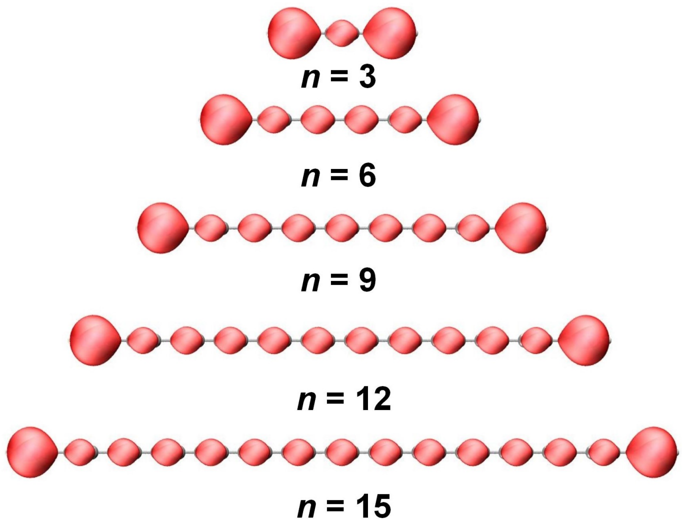

Fig. S2 Hyperpolarizability density $-x^{(3)}_{xxx}(\vec{\ })$ isosurfaces (isovalue = 10 au) of the carbon chains H-(C≡C)$_n$-H (n = 3, 6, 9, 12, and 15) at the ωB97XD/LPol-ds level. Red and blue regions denote positive and negative spatial contributions, respectively.

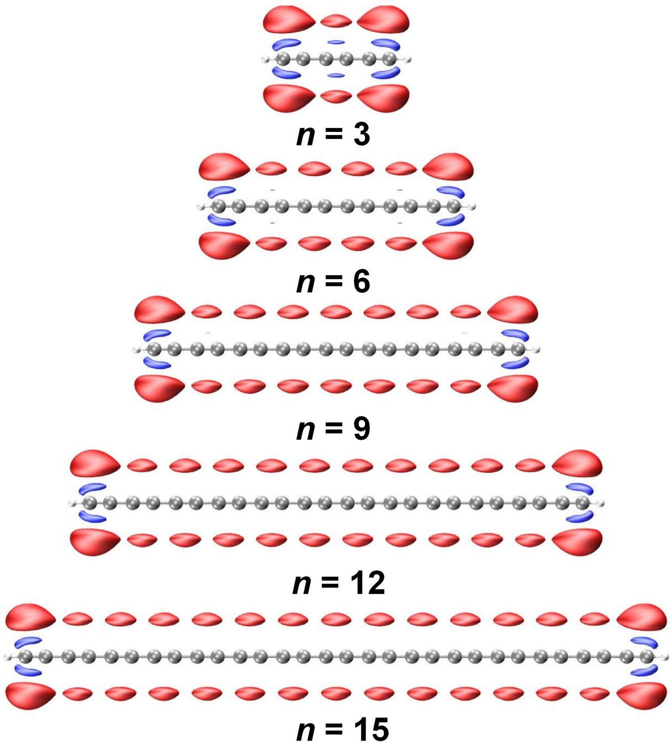

Fig. S3 Hyperpolarizability density $-y^{(3)}_{yyy}(\vec{\ })$ isosurfaces (isovalue = 10 au) of the carbon chains H-(C≡C)$_n$-H (*n* = 3, 6, 9, 12, and 15) at the ωB97XD/LPol-ds level. Red and blue regions denote positive and negative spatial contributions, respectively.

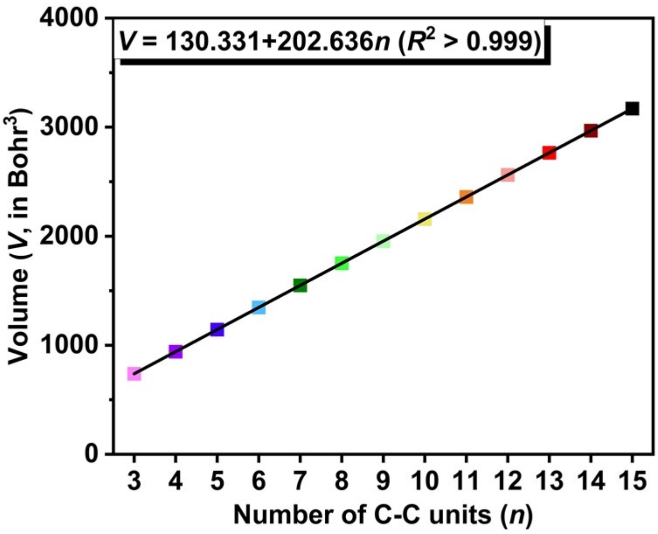

Fig. S4 Volumes of the carbon chains H-(C≡C)$_n$-H (n = 3-15) as a function of the number of C-C units in the chains.

Table S4 Total molecular polarizability [$\alpha_{iso}(\lambda)$, $\lambda = \infty$, 1907, 1460, 1340, and 1064 nm] and total second hyperpolarizability [$\gamma_{||}(\lambda)$, $\lambda = \infty$, 1907, 1460, 1340, and 1064 nm] versus the number of carbon atoms (n_C) in carbon chains H-(C≡C)$_n$-H (n = 3-15) at the ωB97XD/LPol-ds level

