Supporting Information

Theoretical Determination of Linear and nonlinear Optical Properties as well as Electric Anisotropies of Elements of Periodic Table: A Density Functional Theory Study

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Table S1 Static dipole polarizabilities (ᾱ/a.u.) calculated using the quadruple-zeta quality def2-QZVPPD basis set.

	$\alpha_{0\,(Tl)}$	$\alpha_{0\,(Au)}$	$\alpha_{0\,(At)}$	$\alpha_{0\,(Ba)}$	$\alpha_{0\,(Bi)}$	$\alpha_{0\,(Hg)}$
B3LYP	79.26	35.52	40.70	256.8	53.04	35.76
B3PW	78.34	36.03	40.11	273.8	52.32	35.68
BLYP	86.02	34.76	42.38	250.3	55.60	34.95
BP86	83.09	34.93	41.74	253.6	54.75	34.60
M06L	80.32	38.87	42.56	205.3	55.33	35.74
B1LYP	79.34	35.97	40.62	260.0	52.83	36.27
mPW1LYP	78.81	35.97	40.70	259.2	52.74	36.36
mPW1PW	77.23	36.54	39.88	279.5	51.78	36.20
PBEPBE	84.81	35.33	41.93	269.4	54.95	35.10
TPSSTPSS	82.31	35.99	41.30	308.5	53.63	35.80
TPSSh	79.82	36.47	40.65	309.0	52.63	36.29
LC-BLYP	63.57	38.05	37.76	274.1	47.80	37.37
BHandHLYP	73.93	37.64	39.07	270.6	50.44	37.80
wB97X	72.35	38.15	40.05	245.3	51.09	37.75
CAM-B3LYP	71.60	36.65	39.58	263.9	50.89	36.62
Other theor. /exp. results	70.05[a]	36.06±0.54[b]	40.7±2.0[c]	262.2[d]	50[e]	39.1[f], 34.42[g]
	$\alpha_{0\,(Kr)}$	$\alpha_{0\,(Br)}$	$\alpha_{0\,(Po)}$	$\alpha_{0\,(I)}$	$\alpha_{0\,(Rn)}$	$\alpha_{0\,(Xe)}$
B3LYP	17.64	21.77	47.59	33.77	34.84	28.36
B3PW	17.32	21.35	46.90	33.15	34.33	27.86
BLYP	18.29	22.64	49.82	35.08	36.13	29.37
BP86	18.04	22.32	49.03	34.49	35.60	28.90
M06L	17.52	21.62	49.23	37.48	35.88	30.76
B1LYP	17.59	21.71	47.49	33.71	34.77	28.31
mPW1LYP	17.58	21.70	47.55	33.77	34.83	28.35
mPW1PW	17.17	21.16	46.58	32.96	34.14	27.70
PBEPBE	18.00	22.27	49.26	34.55	35.73	28.92
TPSSTPSS	17.59	21.72	48.46	33.84	35.23	28.36
TPSSh	17.36	21.41	47.59	33.36	34.73	28.00
LC-BLYP	16.97	20.64	43.28	31.69	32.87	27.01
BHandHLYP	16.98	20.90	45.45	32.52	33.56	27.38
wB97X	17.45	21.39	46.04	33.23	34.54	28.10
CAM-B3LYP	17.40	21.36	45.87	32.97	34.11	27.85
Other theor. /exp. results	17.075[h], 16.8[i]	21.13±0.42[j]	46[k]	32.9 ±1.3[l]	34.2[m]	27.82[n]

a. Configuration interaction (CI)[26]; b. CCSD(T)[60,89]; c. Cowan-Griffin, HF only[90,91]; d. CI, MBPT[92,93]; e. Dirac, LDA[94,95]; f. RPA, PolPlot[96]; g. CCSD(T)[97]; h. exp.[98]; i. CCSD(T)[98]; j. CASPT2[21]; k. Dirac[94,95]; l. exp.[99]; m. RPA, PolPlot[96]; n. exp.[98]

Table S2 Mean static polarizabilities (α(0;0)≡ᾱ/a.u.) together with the corresponding components for the elements of Periodic Table H-Rn (except lanthanides) computed at the CAM-B3LYP/def2-QZVPPD level.

	α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$		α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$
1					11				
$_{1}$H	5.2122	5.2122	5.2122	5.2122	$_{29}$Cu	45.840	45.840	45.840	45.840
$_{3}$Li	146.49	146.49	146.49	146.49	$_{47}$Ag	49.769	49.769	49.769	49.769
$_{11}$Na	146.21	146.21	146.21	146.21	$_{79}$Au	36.645	36.645	36.645	36.645
$_{19}$K	291.01	291.01	291.01	291.01	12				
$_{37}$Rb	318.62	318.62	318.62	318.62	$_{30}$Zn	41.594	41.594	41.594	41.594
$_{55}$Cs	418.90	418.90	418.90	418.90	$_{48}$Cd	48.334	48.334	48.334	48.334
2					$_{80}$Hg	36.614	36.614	36.614	36.614
$_{4}$Be	43.057	43.057	43.057	43.057	13				
$_{12}$Mg	72.086	72.086	72.086	72.086	$_{5}$B	19.970	19.970	25.877	21.939
$_{20}$Ca	156.29	156.29	156.29	156.29	$_{13}$Al	50.593	50.593	79.850	60.345
$_{38}$Sr	190.40	190.40	190.40	190.40	$_{31}$Ga	43.063	43.063	77.646	54.591
$_{56}$Ba	263.81	263.81	263.81	263.81	$_{49}$In	55.108	55.108	98.671	69.629
3					$_{81}$Tl	52.233	52.233	110.35	71.605
$_{21}$Sc	122.08	122.08	129.57	124.58	14				
$_{39}$Y	146.53	146.53	149.18	147.41	$_{6}$C	13.126	13.126	10.910	12.387
$_{57}$La	198.26	198.26	212.94	203.15	$_{14}$Si	42.121	42.121	32.554	38.932
4					$_{32}$Ge	45.054	45.054	32.914	41.007
$_{22}$Ti	106.74	106.74	101.16	104.88	$_{50}$Sn	61.752	61.752	45.066	56.190
$_{40}$Zr	120.69	120.69	114.09	118.49	$_{82}$Pb	69.879	69.879	45.767	61.842
$_{72}$Hf	100.02	100.02	100.01	100.02	15				
5					$_{7}$N	7.6294	7.6294	7.6294	7.6294
$_{23}$V	91.155	91.155	94.377	92.229	$_{15}$P	26.114	26.114	26.114	26.114
$_{41}$Nb	94.291	94.291	100.93	96.504	$_{33}$As	31.036	31.036	31.036	31.036
$_{73}$Ta	82.951	82.951	82.951	82.951	$_{51}$Sb	44.701	44.701	44.701	44.701
6					$_{83}$Bi	50.888	50.888	50.888	50.888
$_{24}$Cr	77.338	77.338	77.338	77.338	16				
$_{42}$Mo	78.705	78.705	78.705	78.705	$_{8}$O	5.1454	5.1454	6.2111	5.5006
$_{74}$W	67.030	67.030	67.030	67.030	$_{16}$S	18.837	18.837	22.852	20.175
7					$_{34}$Se	24.487	24.487	29.921	26.298
$_{25}$Mn	67.827	67.846	68.441	68.038	$_{52}$Te	36.580	36.580	44.180	39.113
$_{43}$Tc	80.864	80.864	80.864	80.864	$_{84}$Po	42.342	42.342	52.939	45.874
$_{75}$Re	64.614	64.614	64.614	64.614	17				
8					$_{9}$F	4.0685	4.0685	3.5556	3.8975
$_{26}$Fe	59.142	59.142	62.052	60.112	$_{17}$Cl	15.806	15.806	13.813	15.142
$_{44}$Ru	62.109	62.109	64.404	62.874	$_{35}$Br	22.321	22.321	19.443	21.362
$_{76}$Os	55.500	55.500	56.932	55.977	$_{53}$I	34.381	34.381	30.132	32.965
9					$_{85}$At	41.640	41.640	35.474	39.585
$_{27}$Co	53.685	53.685	53.685	53.685	18				
$_{45}$Rh	57.626	57.626	57.626	57.626	$_{2}$He	1.5020	1.5020	1.5020	1.5020
$_{77}$Ir	44.239	44.239	44.239	44.239	$_{10}$Ne	2.8339	2.8339	2.8339	2.8339
10					$_{18}$Ar	11.517	11.517	11.517	11.517
$_{28}$Ni	49.743	49.743	48.042	49.176	$_{36}$Kr	17.402	17.402	17.402	17.402
$_{46}$Pd	24.160	24.160	24.160	24.160	$_{54}$Xe	27.846	27.846	27.846	27.846
$_{78}$Pt	40.120	40.120	40.342	40.194	$_{86}$Rn	34.114	34.114	34.114	34.114