	Incident wavelength (λ, in nm)	$\alpha_{iso}(\lambda)/n_C$ (in 10^{-23}esu)	$\gamma_{||}(\lambda)/n_C$ (in 10^{-34}esu)
H-(C≡C)$_3$-H	∞	0.207	0.026
	1907	0.208	0.027
	1460	0.209	0.029
	1340	0.210	0.029
	1064	0.211	0.032
H-(C≡C)$_4$-H	∞	0.238	0.046
	1907	0.240	0.050
	1460	0.241	0.053
	1340	0.242	0.054
	1064	0.244	0.060
H-(C≡C)$_5$-H	∞	0.270	0.082
	1907	0.272	0.091
	1460	0.274	0.098
	1340	0.275	0.101
	1064	0.279	0.116
H-(C≡C)$_6$-H	∞	0.301	0.141
	1907	0.304	0.159
	1460	0.307	0.174
	1340	0.308	0.182
	1064	0.313	0.213
H-(C≡C)$_7$-H	∞	0.330	0.227
	1907	0.335	0.261
	1460	0.339	0.290
	1340	0.340	0.305
	1064	0.346	0.369
H-(C≡C)$_8$-H	∞	0.358	0.343

	1907	0.364	0.402
	1460	0.368	0.453
	1340	0.370	0.479
	1064	0.378	0.597
H-$(C\equiv C)_9$-H	∞	0.383	0.489
	1907	0.390	0.582
	1460	0.396	0.664
	1340	0.398	0.707
	1064	0.407	0.905
H-$(C\equiv C)_{10}$-H	∞	0.407	0.663
	1907	0.415	0.802
	1460	0.421	0.926
	1340	0.424	0.992
	1064	0.435	1.303
H-$(C\equiv C)_{11}$-H	∞	0.429	0.860
	1907	0.438	1.055
	1460	0.445	1.232
	1340	0.448	1.327
	1064	0.460	1.787
H-$(C\equiv C)_{12}$-H	∞	0.448	1.076
	1907	0.459	1.336
	1460	0.466	1.577
	1340	0.470	1.707
	1064	0.484	2.352
H-$(C\equiv C)_{13}$-H	∞	0.466	1.306
	1907	0.478	1.640
	1460	0.486	1.954
	1340	0.490	2.125
	1064	0.505	2.989
H-$(C\equiv C)_{14}$-H	∞	0.482	1.546
	1907	0.495	1.961
	1460	0.504	2.356
	1340	0.508	2.573

	1064	0.525	3.690
	∞	0.497	1.789
	1907	0.510	2.290
H-(C≡C)$_{15}$-H	1460	0.520	2.772
	1340	0.525	3.039
	1064	0.543	4.439

Table S5 Vertical ionization energy (*VIE*) and vertical electron affinity (*VEA*) of the carbon chains H-(C≡C)$_n$-H (n = 3-15) at the ωB97XD/def2-TZVP level

	$E_{[\text{H-(C≡C)}_n\text{-H}]^+}$ (in eV)	$E_{[\text{H-(C≡C)}_n\text{-H}]}$ (in eV)	$E_{[\text{H-(C≡C)}_n\text{-H}]^-}$ (in eV)	*VIE* (in eV)	*VEA* (in eV)
H-(C≡C)$_3$-H	-6239.079	-6248.404	-6248.195	9.325	-0.209
H-(C≡C)$_4$-H	-8311.640	-8320.542	-8321.033	8.902	0.490
H-(C≡C)$_5$-H	-10384.068	-10392.685	-10393.658	8.617	0.973
H-(C≡C)$_6$-H	-12456.414	-12464.829	-12466.150	8.415	1.321
H-(C≡C)$_7$-H	-14528.705	-14536.973	-14538.554	8.268	1.581
H-(C≡C)$_8$-H	-16600.958	-16609.117	-16610.898	8.159	1.781
H-(C≡C)$_9$-H	-18673.182	-18681.261	-18683.196	8.079	1.935
H-(C≡C)$_{10}$-H	-20745.389	-20753.405	-20755.466	8.016	2.061
H-(C≡C)$_{11}$-H	-22817.579	-22825.548	-22827.709	7.969	2.161
H-(C≡C)$_{12}$-H	-24889.758	-24897.692	-24899.933	7.934	2.241
H-(C≡C)$_{13}$-H	-26961.928	-26969.835	-26972.142	7.907	2.307
H-(C≡C)$_{14}$-H	-29034.092	-29041.978	-29044.340	7.887	2.361
H-(C≡C)$_{15}$-H	-31106.250	-31114.122	-31116.528	7.872	2.406

Table S6 Calculated polarizability of the carbon chains H-(C≡C)$_n$-H (n = 3-15) in the zero-frequency limit (λ = ∞ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the ωB97XD/aug-cc-pVTZ level

	Incident wavelength (λ, in nm)	$\alpha_{iso}(\lambda)$ (in 10^{-23}esu)	$\alpha_{xx}(\lambda) = \alpha_{yy}(\lambda)$ (in 10^{-23}esu)	$\alpha_{zz}(\lambda)$ (in 10^{-23}esu)
H-(C≡C)$_3$-H	∞	1.243	0.616	2.496
	1907	1.250	0.617	2.516
	1460	1.255	0.618	2.529
	1340	1.258	0.619	2.536
	1064	1.267	0.620	2.559
H-(C≡C)$_4$-H	∞	1.904	0.778	4.156
	1907	1.919	0.780	4.198
	1460	1.930	0.781	4.228
	1340	1.935	0.781	4.242
	1064	1.954	0.784	4.295
H-(C≡C)$_5$-H	∞	2.698	0.940	6.212
	1907	2.725	0.942	6.291
	1460	2.745	0.944	6.347
	1340	2.754	0.944	6.373
	1064	2.788	0.947	6.472
H-(C≡C)$_6$-H	∞	3.610	1.102	8.626
	1907	3.655	1.104	8.755
	1460	3.687	1.106	8.849
	1340	3.702	1.107	8.893
	1064	3.759	1.109	9.057
H-(C≡C)$_7$-H	∞	4.626	1.264	11.350
	1907	4.693	1.267	11.550
	1460	4.742	1.268	11.690
	1340	4.765	1.269	11.750
	1064	4.851	1.272	12.010
H-(C≡C)$_8$-H	∞	5.731	1.426	14.340
	1907	5.825	1.429	14.620