Table S3 Mean static polarizabilities (α(0;0)≡ᾱ/a.u.) together with the corresponding components for the elements of Periodic Table H-Rn (except lanthanides) computed at the B3LYP/def2-QZVPPD level.

	α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$		α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$
1					11				
$_{1}$H	5.1721	5.1721	5.1721	5.1721	$_{29}$Cu	44.193	44.193	44.193	44.193
$_{3}$Li	142.48	142.48	142.48	142.48	$_{47}$Ag	47.447	47.447	47.447	47.447
$_{11}$Na	144.60	144.60	144.60	144.60	$_{79}$Au	35.515	35.515	35.515	35.515
$_{19}$K	277.29	277.29	277.29	277.29	12				
$_{37}$Rb	303.93	303.93	303.93	303.93	$_{30}$Zn	41.064	41.064	41.064	41.064
$_{55}$Cs	393.11	393.11	393.11	393.11	$_{48}$Cd	47.170	47.170	47.170	47.170
2					$_{80}$Hg	35.755	35.755	35.755	35.755
$_{4}$Be	43.004	43.004	43.004	43.004	13				
$_{12}$Mg	72.245	72.245	72.245	72.245	$_{5}$B	20.189	20.189	26.842	22.407
$_{20}$Ca	154.06	154.06	154.06	154.06	$_{13}$Al	52.146	52.146	85.389	63.227
$_{38}$Sr	187.06	187.06	187.06	187.06	$_{31}$Ga	44.921	44.921	84.901	58.248
$_{56}$Ba	256.69	256.69	256.69	256.69	$_{49}$In	58.031	58.031	110.12	75.394
3					$_{81}$Tl	56.249	56.249	125.28	79.259
$_{21}$Sc	119.38	119.38	127.01	121.92	14				
$_{39}$Y	142.21	142.21	144.57	143.00	$_{6}$C	13.389	13.389	10.982	12.587
$_{57}$La	187.03	187.03	198.75	190.94	$_{14}$Si	43.662	43.662	33.188	40.171
4					$_{32}$Ge	47.161	47.161	33.780	42.701
$_{22}$Ti	104.64	104.64	98.841	102.71	$_{50}$Sn	65.443	65.443	46.606	59.164
$_{40}$Zr	117.04	117.04	109.92	114.67	$_{82}$Pb	74.747	74.747	47.984	65.826
$_{72}$Hf	97.223	97.223	98.702	97.716	15				
5					$_{7}$N	7.7056	7.7056	7.7056	7.7056
$_{23}$V	89.739	89.739	89.739	89.739	$_{15}$P	26.653	26.653	26.653	26.653
$_{41}$Nb	86.371	86.371	92.745	88.496	$_{33}$As	31.908	31.908	31.908	31.908
$_{73}$Ta	81.091	81.091	81.091	81.091	$_{51}$Sb	46.266	46.266	46.266	46.266
6					$_{83}$Bi	53.038	53.038	53.038	53.038
$_{24}$Cr	71.308	71.308	71.308	71.308	16				
$_{42}$Mo	72.178	72.178	72.178	72.178	$_{8}$O	5.1707	5.1707	6.2839	5.5418
$_{74}$W	62.987	62.987	62.987	62.987	$_{16}$S	19.130	19.130	23.432	20.564
7					$_{34}$Se	25.015	25.015	30.924	26.985
$_{25}$Mn	71.472	71.472	71.472	71.472	$_{52}$Te	37.566	37.566	45.935	40.356
$_{43}$Tc	79.368	79.368	69.897	76.211	$_{84}$Po	43.752	43.752	55.268	47.591
$_{75}$Re	63.453	63.453	63.453	63.453	17				
8					$_{9}$F	4.0865	4.0865	3.5582	3.9104
$_{26}$Fe	56.323	56.323	54.932	55.859	$_{17}$Cl	16.044	16.044	13.943	15.344
$_{44}$Ru	58.846	58.846	58.836	58.843	$_{35}$Br	22.792	22.792	19.723	21.769
$_{76}$Os	54.320	54.320	55.768	54.803	$_{53}$I	35.290	35.290	30.710	33.763
9					$_{85}$At	42.892	42.892	36.342	40.709
$_{27}$Co	50.905	50.905	50.905	50.905	18				
$_{45}$Rh	54.188	54.188	54.188	54.188	$_{2}$He	1.4829	1.4829	1.4829	1.4829
$_{77}$Ir	43.162	43.162	43.162	43.162	$_{10}$Ne	2.8344	2.8344	2.8344	2.8344
10					$_{18}$Ar	11.617	11.617	11.617	11.617
$_{28}$Ni	47.846	47.846	46.308	47.333	$_{36}$Kr	17.639	17.639	17.639	17.639
$_{46}$Pd	25.005	25.005	25.005	25.005	$_{54}$Xe	28.352	28.352	28.352	28.352
$_{78}$Pt	38.904	38.904	39.163	38.990	$_{86}$Rn	34.843	34.843	34.843	34.843

Table S4 Electric anisotropies (α_2/a.u.) of the elements of Periodic Table H-Rn (except lanthanides) computed at the B3LYP/def2-QZVPPD and CAM-B3LYP/def2-QZVPPD levels.

	State	α_2	State	α_2		State	α_2	State	α_2
	B3LYP		CAM-B3LYP			B3LYP		CAM-B3LYP	
1					$_{29}$Cu	^{2}S	0.0000	^{2}S	0.0000
$_1$H	^{2}S	0.0000	^{2}S	0.0000	$_{47}$Ag	^{2}S	0.0000	^{2}S	0.0000
$_3$Li	^{2}S	0.0000	^{2}S	0.0000	$_{79}$Au	^{2}S	0.0000	^{2}S	0.0000
$_{11}$Na	^{2}S	0.0000	^{2}S	0.0000	12				
$_{19}$K	^{2}S	0.0000	^{2}S	0.0000	$_{30}$Zn	^{1}S	0.0000	^{1}S	0.0000
$_{37}$Rb	^{2}S	0.0000	^{2}S	0.0000	$_{48}$Cd	^{1}S	0.0000	^{1}S	0.0000
$_{55}$Cs	^{2}S	0.0000	^{2}S	0.0000	$_{80}$Hg	^{1}S	0.0000	^{1}S	0.0000
2					13				
$_4$Be	^{1}S	0.0000	^{1}S	0.0000	$_5$B	^{2}P	6.6531	^{2}P	5.9071
$_{12}$Mg	^{1}S	0.0000	^{1}S	0.0000	$_{13}$Al	^{2}P	33.243	^{2}P	29.257
$_{20}$Ca	^{1}S	0.0000	^{1}S	0.0000	$_{31}$Ga	^{2}P	39.979	^{2}P	34.583
$_{38}$Sr	^{1}S	0.0000	^{1}S	0.0000	$_{49}$In	^{2}P	52.088	^{2}P	43.564
$_{56}$Ba	^{1}S	0.0000	^{1}S	0.0000	$_{81}$Tl	^{2}P	69.035	^{2}P	58.113
3					14				
$_{21}$Sc	^{2}D	7.6260	^{2}D	7.4861	$_6$C	^{3}P	-2.4070	^{3}P	-2.2160
$_{39}$Y	^{2}D	2.3633	^{2}D	2.6517	$_{14}$Si	^{3}P	-10.475	^{3}P	-9.5666
4					$_{32}$Ge	^{3}P	-13.382	^{3}P	-12.139
$_{22}$Ti	^{3}F	-5.7992	^{3}F	-5.5811	$_{50}$Sn	^{3}P	-18.837	^{3}P	-16.686
$_{40}$Zr	^{3}F	-7.1173	^{3}F	-6.6008	$_{82}$Pb	^{3}P	-26.763	^{3}P	-24.113
$_{72}$Hf	^{3}F	1.4791	^{3}F	-0.0019	15				
5					$_7$N	^{4}S	0.0000	^{4}S	0.0000
$_{23}$V	^{4}F	0.0000	^{6}D	3.2218	$_{15}$P	^{4}S	0.0000	^{4}S	0.0000
$_{41}$Nb	^{6}D	6.3736	^{6}D	6.6427	$_{33}$As	^{4}S	0.0000	^{4}S	0.0000
$_{73}$Ta	^{4}F	0.0000	^{4}F	-0.0002	$_{51}$Sb	^{4}S	0.0000	^{4}S	0.0000
6					$_{83}$Bi	^{4}S	0.0000	^{4}S	0.0000
$_{24}$Cr	^{7}S	0.0000	^{7}S	0.0000	16				
$_{42}$Mo	^{7}S	0.0000	^{7}S	0.0000	$_8$O	^{3}P	1.1132	^{3}P	1.0658
$_{74}$W	^{7}S	0.0000	^{7}S	0.0000	$_{16}$S	^{3}P	4.3019	^{3}P	4.0148
7					$_{34}$Se	^{3}P	5.9091	^{3}P	5.4341
$_{25}$Mn	^{6}S	0.0000	^{6}D	0.6049	$_{52}$Te	^{3}P	8.3686	^{3}P	7.6005
$_{43}$Tc	-	-9.4714	^{6}S	0.0000	$_{84}$Po	^{3}P	11.516	^{3}P	10.597
$_{75}$Re	^{6}S	0.0000	^{6}S	0.0000	17				
8					$_9$F	^{2}P	-0.5283	^{2}P	-0.5129
$_{26}$Fe	^{5}F	-1.3915	^{5}F	2.9100	$_{17}$Cl	^{2}P	-2.1006	^{2}P	-1.9926
$_{44}$Ru	^{5}F	-0.0097	^{5}F	2.2949	$_{35}$Br	^{2}P	-3.0690	^{2}P	-2.8781
$_{76}$Os	^{5}D	1.4473	^{5}D	1.4324	$_{53}$I	^{2}P	-4.5797	^{2}P	-4.2491
9					$_{85}$At	^{2}P	-6.5509	^{2}P	-6.1661
$_{27}$Co	^{4}F	0.0000	^{4}F	0.0000	18				
$_{45}$Rh	^{4}F	0.0000	^{4}F	0.0000	$_2$He	^{1}S	0.0000	^{1}S	0.0000
$_{77}$Ir	^{4}F	0.0000	^{4}F	0.0000	$_{10}$Ne	^{1}S	0.0000	^{1}S	0.0000
10					$_{18}$Ar	^{1}S	0.0000	^{1}S	0.0000
$_{28}$Ni	^{3}D	-1.5377	^{3}D	-1.7011	$_{36}$Kr	^{1}S	0.0000	^{1}S	0.0000
$_{46}$Pd	^{1}S	0.0000	^{1}S	0.0000	$_{54}$Xe	^{1}S	0.0000	^{1}S	0.0000
$_{78}$Pt	^{3}D	0.2587	^{3}D	0.2228	$_{86}$Rn	^{1}S	0.0000	^{1}S	0.0000
11									

At the B3LYP/def2-QZVPPD level of theory, the ground-configuration for Tc is $[Kr]4d^{5.21}5S^{1.78}$; so that 0.16 and 0.05 electrons occupy the β-spin d_{x2+y2} and d_{z2} orbitals, respectively, resulting in a negative anisotropy for this atom.

Table S5 Frequency-dependent mean static polarizabilities ($\alpha(-\omega;\omega)\equiv\bar{\alpha}$/a.u.) together with the corresponding components and polarizability anisotropies (α_2/a.u.) for the elements of Periodic Table H-Rn (except lanthanides) computed at the CAM-B3LYP/def2-QZVPPD level (λ = 1064.0 nm).