	1460	5.894	1.431	14.820
	1340	5.926	1.432	14.920
	1064	6.049	1.435	15.280
	∞	6.904	1.588	17.540
	1907	7.030	1.591	17.910
H-(C≡C)$_9$-H	1460	7.124	1.593	18.180
	1340	7.167	1.594	18.310
	1064	7.333	1.598	18.800
	∞	8.147	1.750	20.940
	1907	8.311	1.753	21.430
H-(C≡C)$_{10}$-H	1460	8.432	1.755	21.780
	1340	8.488	1.757	21.950
	1064	8.704	1.761	22.590
	∞	9.435	1.911	24.480
	1907	9.640	1.915	25.090
H-(C≡C)$_{11}$-H	1460	9.791	1.918	25.540
	1340	9.862	1.919	25.750
	1064	10.130	1.924	26.560
	∞	10.760	2.073	28.140
	1907	11.010	2.077	28.880
H-(C≡C)$_{12}$-H	1460	11.200	2.080	29.430
	1340	11.280	2.081	29.690
	1064	11.620	2.086	30.680
	∞	12.120	2.235	31.900
	1907	12.420	2.239	32.790
H-(C≡C)$_{13}$-H	1460	12.640	2.242	33.440
	1340	12.750	2.244	33.750
	1064	13.150	2.249	34.940
	∞	13.510	2.397	35.740
	1907	13.860	2.401	36.780
H-(C≡C)$_{14}$-H	1460	14.120	2.405	37.550
	1340	14.240	2.406	37.910
	1064	14.710	2.412	39.310

	∞	14.920	2.559	39.650
	1907	15.320	2.563	40.840
H-(C≡C)$_{15}$-H	1460	15.620	2.567	41.730
	1340	15.760	2.569	42.150
	1064	16.310	2.575	43.770

Table S7 Calculated second hyperpolarizability of the carbon chains H-(C≡C)$_n$-H (n = 3-15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the ωB97XD/aug-cc-pVTZ level

	Incident wavelength (λ, in nm)	$\gamma_{\|\|}(\lambda)$ (in 10^{-34}esu)	$\gamma_{xxxx}(\lambda) = \gamma_{yyyy}(\lambda)$ (in 10^{-34}esu)	$\gamma_{zzzz}(\lambda)$ (in 10^{-34}esu)
	∞	0.150	0.041	0.476
	1907	0.160	0.043	0.511
H-(C≡C)$_3$-H	1460	0.168	0.044	0.538
	1340	0.172	0.045	0.551
	1064	0.186	0.047	0.602
	∞	0.362	0.048	1.447
	1907	0.393	0.050	1.579
H-(C≡C)$_4$-H	1460	0.417	0.051	1.682
	1340	0.429	0.052	1.732
	1064	0.476	0.055	1.937
	∞	0.819	0.055	3.639
	1907	0.905	0.057	4.043
H-(C≡C)$_5$-H	1460	0.975	0.059	4.368
	1340	1.009	0.059	4.527
	1064	1.152	0.062	5.199
	∞	1.691	0.062	7.911
	1907	1.908	0.064	8.956
H-(C≡C)$_6$-H	1460	2.086	0.066	9.817
	1340	2.174	0.067	10.250
	1064	2.557	0.070	12.100
	∞	3.181	0.069	15.270
	1907	3.658	0.071	17.610
H-(C≡C)$_7$-H	1460	4.060	0.073	19.580
	1340	4.263	0.074	20.580
	1064	5.161	0.077	24.990
	∞	5.494	0.076	26.750
H-(C≡C)$_8$-H	1907	6.434	0.079	31.390
	1460	7.242	0.080	35.390

	1340	7.656	0.081	37.430
	1064	9.538	0.085	46.750
	∞	8.798	0.083	43.180
	1907	10.480	0.086	51.500
H-(C≡C)$_9$-H	1460	11.950	0.088	58.810
	1340	12.710	0.089	62.610
	1064	16.280	0.093	80.300
	∞	13.260	0.090	65.420
	1907	16.040	0.093	79.210
H-(C≡C)$_{10}$-H	1460	18.520	0.095	91.560
	1340	19.820	0.096	98.050
	1064	26.050	0.100	129.000
	∞	18.930	0.097	93.680
	1907	23.210	0.100	115.000
H-(C≡C)$_{11}$-H	1460	27.110	0.102	134.400
	1340	29.180	0.104	144.700
	1064	39.280	0.108	195.100
	∞	25.850	0.104	128.200
	1907	32.090	0.107	159.300
H-(C≡C)$_{12}$-H	1460	37.860	0.110	188.100
	1340	40.970	0.111	203.600
	1064	56.410	0.116	280.600
	∞	33.990	0.111	168.800
	1907	42.670	0.115	212.100
H-(C≡C)$_{13}$-H	1460	50.820	0.117	252.800
	1340	55.250	0.118	274.900
	1064	77.690	0.123	386.800
	∞	43.310	0.118	215.300
	1907	54.920	0.122	273.200
H-(C≡C)$_{14}$-H	1460	65.960	0.125	328.400
	1340	72.020	0.126	358.600
	1064	103.200	0.131	514.400
H-(C≡C)$_{15}$-H	∞	53.730	0.125	267.300

	1907	68.760	0.129	342.300
	1460	83.220	0.132	414.500
	1340	91.220	0.133	454.500
	1064	133.100	0.139	663.700

Table S8 Calculated polarizability of the carbon chains H-(C≡C)$_n$-H (n = 3- 15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the CAM-B3LYP/LPol-ds level