	α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$	α_2		α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$	α_2
1						11					
$_{1}$H	5.2806	5.2806	5.2806	5.2806	0.0000	$_{29}$Cu	49.526	49.526	49.526	49.526	0.0000
$_{3}$Li	227.79	227.79	227.79	227.79	0.0000	$_{47}$Ag	53.628	53.628	53.628	53.628	0.0000
$_{11}$Na	201.96	201.96	201.96	201.96	0.0000	$_{79}$Au	37.875	37.875	37.875	37.875	0.0000
$_{19}$K	606.80	606.80	606.80	606.80	0.0000	12					
$_{37}$Rb	686.86	686.86	686.86	686.86	0.0000	$_{30}$Zn	43.329	43.329	43.329	43.329	0.0000
$_{55}$Cs	1313.5	1313.5	1313.5	1313.5	0.0000	$_{48}$Cd	50.456	50.456	50.456	50.456	0.0000
2						$_{80}$Hg	37.525	37.525	37.525	37.525	0.0000
$_{4}$Be	45.577	45.577	45.577	45.577	0.0000	13					
$_{12}$Mg	77.666	77.666	77.666	77.666	0.0000	$_{5}$B	20.398	20.398	26.813	22.536	6.4152
$_{20}$Ca	182.39	182.39	182.39	182.39	0.0000	$_{13}$Al	52.922	52.922	87.959	64.601	35.037
$_{38}$Sr	227.21	227.21	227.21	227.21	0.0000	$_{31}$Ga	44.872	44.872	86.229	58.658	41.357
$_{56}$Ba	335.06	335.06	335.06	335.06	0.0000	$_{49}$In	58.003	58.003	111.61	75.872	53.609
3						$_{81}$Tl	55.193	55.193	128.81	79.732	73.617
$_{21}$Sc	137.28	137.28	148.03	140.86	10.754	14					
$_{39}$Y	181.36	181.36	171.58	178.10	-9.7788	$_{6}$C	13.320	13.320	11.020	12.553	-2.2998
$_{57}$La	240.23	240.23	266.10	248.85	25.862	$_{14}$Si	43.552	43.552	33.246	40.117	-10.306
4						$_{32}$Ge	46.721	46.721	33.618	42.353	-13.103
$_{22}$Ti	118.89	118.89	111.67	116.48	-7.2185	$_{50}$Sn	64.652	64.652	46.339	58.548	-18.313
$_{40}$Zr	134.04	134.04	125.28	131.12	-8.7601	$_{82}$Pb	73.928	73.928	47.181	65.012	-26.747
$_{72}$Hf	110.94	110.94	110.94	110.94	-0.0034	15					
5						$_{7}$N	7.6851	7.6851	7.6851	7.6851	0.0000
$_{23}$V	106.33	106.33	110.57	107.74	4.2416	$_{15}$P	26.528	26.528	26.528	26.528	0.0000
$_{41}$Nb	106.82	106.82	115.68	109.77	8.8650	$_{33}$As	31.598	31.598	31.598	31.598	0.0000
$_{73}$Ta	88.161	88.161	88.161	88.161	-0.0002	$_{51}$Sb	45.768	45.768	45.768	45.768	0.0000
6						$_{83}$Bi	52.343	52.343	52.343	52.343	0.0000
$_{24}$Cr	87.771	87.771	87.771	87.771	0.0000	16					
$_{42}$Mo	86.834	86.834	86.834	86.834	0.0000	$_{8}$O	5.1665	5.1665	6.2540	5.5290	1.0875
$_{74}$W	72.504	72.504	72.504	72.504	0.0000	$_{16}$S	19.020	19.020	23.177	20.406	4.1571
7						$_{34}$Se	24.770	24.770	30.430	26.657	5.6599
$_{25}$Mn	77.122	77.214	76.919	77.085	-0.2620	$_{52}$Te	37.155	37.155	45.164	39.825	8.0087
$_{43}$Tc	87.377	87.377	87.377	87.377	0.0000	$_{84}$Po	43.126	43.126	54.367	46.873	11.240
$_{75}$Re	67.756	67.756	67.756	67.756	0.0000	17					
8						$_{9}$F	4.0834	4.0834	3.5643	3.9104	-0.5190
$_{26}$Fe	65.011	65.011	69.541	66.521	4.5300	$_{17}$Cl	15.932	15.932	13.898	15.254	-2.0347
$_{44}$Ru	66.814	66.814	71.384	68.337	4.5695	$_{35}$Br	22.545	22.545	19.593	21.561	-2.9522
$_{76}$Os	57.660	57.660	59.348	58.223	1.6881	$_{53}$I	34.852	34.852	30.458	33.387	-4.3937
9						$_{85}$At	42.323	42.323	35.925	40.190	-6.3990
$_{27}$Co	58.719	58.719	58.719	58.719	0.0000	18					
$_{45}$Rh	62.289	62.289	62.289	62.289	0.0000	$_{2}$He	1.5054	1.5054	1.5054	1.5054	0.0000
$_{77}$Ir	45.919	45.919	45.919	45.919	0.0000	$_{10}$Ne	2.8401	2.8401	2.8401	2.8401	0.0000
10						$_{18}$Ar	11.574	11.574	11.574	11.574	0.0000
$_{28}$Ni	53.983	53.983	51.947	53.304	-2.0366	$_{36}$Kr	17.516	17.516	17.516	17.516	0.0000
$_{46}$Pd	24.974	24.974	24.974	24.974	0.0000	$_{54}$Xe	28.103	28.103	28.103	28.103	0.0000
$_{78}$Pt	41.510	41.510	41.751	41.590	0.2408	$_{86}$Rn	34.492	34.492	34.492	34.492	0.0000

Table S6 Frequency-dependent mean static polarizabilities ($\alpha(-\omega;\omega)\equiv\bar{\alpha}$/a.u.) together with the corresponding components and polarizability anisotropies (α_2/a.u.) for the elements of Periodic Table H-Rn (except lanthanides) computed at the B3LYP/def2-QZVPPD level (λ = 1064.0 nm).

	α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$	α_2		α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$	α_2
1						11					
$_{1}$H	5.2400	5.2400	5.2400	5.2400	0.0000	$_{29}$Cu	47.536	47.536	47.536	47.536	0.0000
$_{3}$Li	217.36	217.36	217.36	217.36	0.0000	$_{47}$Ag	50.820	50.820	50.820	50.820	0.0000
$_{11}$Na	199.29	199.29	199.29	199.29	0.0000	$_{79}$Au	36.616	36.616	36.616	36.616	0.0000
$_{19}$K	552.77	552.77	552.77	552.77	0.0000	12					
$_{37}$Rb	627.06	627.06	627.06	627.06	0.0000	$_{30}$Zn	42.755	42.755	42.755	42.755	0.0000
$_{55}$Cs	1099.4	1099.4	1099.4	1099.4	0.0000	$_{48}$Cd	49.174	49.174	49.174	49.174	0.0000
2						$_{80}$Hg	36.610	36.610	36.610	36.610	0.0000
$_{4}$Be	45.513	45.513	45.513	45.513	0.0000	13					
$_{12}$Mg	77.889	77.889	77.889	77.889	0.0000	$_{5}$B	20.628	20.628	27.902	23.053	7.2736
$_{20}$Ca	179.55	179.55	179.55	179.55	0.0000	$_{13}$Al	54.797	54.797	95.606	68.400	40.810
$_{38}$Sr	222.75	222.75	222.75	222.75	0.0000	$_{31}$Ga	47.084	47.084	96.384	63.517	49.300
$_{56}$Ba	324.34	324.34	324.34	324.34	0.0000	$_{49}$In	61.684	61.684	128.49	83.953	66.807
3						$_{81}$Tl	60.265	60.265	152.75	91.093	92.489
$_{21}$Sc	133.96	133.96	145.05	137.66	11.096	14					
$_{39}$Y	172.33	172.33	165.27	169.98	-7.0563	$_{6}$C	13.596	13.596	11.094	12.762	-2.5021
$_{57}$La	224.17	224.17	241.63	229.99	17.460	$_{14}$Si	45.281	45.281	33.930	41.497	-11.351
4						$_{32}$Ge	49.100	49.100	34.554	44.251	-14.546
$_{22}$Ti	116.39	116.39	108.84	113.87	-7.5424	$_{50}$Sn	68.938	68.938	48.054	61.977	-20.884
$_{40}$Zr	129.66	129.66	120.34	126.55	-9.3213	$_{82}$Pb	79.720	79.720	49.665	69.702	-30.055
$_{72}$Hf	107.18	107.18	111.88	108.75	4.6979	15					
5						$_{7}$N	7.7632	7.7632	7.7632	7.7632	0.0000
$_{23}$V	98.426	98.426	98.426	98.426	0.0000	$_{15}$P	27.097	27.097	27.097	27.097	0.0000
$_{41}$Nb	96.730	96.730	105.29	99.583	8.5588	$_{33}$As	32.523	32.523	32.523	32.523	0.0000
$_{73}$Ta	86.135	86.135	86.135	86.135	0.0000	$_{51}$Sb	47.459	47.459	47.459	47.459	0.0000
6						$_{83}$Bi	54.690	54.690	54.690	54.690	0.0000
$_{24}$Cr	79.977	79.977	79.977	79.977	0.0000	16					
$_{42}$Mo	78.744	78.744	78.744	78.744	0.0000	$_{8}$O	5.1923	5.1923	6.3284	5.5710	1.1361
$_{74}$W	0.67.704	67.704	67.704	67.704	0.0000	$_{16}$S	19.322	19.322	23.783	20.809	4.4615
7						$_{34}$Se	25.317	25.317	31.485	27.373	6.1679
$_{25}$Mn	77.135	77.135	77.135	77.135	0.0000	$_{52}$Te	38.192	38.192	47.041	41.142	8.8490
$_{43}$Tc	85.740	85.740	75.208	82.229	-10.532	$_{84}$Po	44.616	44.616	56.886	48.706	12.270
$_{75}$Re	66.524	66.524	66.524	66.524	0.0000	17					
8						$_{9}$F	4.1016	4.1016	3.5670	3.9234	-0.5346
$_{26}$Fe	61.751	61.751	59.645	61.049	-2.1059	$_{17}$Cl	16.177	16.177	14.030	15.461	-2.1464
$_{44}$Ru	63.177	63.177	63.169	63.174	-0.0085	$_{35}$Br	23.030	23.030	19.880	21.980	-3.1508
$_{76}$Os	56.388	56.388	58.073	56.950	1.6849	$_{53}$I	35.799	35.799	31.056	34.218	-4.7429
9						$_{85}$At	43.637	43.637	36.826	41.367	-6.8108
$_{27}$Co	55.326	55.326	55.326	55.326	0.0000	18					
$_{45}$Rh	58.101	58.101	58.101	58.101	0.0000	$_{2}$He	1.4863	1.4863	1.4863	1.4863	0.0000
$_{77}$Ir	44.783	44.783	44.783	44.783	0.0000	$_{10}$Ne	2.8406	2.8406	2.8406	2.8406	0.0000
10						$_{18}$Ar	11.677	11.677	11.677	11.677	0.0000
$_{28}$Ni	51.663	51.663	49.829	51.052	-1.8346	$_{36}$Kr	17.758	17.758	17.758	17.758	0.0000
$_{46}$Pd	25.912	25.912	25.912	25.912	0.0000	$_{54}$Xe	28.624	28.624	28.624	28.624	0.0000
$_{78}$Pt	40.151	40.151	40.431	40.244	0.2797	$_{86}$Rn	35.244	35.244	35.244	35.244	0.0000

Table S7 Isotropic average of scalar component for static $\gamma_{||}(0;0,0,0)$, dc-Kerr $\gamma_{||}(-\omega;\omega,0,0)$, and electric field-induced second-harmonic generation $\gamma_{||}(-2\omega;\omega,\omega,0)$ second-order hyperpolarizabilities for the elements of Periodic Table H-Rn (except lanthanides) calculated at the CAM-B3LYP/def2-QZVPPD level ($\gamma_{||}$/a.u.), in the wavelength λ = 1064.0 nm. A(n) means $A\times10^{n}$.

	$\gamma_{\|\|}(0;0,0,0)$	$\gamma_{\|\|}(-\omega;\omega,0,0)$	$\gamma_{\|\|}(-2\omega;\omega,\omega,0)$		$\gamma_{\|\|}(0;0,0,0)$	$\gamma_{\|\|}(-\omega;\omega,0,0)$	$\gamma_{\|\|}(-2\omega;\omega,\omega,0)$
1				11			
$_{1}$H	5.00(2)	5.24(2)	5.72(2)	$_{29}$Cu	5.04(4)	6.58(4)	1.06(5)
$_{3}$Li	-3.47(5)	-1.16(6)	2.62(6)	$_{47}$Ag	1.80(4)	2.63(4)	4.61(4)
$_{11}$Na	5.77(2)	1.73(5)	-6.71(6)	$_{79}$Au	8.96(3)	1.08(4)	1.40(4)
$_{19}$K	-1.70(6)	-5.16(6)	7.30(5)	12			
$_{37}$Rb	5.05(6)	3.72(7)	-2.79(8)	$_{30}$Zn	2.26(4)	2.64(4)	3.32(4)
$_{55}$Cs	1.01(7)	2.26(8)	2.67(8)	$_{48}$Cd	1.44(4)	1.74(4)	2.34(4)
2				$_{80}$Hg	1.14(4)	1.28(4)	1.52(4)
$_{4}$Be	1.61(4)	1.81(4)	2.33(4)	13			
$_{12}$Mg	8.40(4)	1.06(5)	1.61(5)	$_{5}$B	-1.61(5)	-4.78(4)	1.63(4)
$_{20}$Ca	3.30(5)	5.45(5)	1.01(6)	$_{13}$Al	-4.20(7)	-1.62(7)	3.22(5)
$_{38}$Sr	7.43(5)	1.29(6)	6.48(6)	$_{31}$Ga	-3.50(7)	-1.28(7)	1.81(5)
$_{56}$Ba	6.84(5)	2.06(6)	1.81(7)	$_{49}$In	-5.51(7)	-2.18(7)	1.28(6)
3				$_{81}$Tl	-3.09(7)	-1.24(7)	3.25(6)
$_{21}$Sc	-2.30(5)	2.75(5)	9.55(5)	14			
$_{39}$Y	2.46(5)	3.03(5)	-2.32(5)	$_{6}$C	-5.05(4)	-1.56(4)	2.74(3)
$_{57}$La	-3.33(6)	-1.32(6)	3.43(6)	$_{14}$Si	4.66(6)	1.73(6)	3.12(4)
4				$_{32}$Ge	-3.80(8)	6.03(7)	2.60(4)
$_{22}$Ti	1.39(5)	2.31(5)	3.93(5)	$_{50}$Sn	-5.98(7)	2.04(7)	1.31(5)
$_{40}$Zr	-1.06(5)	1.99(5)	3.93(5)	$_{82}$Pb	-1.81(8)	-5.19(7)	1.87(5)
$_{72}$Hf	6.12(4)	6.02(4)	1.74(5)	15			
5				$_{7}$N	5.88(2)	6.03(2)	6.36(2)
$_{23}$V	-4.51(6)	-2.88(6)	5.08(5)	$_{15}$P	5.38(3)	5.82(3)	6.58(3)
$_{41}$Nb	-2.94(9)	2.17(8)	8.15(5)	$_{33}$As	8.27(3)	9.10(3)	1.05(4)
$_{73}$Ta	4.99(4)	6.17(4)	1.95(5)	$_{51}$Sb	2.39(4)	2.71(4)	3.33(4)
6				$_{83}$Bi	3.15(4)	3.65(4)	4.64(4)
$_{24}$Cr	8.36(4)	1.26(5)	3.15(5)	16			
$_{42}$Mo	4.34(4)	6.48(4)	1.45(5)	$_{8}$O	-2.23(3)	-3.98(2)	5.66(2)
$_{74}$W	1.54(2)	-2.76(2)	-1.05(4)	$_{16}$S	2.16(6)	7.41(5)	6.08(3)
7				$_{34}$Se	-1.58(6)	-5.35(5)	1.04(4)
$_{25}$Mn	-6.74(5)	-2.29(7)	-8.32(6)	$_{52}$Te	-4.03(6)	-1.38(6)	2.62(4)
$_{43}$Tc	1.27(5)	9.62(4)	1.26(5)	$_{84}$Po	-1.76(6)	-5.97(5)	4.04(4)
$_{75}$Re	3.16(4)	4.49(4)	5.53(4)	17			
8				$_{9}$F	-1.05(3)	-2.21(2)	2.06(2)
$_{26}$Fe	1.43(5)	1.36(5)	1.60(5)	$_{17}$Cl	1.04(5)	3.66(4)	2.27(3)
$_{44}$Ru	3.55(4)	4.54(4)	8.22(4)	$_{35}$Br	-3.91(6)	-1.29(6)	4.43(3)
$_{76}$Os	2.13(4)	2.40(4)	2.95(4)	$_{53}$I	6.56(7)	1.53(7)	1.17(4)
9				$_{85}$At	-4.31(7)	-1.18(7)	1.85(4)
$_{27}$Co	6.50(4)	7.96(4)	1.27(5)	18			
$_{45}$Rh	2.15(4)	3.46(4)	5.56(4)	$_{2}$He	1.30(1)	1.31(1)	1.33(1)
$_{77}$Ir	2.61(4)	1.71(4)	1.62(4)	$_{10}$Ne	8.02(1)	8.12(1)	8.31(1)
10				$_{18}$Ar	8.48(2)	8.79(2)	9.25(2)
$_{28}$Ni	1.34(5)	9.65(4)	1.01(5)	$_{36}$Kr	1.80(3)	1.88(3)	2.01(3)
$_{46}$Pd	1.69(4)	-1.26(5)	-1.65(5)	$_{54}$Xe	4.75(3)	5.06(3)	5.56(3)
$_{78}$Pt	1.09(4)	1.24(4)	1.54(4)	$_{86}$Rn	7.52(3)	8.12(3)	9.08(3)