	Incident wavelength (λ, in nm)	$\alpha_{iso}(\lambda)$ (in 10^{-23}esu)	$\alpha_{xx}(\lambda) = \alpha_{yy}(\lambda)$ (in 10^{-23}esu)	$\alpha_{zz}(\lambda)$ (in 10^{-23}esu)
H-(C≡C)$_3$-H	∞	1.243	0.615	2.498
	1907	1.250	0.616	2.517
	1460	1.255	0.617	2.531
	1340	1.258	0.618	2.538
	1064	1.267	0.620	2.562
H-(C≡C)$_4$-H	∞	1.906	0.777	4.163
	1907	1.921	0.779	4.206
	1460	1.932	0.780	4.236
	1340	1.937	0.780	4.251
	1064	1.956	0.782	4.304
H-(C≡C)$_5$-H	∞	2.704	0.939	6.235
	1907	2.732	0.941	6.314
	1460	2.752	0.942	6.372
	1340	2.761	0.943	6.399
	1064	2.796	0.945	6.499
H-(C≡C)$_6$-H	∞	3.626	1.100	8.676
	1907	3.671	1.102	8.809
	1460	3.704	1.104	8.905
	1340	3.720	1.105	8.949
	1064	3.778	1.108	9.118
H-(C≡C)$_7$-H	∞	4.656	1.262	11.440
	1907	4.725	1.264	11.650
	1460	4.775	1.266	11.790
	1340	4.798	1.267	11.860
	1064	4.887	1.270	12.120
H-(C≡C)$_8$-H	∞	5.780	1.423	14.490
	1907	5.878	1.426	14.780

	1460	5.949	1.428	14.990
	1340	5.983	1.429	15.090
	1064	6.110	1.432	15.460
H-(C≡C)$_9$-H	∞	6.978	1.584	17.760
	1907	7.110	1.587	18.150
	1460	7.207	1.590	18.440
	1340	7.252	1.591	18.570
	1064	7.425	1.595	19.090
H-(C≡C)$_{10}$-H	∞	8.251	1.746	21.260
	1907	8.423	1.749	21.770
	1460	8.550	1.752	22.150
	1340	8.608	1.753	22.320
	1064	8.836	1.757	22.990
H-(C≡C)$_{11}$-H	∞	9.573	1.907	24.910
	1907	9.789	1.911	25.550
	1460	9.949	1.914	26.020
	1340	10.020	1.915	26.240
	1064	10.310	1.919	27.100
H-(C≡C)$_{12}$-H	∞	10.940	2.068	28.680
	1907	11.200	2.073	29.470
	1460	11.400	2.076	30.050
	1340	11.490	2.077	30.320
	1064	11.850	2.082	31.380
H-(C≡C)$_{13}$-H	∞	12.340	2.230	32.570
	1907	12.660	2.234	33.510
	1460	12.890	2.237	34.210
	1340	13.000	2.239	34.530
	1064	13.430	2.244	35.810
H-(C≡C)$_{14}$-H	∞	13.780	2.391	36.550
	1907	14.150	2.396	37.650
	1460	14.420	2.399	38.470
	1340	14.550	2.401	38.860
	1064	15.060	2.406	40.360

	∞	15.240	2.553	40.600
	1907	15.660	2.558	41.880
H-(C≡C)$_{15}$-H	1460	15.980	2.561	42.830
	1340	16.130	2.563	43.270
	1064	16.720	2.569	45.020

Table S9 Calculated second hyperpolarizability of the carbon chains H-(C≡C)$_n$-H (n = 3-15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the CAM-B3LYP/LPol-ds level

	Incident wavelength (λ, in nm)	$\gamma_{\|\|}(\lambda)$ (in 10^{-34}esu)	$\gamma_{xxxx}(\lambda) = \gamma_{yyyy}(\lambda)$ (in 10^{-34}esu)	$\gamma_{zzzz}(\lambda)$ (in 10^{-34}esu)
H-(C≡C)$_3$-H	∞	0.159	0.043	0.499
	1907	0.170	0.045	0.537
	1460	0.179	0.047	0.566
	1340	0.183	0.047	0.580
	1064	0.199	0.050	0.636
H-(C≡C)$_4$-H	∞	0.381	0.049	1.523
	1907	0.415	0.051	1.666
	1460	0.441	0.052	1.779
	1340	0.454	0.053	1.834
	1064	0.506	0.056	2.060
H-(C≡C)$_5$-H	∞	0.872	0.056	3.876
	1907	0.969	0.058	4.323
	1460	1.046	0.060	4.685
	1340	1.084	0.061	4.863
	1064	1.245	0.064	5.616
H-(C≡C)$_6$-H	∞	1.824	0.063	8.540
	1907	2.069	0.065	9.722
	1460	2.271	0.067	10.700
	1340	2.373	0.068	11.190
	1064	2.813	0.071	13.320
H-(C≡C)$_7$-H	∞	3.484	0.069	16.740
	1907	4.036	0.072	19.450
	1460	4.505	0.074	21.760
	1340	4.744	0.075	22.930
	1064	5.811	0.079	28.170
H-(C≡C)$_8$-H	∞	6.105	0.066	29.790
	1907	7.221	0.068	35.300
	1460	8.191	0.070	40.100