Table S8 Isotropic average of scalar component for static $\gamma_{||}(0;0,0,0)$, dc-Kerr $\gamma_{||}(-\omega;\omega,0,0)$, and electric field-induced second-harmonic generation $\gamma_{||}(-2\omega;\omega,\omega,0)$ second-order hyperpolarizabilities for the elements of Periodic Table H-Rn (except lanthanides) calculated at the B3LYP/def2-QZVPPD level ($\gamma_{||}$/a.u.), in the wavelength λ = 1064.0 nm. A(n) means A×10^n.

	$\gamma_{||}(0;0,0,0)$	$\gamma_{||}(-\omega;\omega,0,0)$	$\gamma_{||}(-2\omega;\omega,\omega,0)$		$\gamma_{||}(0;0,0,0)$	$\gamma_{||}(-\omega;\omega,0,0)$	$\gamma_{||}(-2\omega;\omega,\omega,0)$
1				11			
$_{1}$H	5.14(2)	5.52(2)	6.03(2)	$_{29}$Cu	5.41(4)	7.37(4)	1.17(5)
$_{3}$Li	-1.42(5)	-9.71(5)	2.60(6)	$_{47}$Ag	2.24(4)	3.33(4)	5.61(4)
$_{11}$Na	9.59(4)	3.62(5)	-1.07(7)	$_{79}$Au	9.56(3)	1.20(4)	1.52(4)
$_{19}$K	-1.09(6)	-2.26(6)	-2.14(6)	12			
$_{37}$Rb	5.54(6)	4.31(7)	4.72(7)	$_{30}$Zn	2.42(4)	2.94(4)	3.71(4)
$_{55}$Cs	1.05(7)	2.27(8)	1.88(8)	$_{48}$Cd	1.56(4)	1.95(4)	2.61(4)
2				$_{80}$Hg	1.16(4)	1.34(4)	1.59(4)
$_{4}$Be	1.75(4)	2.03(4)	2.62(4)	13			
$_{12}$Mg	9.23(4)	1.23(5)	1.89(5)	$_{5}$B	-1.30(5)	-3.69(4)	2.12(4)
$_{20}$Ca	3.63(5)	6.41(5)	1.41(6)	$_{13}$Al	-1.67(7)	-7.27(6)	5.01(5)
$_{38}$Sr	8.04(5)	1.49(6)	8.86(6)	$_{31}$Ga	-2.02(7)	-8.62(6)	2.19(5)
$_{56}$Ba	7.41(5)	2.44(6)	1.99(7)	$_{49}$In	-2.88(7)	-1.29(7)	3.65(6)
3				$_{81}$Tl	-1.69(7)	-7.28(6)	-2.03(7)
$_{21}$Sc	-4.11(6)	-1.54(6)	9.17(5)	14			
$_{39}$Y	-1.04(5)	8.08(5)	3.63(5)	$_{6}$C	-3.62(4)	-1.09(4)	3.22(3)
$_{57}$La	-1.52(6)	-1.13(5)	3.42(6)	$_{14}$Si	1.26(7)	4.91(6)	3.88(4)
4				$_{32}$Ge	-3.41(8)	-6.46(7)	3.04(4)
$_{22}$Ti	9.54(4)	2.53(5)	5.36(5)	$_{50}$Sn	-7.17(8)	-1.21(8)	1.82(5)
$_{40}$Zr	-9.15(4)	2.81(5)	1.90(5)	$_{82}$Pb	-6.10(7)	-2.32(7)	2.59(5)
$_{72}$Hf	6.07(4)	5.99(4)	2.00(5)	15			
5				$_{7}$N	6.42(2)	6.73(2)	7.11(2)
$_{23}$V	1.65(5)	2.09(5)	4.62(5)	$_{15}$P	5.95(3)	6.72(3)	7.66(3)
$_{41}$Nb	3.81(7)	2.20(7)	1.11(5)	$_{33}$As	9.19(3)	1.06(4)	1.24(4)
$_{73}$Ta	5.77(4)	7.36(4)	2.06(5)	$_{51}$Sb	2.76(4)	3.28(4)	4.11(4)
6				$_{83}$Bi	3.51(4)	4.31(4)	5.60(4)
$_{24}$Cr	9.60(4)	1.47(5)	3.16(5)	16			
$_{42}$Mo	5.66(4)	8.34(4)	1.61(5)	$_{8}$O	-1.79(3)	-2.17(2)	6.21(2)
$_{74}$W	7.11(3)	9.67(3)	1.20(4)	$_{16}$S	-6.26(5)	-2.10(5)	7.16(3)
7				$_{34}$Se	-7.97(5)	-2.67(5)	1.24(4)
$_{25}$Mn	1.03(5)	-1.58(5)	-3.38(5)	$_{52}$Te	-2.03(6)	-6.93(5)	3.30(4)
$_{43}$Tc	-2.69(8)	-8.27(7)	1.45(5)	$_{84}$Po	-8.86(5)	-2.86(5)	5.09(4)
$_{75}$Re	3.58(4)	5.67(4)	7.15(4)	17			
8				$_{9}$F	-8.40(2)	-1.42(2)	2.20(2)
$_{26}$Fe	-8.20(4)	-9.43(2)	5.17(5)	$_{17}$Cl	1.67(5)	5.82(4)	2.57(3)
$_{44}$Ru	3.93(4)	5.40(4)	9.39(4)	$_{35}$Br	-1.99(6)	-6.67(5)	5.09(3)
$_{76}$Os	-2.67(5)	-8.00(4)	4.12(4)	$_{53}$I	-2.80(7)	1.19(7)	1.39(4)
9				$_{85}$At	-5.90(6)	-2.00(6)	2.19(4)
$_{27}$Co	7.73(4)	1.02(5)	1.58(5)	18			
$_{45}$Rh	1.85(4)	3.42(4)	6.42(4)	$_{2}$He	1.31(1)	1.33(1)	1.35(1)
$_{77}$Ir	3.15(5)	1.24(5)	2.83(4)	$_{10}$Ne	8.31(1)	8.51(1)	8.71(1)
10				$_{18}$Ar	9.17(2)	9.67(2)	1.02(3)
$_{28}$Ni	6.42(5)	3.05(5)	1.11(5)	$_{36}$Kr	1.97(3)	2.10(3)	2.25(3)
$_{46}$Pd	2.14(4)	-5.12(4)	-7.74(4)	$_{54}$Xe	5.29(3)	5.77(3)	6.38(3)
$_{78}$Pt	1.88(4)	1.70(4)	1.76(4)	$_{86}$Rn	8.33(3)	9.23(3)	1.04(4)

Table S9 Isotropic average of scalar component for static $\gamma_{||}(0;0,0,0)$, dc-Kerr $\gamma_{||}(-\omega;\omega,0,0)$, and electric field-induced second-harmonic generation $\gamma_{||}(-2\omega;\omega,\omega,0)$ second-order hyperpolarizabilities for the lanthanides calculated at the B3LYP/def2-QZVPP and CAM-B3LYP/def2-QZVPP levels ($\gamma_{||}$/a.u.), in the wavelength λ = 1064.0 nm. A(n) means A×10^n.