	1340	8.693	0.071	42.580
	1064	11.000	0.074	54.020
	∞	9.952	0.082	48.900
	1907	11.990	0.086	59.030
H-(C≡C)$_9$-H	1460	13.810	0.088	68.070
	1340	14.770	0.089	72.810
	1064	19.290	0.093	95.300
	∞	15.240	0.089	75.250
	1907	18.700	0.092	92.440
H-(C≡C)$_{10}$-H	1460	21.850	0.095	108.100
	1340	23.530	0.096	116.500
	1064	31.700	0.100	157.200
	∞	22.080	0.096	109.400
	1907	27.540	0.099	136.500
H-(C≡C)$_{11}$-H	1460	32.610	0.102	161.800
	1340	35.350	0.103	175.500
	1064	49.060	0.107	243.800
	∞	30.570	0.103	151.700
	1907	38.690	0.106	192.200
H-(C≡C)$_{12}$-H	1460	46.390	0.109	230.600
	1340	50.600	0.110	251.600
	1064	72.190	0.115	359.300
	∞	40.720	0.109	202.300
	1907	52.230	0.113	259.800
H-(C≡C)$_{13}$-H	1460	63.340	0.116	315.200
	1340	69.490	0.118	345.900
	1064	101.800	0.123	507.000
	∞	52.450	0.116	260.900
	1907	68.110	0.120	339.100
H-(C≡C)$_{14}$-H	1460	83.460	0.123	415.700
	1340	92.040	0.125	458.600
	1064	138.100	0.130	688.600
H-(C≡C)$_{15}$-H	∞	65.770	0.169	327.300

	1907	86.350	0.176	430.000
	1460	106.800	0.181	532.100
	1340	118.400	0.183	589.900
	1064	181.600	0.192	906.000

Table S10 Calculated polarizability of the carbon chains H-(C≡C)$_n$-H (n = 3- 15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the CAM-B3LYP/aug-cc-pVTZ level

	Incident wavelength (λ, in nm)	$\alpha_{iso}(\lambda)$ (in 10^{-23}esu)	$\alpha_{xx}(\lambda) = \alpha_{yy}(\lambda)$ (in 10^{-23}esu)	$\alpha_{zz}(\lambda)$ (in 10^{-23}esu)
	∞	1.242	0.612	2.501
	1907	1.249	0.613	2.520
H-(C≡C)$_3$-H	1460	1.254	0.614	2.534
	1340	1.257	0.615	2.541
	1064	1.266	0.616	2.565
	∞	1.905	0.773	4.168
	1907	1.920	0.775	4.211
H-(C≡C)$_4$-H	1460	1.931	0.776	4.242
	1340	1.936	0.777	4.256
	1064	1.956	0.779	4.309
	∞	2.704	0.935	6.243
	1907	2.732	0.937	6.322
H-(C≡C)$_5$-H	1460	2.752	0.938	6.380
	1340	2.761	0.939	6.407
	1064	2.796	0.941	6.507
	∞	3.627	1.096	8.688
	1907	3.672	1.098	8.820
H-(C≡C)$_6$-H	1460	3.705	1.100	8.916
	1340	3.720	1.100	8.961
	1064	3.779	1.103	9.129
	∞	4.658	1.257	11.460
	1907	4.727	1.259	11.662
H-(C≡C)$_7$-H	1460	4.777	1.261	11.809
	1340	4.800	1.262	11.877
	1064	4.889	1.265	12.137
	∞	5.783	1.418	14.514
H-(C≡C)$_8$-H	1907	5.881	1.421	14.802
	1460	5.953	1.423	15.013

	1340	5.986	1.424	15.111
	1064	6.113	1.427	15.486
	∞	6.983	1.579	17.791
	1907	7.115	1.582	18.181
H-(C≡C)$_9$-H	1460	7.212	1.584	18.468
	1340	7.257	1.585	18.601
	1064	7.430	1.589	19.113
	∞	8.258	1.740	21.296
	1907	8.430	1.743	21.804
H-(C≡C)$_{10}$-H	1460	8.557	1.746	22.179
	1340	8.616	1.747	22.354
	1064	8.843	1.751	23.028
	∞	9.583	1.901	24.948
	1907	9.799	1.905	25.588
H-(C≡C)$_{11}$-H	1460	9.959	1.907	26.062
	1340	10.033	1.908	26.282
	1064	10.321	1.913	27.138
	∞	10.953	2.062	28.735
	1907	11.217	2.066	29.519
H-(C≡C)$_{12}$-H	1460	11.413	2.069	30.101
	1340	11.504	2.070	30.372
	1064	11.859	2.075	31.429
	∞	12.359	2.223	32.632
	1907	12.675	2.227	33.571
H-(C≡C)$_{13}$-H	1460	12.910	2.230	34.269
	1340	13.020	2.231	34.596
	1064	13.448	2.237	35.869
	∞	13.795	2.384	36.617
	1907	14.166	2.388	37.720
H-(C≡C)$_{14}$-H	1460	14.442	2.392	38.543
	1340	14.571	2.393	38.928
	1064	15.077	2.399	40.434
H-(C≡C)$_{15}$-H	∞	15.256	2.545	40.680

	1907	15.685	2.550	41.955
	1460	16.005	2.553	42.909
	1340	16.155	2.555	43.355
	1064	16.742	2.560	45.106

Table S11 Calculated second hyperpolarizability of the carbon chains H-(C≡C)$_n$-H (n = 3-15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the CAM-B3LYP/aug-cc-pVTZ level

	Incident wavelength (λ, in nm)	$\gamma_{\|\|}(\lambda)$ (in 10^{-34}esu)	$\gamma_{xxxx}(\lambda) = \gamma_{yyyy}(\lambda)$ (in 10^{-34}esu)	$\gamma_{zzzz}(\lambda)$ (in 10^{-34}esu)
	∞	0.152	0.038	0.498
	1907	0.163	0.040	0.535
H-(C≡C)$_3$-H	1460	0.171	0.041	0.564
	1340	0.175	0.041	0.578
	1064	0.190	0.044	0.634
	∞	0.375	0.044	1.525
	1907	0.408	0.046	1.668
H-(C≡C)$_4$-H	1460	0.434	0.048	1.781
	1340	0.447	0.048	1.836
	1064	0.499	0.051	2.062
	∞	0.863	0.051	3.874
	1907	0.958	0.053	4.321
H-(C≡C)$_5$-H	1460	1.035	0.054	4.682
	1340	1.073	0.055	4.860
	1064	1.232	0.057	5.611
	∞	1.813	0.057	8.537
	1907	2.056	0.059	9.716
H-(C≡C)$_6$-H	1460	2.258	0.061	10.694
	1340	2.359	0.061	11.183
	1064	2.796	0.064	13.309
	∞	3.470	0.063	16.733
	1907	4.020	0.065	19.436
H-(C≡C)$_7$-H	1460	4.487	0.067	21.735
	1340	4.724	0.068	22.902
	1064	5.786	0.071	28.133
	∞	6.098	0.069	29.790
H-(C≡C)$_8$-H	1907	7.210	0.072	35.291
	1460	8.178	0.074	40.084

	1340	8.678	0.075	42.560
	1064	10.980	0.078	53.971
H-(C≡C)$_9$-H	∞	9.938	0.076	48.900
	1907	11.974	0.078	59.013
	1460	13.789	0.080	68.033
	1340	14.741	0.081	72.765
	1064	19.251	0.085	95.192
H-(C≡C)$_{10}$-H	∞	15.229	0.082	75.267
	1907	18.679	0.085	92.440
	1460	21.824	0.087	108.100
	1340	23.498	0.088	116.439
	1064	31.652	0.092	157.060
H-(C≡C)$_{11}$-H	∞	22.079	0.088	109.432
	1907	27.525	0.091	136.575
	1460	32.592	0.093	161.834
	1340	35.328	0.095	175.473
	1064	49.000	0.099	243.662
H-(C≡C)$_{12}$-H	∞	30.579	0.095	151.849
	1907	38.692	0.098	192.314
	1460	46.384	0.100	230.684
	1340	50.590	0.101	251.669
	1064	72.139	0.105	359.215
H-(C≡C)$_{13}$-H	∞	40.740	0.101	202.569
	1907	52.249	0.104	259.999
	1460	63.347	0.107	315.394
	1340	69.489	0.108	346.052
	1064	101.701	0.112	506.878
H-(C≡C)$_{14}$-H	∞	52.520	0.107	261.387
	1907	68.183	0.111	339.573
	1460	83.525	0.113	416.174
	1340	92.108	0.115	459.032
	1064	138.121	0.119	688.829
H-(C≡C)$_{15}$-H	∞	65.853	0.114	327.967

- - - - - - - - - -	1907	86.439	0.117	430.756
	1460	106.893	0.120	532.903
	1340	118.451	0.121	590.629
	1064	181.693	0.126	906.541

Table S12 Calculated polarizability of the carbon chains H-(C≡C)$_n$-H (n = 3- 15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the PBE38/LPol-ds level

	Incident wavelength (λ, in nm)	$\alpha_{iso}(\lambda)$ (in 10^{-23}esu)	$\alpha_{xx}(\lambda) = \alpha_{yy}(\lambda)$ (in 10^{-23}esu)	$\alpha_{zz}(\lambda)$ (in 10^{-23}esu)
	∞	1.246	0.606	2.524
	1907	1.253	0.607	2.544
H-(C≡C)$_3$-H	1460	1.258	0.608	2.558
	1340	1.261	0.609	2.565
	1064	1.270	0.610	2.589
	∞	1.928	0.766	4.252
	1907	1.944	0.768	4.297
H-(C≡C)$_4$-H	1460	1.956	0.769	4.329
	1340	1.961	0.769	4.343
	1064	1.981	0.771	4.399
	∞	2.762	0.926	6.435
	1907	2.792	0.928	6.520
H-(C≡C)$_5$-H	1460	2.813	0.929	6.582
	1340	2.823	0.930	6.610
	1064	2.860	0.932	6.717
	∞	3.739	1.085	9.046
	1907	3.789	1.088	9.191
H-(C≡C)$_6$-H	1460	3.825	1.089	9.296
	1340	3.841	1.090	9.345
	1064	3.905	1.093	9.530
	∞	4.847	1.245	12.049
	1907	4.923	1.248	12.275
H-(C≡C)$_7$-H	1460	4.979	1.249	12.440
	1340	5.005	1.250	12.516
	1064	5.105	1.253	12.808
	∞	6.070	1.405	15.402
H-(C≡C)$_8$-H	1907	6.182	1.407	15.731
	1460	6.264	1.409	15.973

	1340	6.302	1.410	16.085
	1064	6.448	1.413	16.516
	∞	7.391	1.564	19.044
	1907	7.544	1.567	19.499
H-(C≡C)$_9$-H	1460	7.657	1.569	19.834
	1340	7.710	1.570	19.990
	1064	7.914	1.574	20.593
	∞	8.809	1.724	22.981
	1907	9.013	1.727	23.585
H-(C≡C)$_{10}$-H	1460	9.164	1.729	24.033
	1340	9.234	1.730	24.241
	1064	9.507	1.734	25.052
	∞	10.297	1.883	27.124
	1907	10.557	1.887	27.898
H-(C≡C)$_{11}$-H	1460	10.751	1.889	28.474
	1340	10.841	1.890	28.743
	1064	11.194	1.895	29.793
	∞	11.847	2.043	31.457
	1907	12.171	2.046	32.421
H-(C≡C)$_{12}$-H	1460	12.413	2.049	33.141
	1340	12.526	2.050	33.478
	1064	12.969	2.055	34.798
	∞	13.451	2.202	35.948
	1907	13.844	2.206	37.119
H-(C≡C)$_{13}$-H	1460	14.138	2.209	37.997
	1340	14.277	2.210	38.409
	1064	14.820	2.215	40.028
	∞	15.099	2.362	40.573
	1907	15.566	2.366	41.967
H-(C≡C)$_{14}$-H	1460	15.918	2.369	43.015
	1340	16.083	2.370	43.508
	1064	16.735	2.376	45.453
H-(C≡C)$_{15}$-H	∞	16.784	2.521	45.310