	$\gamma_{||}(0;0,0,0)$	$\gamma_{||}(-\omega;\omega,0,0)$	$\gamma_{||}(-2\omega;\omega,\omega,0)$	$\gamma_{||}(0;0,0,0)$	$\gamma_{||}(-\omega;\omega,0,0)$	$\gamma_{||}(-2\omega;\omega,\omega,0)$
		B3LYP			CAM-B3LYP	
$_{57}$La	-1.52(6)	-1.13(5)	3.42(6)	-3.33(6)	-1.32(6)	3.43(6)
$_{58}$Ce	-5.84(4)	1.91(4)	2.37(5)	-7.94(4)	-9.92(4)	-3.37(5)
$_{59}$Pr	-1.56(5)	-1.89(4)	1.14(5)	-2.37(5)	-9.60(4)	-7.91(4)
$_{60}$Nd	-6.21(4)	-2.64(4)	1.02(5)	-9.26(4)	-1.87(5)	-1.93(5)
$_{61}$Pm	-3.28(4)	-2.06(5)	-5.94(5)	-4.67(4)	-1.14(4)	-5.48(4)
$_{62}$Sm	5.41(4)	1.05(5)	8.46(7)	2.99(3)	5.04(4)	3.33(6)
$_{63}$Eu	2.27(4)	9.57(4)	8.38(5)	1.74(8)	2.24(5)	-7.48(6)
$_{64}$Gd [a]	-1.87(7)	1.48(6)	-2.29(5)	-2.43(5)	-9.15(4)	-8.85(5)
$_{65}$Tb [a]	-1.63(4)	-1.69(3)	-3.87(4)	-2.00(6)	-3.17(4)	-3.50(4)
$_{66}$Dy [a]	-5.30(3)	-5.20(3)	-5.44(3)	-8.86(3)	-7.21(3)	-8.05(3)
$_{67}$Ho	7.45(4)	1.40(5)	5.07(5)	5.67(4)	1.05(5)	4.48(5)
$_{68}$Er	1.07(5)	1.80(5)	2.65(5)	8.63(4)	1.41(5)	4.90(5)
$_{69}$Tm	-5.79(4)	1.35(5)	6.77(5)	1.17(5)	1.79(5)	5.47(5)
$_{70}$Yb	8.15(4)	1.59(5)	4.45(5)	6.80(4)	1.24(5)	3.62(5)
$_{71}$Lu	-6.15(7)	2.61(7)	-3.64(4)	-2.00(7)	5.34(6)	3.82(5)

[a] Response property for the Gd, Tb, and Dy atoms calculated at the all-electron relativistic DKH2-B3LYP/ANO-RCC-MB and DKH2- CAM-B3LYP /ANO-RCC-MB level.

Table S10 Mean static polarizabilities ($\bar{\alpha}$/a.u.) together with the corresponding components and polarizability anisotropies (α_2/a.u.) for the lanthanides calculated at the B3LYP/ def2-QZVPP level of theory.

	Configuration	α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$	α_2
$_{57}$La	[Xe]5d$^{1.3}$6s$^{1.7}$	187.03	187.03	198.75	190.94	11.720
$_{58}$Ce	[Xe]4f^{2}6s^2	210.10	210.10	208.27	209.49	-1.8289
$_{59}$Pr	[Xe]4f^{3}6s^2	202.17	202.17	202.17	202.17	0.0000
$_{60}$Nd	[Xe]4f^{4}6s^2	193.39	193.39	193.39	193.39	0.0000
$_{61}$Pm	[Xe]4f^{5}6s^2	185.59	185.59	188.89	186.69	3.2980
$_{62}$Sm	[Xe]4f^{6}6s^2	179.07	179.07	182.05	180.06	2.9798
$_{63}$Eu	[Xe]4f^{7}6s^2	175.51	175.51	175.51	175.51	0.0000
$_{64}$Gd [a]	[Xe]4f^{7}5d^{1}6s^2	142.18	142.18	149.75	144.70	7.5692
$_{65}$Tb [a]	[Xe]4f^{9}6s^2	162.50	162.51	163.69	162.90	1.1829
$_{66}$Dy [a]	[Xe]4f^{10}6s^2	156.36	156.36	156.36	156.36	0.0003
$_{67}$Ho	[Xe]4f^{11}6s^2	152.60	152.60	152.60	152.60	0.0000
$_{68}$Er	[Xe]4f^{12}6s^2	149.00	149.00	148.33	148.78	-0.6680
$_{69}$Tm	[Xe]4f^{13}6s^2	145.57	145.57	142.44	144.53	-3.1322
$_{70}$Yb	[Xe]4f^{14}6s^2	143.60	143.60	143.60	143.60	0.0000
$_{71}$Lu	[Xe]4f^{14}5d^{1}6s^2	130.47	130.47	106.77	122.57	-23.699

[a] Response property for the Gd, Tb, and Dy atoms calculated at the all-electron relativistic DKH2-B3LYP/SARC-DKH-TZVPP level.

Table S11 Mean static polarizabilities ($\bar{\alpha}$/a.u.) and the corresponding components together with the polarizability anisotropies (α_2/a.u.) for the actinides computed at the all-electron relativistic B3LYP/SARC-DKH-TZVPP level.

	Configuration	α_{xx}	α_{yy}	α_{zz}	$\bar{\alpha}$	α_2
$_{89}$Ac	[Rn]6d^{1}7s^2	170.02	170.02	160.69	166.91	-9.3252
$_{90}$Th	[Rn]5f^{1}6d^{1}7s^2	148.63	151.89	153.14	151.22	4.0336
$_{91}$Pa	[Rn]5f^{2}6d^{1}7s^2	142.29	143.12	145.09	143.50	2.4974
$_{92}$U	[Rn]5f^{4}7s^2	144.13	144.13	144.13	144.13	0.0005
$_{93}$Np	[Rn]5f^{5}7s^2	136.46	136.47	137.87	136.93	1.4012
$_{94}$Pu	[Rn]5f^{6}7s^2	131.12	131.12	131.13	131.12	0.0004
$_{95}$Am	[Rn]5f^{7}7s^2	125.82	125.82	125.82	125.82	0.0000
$_{96}$Cm	[Rn]5f^{7}6d^{1}7s^2	111.27	111.28	122.37	114.97	11.093
$_{97}$Bk	[Rn]5f^{9}7s^2	114.31	114.30	112.92	113.84	-1.3837
$_{98}$Cf	[Rn]5f^{10}7s^2	108.90	108.90	108.90	108.90	0.0009
$_{99}$Es	[Rn]5f^{11}7s^2	104.28	104.28	104.28	104.28	0.0001
$_{100}$Fm	[Rn]5f^{12}7s^2	98.487	98.487	102.28	99.751	3.7922
$_{101}$Md	[Rn]5f^{13}7s^2	95.782	95.782	95.782	95.782	0.0000
$_{102}$No	[Rn]5f^{14}7s^2	91.934	91.934	91.934	91.934	0.0000
$_{103}$Lr	[Rn]5f^{14}6d^{1}7s^2	135.92	135.91	75.270	115.70	-60.649

The ground configuration of Lr atom calculated with the fully variational spin-orbit coupled CASSCF approach (so that the complete active space is the five d-orbitals along with the three p-orbitals, thus resulting in 1-electron in 8 orbitals) is also [Rn]5f^{14}6d^{1}7s^2.