	1907	17.331	2.526	46.942
	1460	17.744	2.529	48.173
	1340	17.938	2.531	48.753
	1064	18.707	2.536	51.048

Table S13 Calculated second hyperpolarizability of the carbon chains H-(C≡C)$_n$-H (n = 3-15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the PBE38/LPol-ds level

	Incident wavelength (λ, in nm)	$\gamma_{\|\|}(\lambda)$ (in 10^{-34}esu)	$\gamma_{xxxx}(\lambda) = \gamma_{yyyy}(\lambda)$ (in 10^{-34}esu)	$\gamma_{zzzz}(\lambda)$ (in 10^{-34}esu)
H-(C≡C)$_3$-H	∞	0.154	0.040	0.490
	1907	0.165	0.042	0.528
	1460	0.173	0.043	0.557
	1340	0.177	0.043	0.570
	1064	0.193	0.046	0.626
H-(C≡C)$_4$-H	∞	0.383	0.046	1.540
	1907	0.418	0.048	1.690
	1460	0.446	0.049	1.809
	1340	0.459	0.050	1.867
	1064	0.514	0.052	2.108
H-(C≡C)$_5$-H	∞	0.904	0.052	4.045
	1907	1.010	0.054	4.543
	1460	1.097	0.056	4.949
	1340	1.139	0.056	5.150
	1064	1.321	0.059	6.009
H-(C≡C)$_6$-H	∞	1.968	0.058	9.264
	1907	2.257	0.060	10.665
	1460	2.499	0.062	11.844
	1340	2.622	0.063	12.439
	1064	3.162	0.066	15.074
H-(C≡C)$_7$-H	∞	3.923	0.064	18.942
	1907	4.621	0.067	22.375
	1460	5.226	0.068	25.358
	1340	5.538	0.069	26.897
	1064	6.971	0.072	33.968
H-(C≡C)$_8$-H	∞	7.196	0.071	35.204
	1907	8.702	0.073	42.670
	1460	10.055	0.075	49.376

	1340	10.767	0.076	52.910
	1064	14.169	0.079	69.804
$H-(C\equiv C)_9-H$	∞	12.250	0.077	60.378
	1907	15.201	0.079	75.049
	1460	17.935	0.081	88.654
	1340	19.407	0.082	95.982
	1064	26.730	0.086	132.444
$H-(C\equiv C)_{10}-H$	∞	19.578	0.083	96.917
	1907	24.905	0.086	123.457
	1460	30.001	0.088	148.854
	1340	32.804	0.089	162.832
	1064	47.337	0.092	235.308
$H-(C\equiv C)_{11}-H$	∞	29.567	0.089	146.765
	1907	38.506	0.092	191.347
	1460	47.316	0.094	235.302
	1340	52.265	0.095	260.000
	1064	78.999	0.099	393.437
$H-(C\equiv C)_{12}-H$	∞	42.557	0.093	211.625
	1907	56.659	0.096	282.012
	1460	70.960	0.099	353.406
	1340	79.157	0.100	394.330
	1064	125.279	0.104	624.667
$H-(C\equiv C)_{13}-H$	∞	58.776	0.101	292.615
	1907	79.880	0.104	397.987
	1460	101.860	0.107	507.760
	1340	114.701	0.108	571.897
	1064	189.931	0.112	947.721
$H-(C\equiv C)_{14}-H$	∞	78.360	0.108	390.424
	1907	108.543	0.112	541.183
	1460	140.779	0.114	702.217
	1340	159.954	0.115	798.015
	1064	276.838	0.120	1382.050
$H-(C\equiv C)_{15}-H$	∞	101.275	0.114	504.914

– – – – – – – – – –	1907	142.786	0.117	712.290
	1460	188.169	0.120	939.037
	1340	215.631	0.121	1076.262
	1064	389.643	0.126	1945.869

Table S14 Calculated polarizability of the carbon chains H-(C≡C)$_n$-H (n = 3-15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the PBE38/aug-cc-pVTZ level

	Incident wavelength (λ, in nm)	$\alpha_{iso}(\lambda)$ (in 10^{-23}esu)	$\alpha_{xx}(\lambda) = \alpha_{yy}(\lambda)$ (in 10^{-23}esu)	$\alpha_{zz}(\lambda)$ (in 10^{-23}esu)
H-(C≡C)$_3$-H	∞	1.245	0.603	2.528
	1907	1.252	0.605	2.547
	1460	1.258	0.606	2.561
	1340	1.260	0.606	2.568
	1064	1.269	0.608	2.592
H-(C≡C)$_4$-H	∞	1.928	0.763	4.258
	1907	1.944	0.765	4.302
	1460	1.955	0.766	4.334
	1340	1.961	0.766	4.349
	1064	1.980	0.768	4.404
H-(C≡C)$_5$-H	∞	2.763	0.923	6.443
	1907	2.792	0.924	6.529
	1460	2.814	0.926	6.590
	1340	2.824	0.926	6.618
	1064	2.861	0.928	6.726
H-(C≡C)$_6$-H	∞	3.741	1.082	9.059
	1907	3.790	1.084	9.203
	1460	3.826	1.085	9.308
	1340	3.843	1.086	9.357
	1064	3.906	1.089	9.542
H-(C≡C)$_7$-H	∞	4.849	1.241	12.066
	1907	4.926	1.243	12.291
	1460	4.982	1.245	12.456
	1340	5.008	1.246	12.532
	1064	5.107	1.249	12.824
H-(C≡C)$_8$-H	∞	6.075	1.400	15.424
	1907	6.186	1.403	15.753
	1460	6.268	1.405	15.995

	1340	6.306	1.406	16.107
	1064	6.452	1.409	16.538
	∞	7.397	1.559	19.073
	1907	7.550	1.562	19.527
H-(C≡C)$_9$-H	1460	7.664	1.564	19.863
	1340	7.716	1.565	20.018
	1064	7.920	1.569	20.622
	∞	8.818	1.718	23.017
	1907	9.021	1.722	23.621
H-(C≡C)$_{10}$-H	1460	9.172	1.724	24.069
	1340	9.242	1.725	24.277
	1064	9.515	1.729	25.089
	∞	10.308	1.877	27.169
	1907	10.568	1.881	27.943
H-(C≡C)$_{11}$-H	1460	10.762	1.883	28.519
	1340	10.852	1.885	28.788
	1064	11.205	1.889	29.839
	∞	11.861	2.037	31.511
	1907	12.185	2.040	32.475
H-(C≡C)$_{12}$-H	1460	12.427	2.043	33.195
	1340	12.540	2.044	33.532
	1064	12.984	2.049	34.853
	∞	13.468	2.196	36.013
	1907	13.861	2.200	37.185
H-(C≡C)$_{13}$-H	1460	14.156	2.203	38.063
	1340	14.294	2.204	38.475
	1064	14.838	2.209	40.095
	∞	15.119	2.355	40.648
	1907	15.587	2.359	42.043
H-(C≡C)$_{14}$-H	1460	15.939	2.362	43.092
	1340	16.104	2.364	43.585
	1064	16.756	2.369	45.532
H-(C≡C)$_{15}$-H	∞	16.808	2.514	45.397

	1907	17.356	2.518	47.031
	1460	17.769	2.522	48.263
	1340	17.963	2.523	48.843
	1064	18.732	2.529	51.140

Table S15 Calculated second hyperpolarizability of the carbon chains H-(C≡C)$_n$-H (n = 3-15) in the zero-frequency limit ($\lambda = \infty$ nm) and in the frequency-dependent fields (λ = 1907, 1460, 1340, and 1064 nm) at the PBE38/aug-cc-pVTZ level

	Incident wavelength (λ, in nm)	$\gamma_{\|\|}(\lambda)$ (in 10^{-34}esu)	$\gamma_{xxxx}(\lambda) = \gamma_{yyyy}(\lambda)$ (in 10^{-34}esu)	$\gamma_{zzzz}(\lambda)$ (in 10^{-34}esu)
	∞	0.149	0.036	0.488
	1907	0.159	0.038	0.525
H-(C≡C)$_3$-H	1460	0.167	0.039	0.554
	1340	0.171	0.039	0.568
	1064	0.186	0.041	0.623
	∞	0.376	0.042	1.537
	1907	0.411	0.044	1.687
H-(C≡C)$_4$-H	1460	0.438	0.045	1.806
	1340	0.451	0.046	1.864
	1064	0.505	0.048	2.103
	∞	0.895	0.048	4.039
	1907	1.000	0.050	4.535
H-(C≡C)$_5$-H	1460	1.086	0.051	4.939
	1340	1.128	0.052	5.139
	1064	1.308	0.054	5.995
	∞	1.956	0.054	9.248
	1907	2.243	0.056	10.645
H-(C≡C)$_6$-H	1460	2.484	0.058	11.819
	1340	2.605	0.058	12.412
	1064	3.143	0.061	15.035
	∞	3.905	0.060	18.902
	1907	4.599	0.062	22.321
H-(C≡C)$_7$-H	1460	5.201	0.064	25.292
	1340	5.511	0.065	26.824
	1064	6.936	0.067	33.861
	∞	7.172	0.066	35.143
	1907	8.672	0.069	42.582
H-(C≡C)$_8$-H	1460	10.018	0.070	49.260
	1340	10.727	0.071	52.779
	1064	14.111	0.074	69.590
H-(C≡C)$_9$-H	∞	12.220	0.072	60.286

	1907	15.159	0.075	74.907
	1460	17.882	0.076	88.459
	1340	19.347	0.077	95.756
	1064	26.632	0.080	132.041
$H\text{-}(C{\equiv}C)_{10}\text{-}H$	∞	19.541	0.078	96.795
	1907	24.850	0.081	123.253
	1460	29.926	0.083	148.557
	1340	32.717	0.084	162.478
	1064	47.178	0.087	234.610
$H\text{-}(C{\equiv}C)_{11}\text{-}H$	∞	29.525	0.084	146.621
	1907	38.437	0.087	191.079
	1460	47.216	0.089	234.888
	1340	52.147	0.090	259.493
	1064	78.756	0.094	392.328
$H\text{-}(C{\equiv}C)_{12}\text{-}H$	∞	42.518	0.090	211.496
	1907	56.586	0.093	281.715
	1460	70.843	0.095	352.898
	1340	79.012	0.096	393.685
	1064	124.934	0.100	623.040
$H\text{-}(C{\equiv}C)_{13}\text{-}H$	∞	58.748	0.096	292.547
	1907	79.810	0.099	397.723
	1460	101.734	0.102	507.226
	1340	114.537	0.103	571.177
	1064	189.477	0.107	945.570
$H\text{-}(C{\equiv}C)_{14}\text{-}H$	∞	78.333	0.102	390.377
	1907	108.466	0.106	540.891
	1460	140.627	0.108	701.562
	1340	159.751	0.109	797.108
	1064	276.208	0.113	1379.028
$H\text{-}(C{\equiv}C)_{15}\text{-}H$	∞	101.307	0.109	505.156
	1907	142.773	0.112	712.315
	1460	188.080	0.114	938.695
	1340	215.485	0.115	1075.638

– – – – – – – – – – –	1064	388.954	0.120	1942.554