Table S12 Molar volumes (V_m/cm^3.mol^{-1}), mean radii (R_s/A°), semimajor axis (R_a/A°), semiminor axis (R_b/A°) and eccentricity ($\epsilon = \sqrt{1-(\frac{R_b}{R_a})^2}$) for the lanthanides. Molar volumes computed using B3LYP/ def2-QZVP method.

	V_m	R_s	R_a	R_b	ϵ
$_{57}$La	52.436	2.7496	2.7824	2.7168	0.2159
$_{58}$Ce	64.963	2.9531	2.9620	2.9442	0.1095
$_{59}$Pr	60.680	2.8867	2.8867	2.8867	0.0000
$_{60}$Nd	52.886	2.7574	2.7574	2.7574	0.0000
$_{61}$Pm	50.164	2.7093	2.7237	2.6949	0.1450
$_{62}$Sm	48.854	2.6855	2.6991	2.6719	0.1416
$_{63}$Eu	46.666	2.6448	2.6448	2.6448	0.0000
$_{64}$Gd [a]	44.169	2.5967	2.6225	2.5709	0.1974
$_{65}$Tb [a]	59.688	2.8709	2.8716	2.8702	0.0312
$_{66}$Dy [a]	51.066	2.7254	2.7254	2.7254	0.0000
$_{67}$Ho	47.799	2.6660	2.6660	2.6660	0.0000
$_{68}$Er	46.542	2.6424	2.6475	2.6373	0.0877
$_{69}$Tm	44.668	2.6065	2.6212	2.5918	0.1494
$_{70}$Yb	44.030	2.5940	2.5940	2.5940	0.0000
$_{71}$Lu	44.030	2.5940	2.6501	2.5379	0.2879

Table S13 Molar volumes (V_m/cm^3.mol^{-1}), mean radii (R_s/A°), semimajor axis (R_a/A°), semiminor axis (R_b/A°) and eccentricity ($\epsilon = \sqrt{1-(\frac{R_b}{R_a})^2}$) for the elements H-Rn (except lanthanides) computed at the B3LYP/def2-QZVPPD level.

	V_m	R_S	R_a	R_b	ϵ		V_m	R_S	R_a	R_b	ϵ
1						11					
$_{1}$H	10.113	1.5886	1.5886	1.5886	0.0000	$_{29}$Cu	28.004	2.2308	2.2308	2.2308	0.0000
$_{3}$Li	28.376	2.2406	2.2406	2.2406	0.0000	$_{47}$Ag	34.844	2.3994	2.3994	2.3994	0.0000
$_{11}$Na	34.049	2.3810	2.3810	2.3810	0.0000	$_{79}$Au	27.310	2.2122	2.2122	2.2122	0.0000
$_{19}$K	39.981	2.5119	2.5119	2.5119	0.0000	12					
$_{37}$Rb	47.722	2.6646	2.6646	2.6646	0.0000	$_{30}$Zn	26.223	2.1825	2.1825	2.1825	0.0000
$_{55}$Cs	55.031	2.7942	2.7942	2.7942	0.0000	$_{48}$Cd	32.168	2.3363	2.3363	2.3363	0.0000
2						$_{80}$Hg	25.621	2.1656	2.1656	2.1656	0.0000
$_{4}$Be	25.964	2.1753	2.1753	2.1753	0.0000	13					
$_{12}$Mg	31.359	2.3165	2.3165	2.3165	0.0000	$_{5}$B	18.869	1.9557	1.9874	1.9240	0.2506
$_{20}$Ca	39.695	2.5059	2.5059	2.5059	0.0000	$_{13}$Al	29.194	2.2620	2.3420	2.1820	0.3633
$_{38}$Sr	46.869	2.6486	2.6486	2.6486	0.0000	$_{31}$Ga	30.813	2.3030	2.3919	2.2141	0.3783
$_{56}$Ba	53.466	2.7674	2.7674	2.7674	0.0000	$_{49}$In	36.968	2.4472	2.5469	2.3475	0.3879
3						$_{81}$Tl	39.061	2.4925	2.6107	2.3743	0.4158
$_{21}$Sc	38.055	2.4709	2.4983	2.4435	0.2083	14					
$_{39}$Y	45.414	2.6209	2.6327	2.6091	0.1336	$_{6}$C	16.965	1.8876	1.9043	1.8709	0.1865
$_{57}$La	51.687	2.7364	2.7694	2.7034	0.2170	$_{14}$Si	26.110	2.1793	2.2177	2.1409	0.2609
4						$_{32}$Ge	27.317	2.2124	2.2570	2.1678	0.2784
$_{22}$Ti	36.996	2.4478	2.4709	2.4247	0.1925	$_{50}$Sn	34.422	2.3896	2.4415	2.3377	0.2885
$_{40}$Zr	44.105	2.5955	2.6205	2.5705	0.1944	$_{82}$Pb	37.410	2.4569	2.5206	2.3932	0.3139
$_{72}$Hf	43.492	2.5834	2.5922	2.5746	0.1163	15					
5						$_{7}$N	14.175	1.7778	1.7778	1.7778	0.0000
$_{23}$V	36.293	2.4322	2.4322	2.4322	0.0000	$_{15}$P	24.311	2.1281	2.1281	2.1281	0.0000
$_{41}$Nb	42.848	2.5706	2.5940	2.5472	0.1891	$_{33}$As	26.440	2.1885	2.1885	2.1885	0.0000
$_{73}$Ta	41.907	2.5516	2.5516	2.5516	0.0000	$_{51}$Sb	33.929	2.3782	2.3782	2.3782	0.0000
6						$_{83}$Bi	35.779	2.4206	2.4206	2.4206	0.0000
$_{24}$Cr	35.065	2.4044	2.4044	2.4044	0.0000	16					
$_{42}$Mo	42.121	2.5560	2.5560	2.5560	0.0000	$_{8}$O	11.908	1.6775	1.6887	1.6663	0.1623
$_{74}$W	40.347	2.5196	2.5196	2.5196	0.0000	$_{16}$S	22.298	2.0676	2.0900	2.0452	0.2059
7						$_{34}$Se	24.346	2.1291	2.1560	2.1022	0.2220
$_{25}$Mn	34.826	2.3990	2.3990	2.3990	0.0000	$_{52}$Te	33.464	2.3673	2.3978	2.3368	0.2241
$_{43}$Tc	40.618	2.5252	2.5562	2.4942	0.2189	$_{84}$Po	34.660	2.3951	2.4324	2.3578	0.2458
$_{75}$Re	38.671	2.4842	2.4842	2.4842	0.0000	17					
8						$_{9}$F	9.627	1.5627	1.5700	1.5554	0.1361
$_{26}$Fe	31.594	2.3223	2.3317	2.3129	0.1267	$_{17}$Cl	20.821	2.0209	2.0351	2.0067	0.1665
$_{44}$Ru	38.924	2.4896	2.4899	2.4893	0.0220	$_{35}$Br	24.197	2.1247	2.1421	2.1073	0.1795
$_{76}$Os	37.475	2.4583	2.4674	2.4492	0.1212	$_{53}$I	31.073	2.3095	2.3304	2.2886	0.1886
9						$_{85}$At	33.630	2.3712	2.3970	2.3454	0.2064
$_{27}$Co	29.639	2.2734	2.2734	2.2734	0.0000	18					
$_{45}$Rh	37.285	2.4541	2.4541	2.4541	0.0000	$_{2}$He	6.924	1.4001	1.4001	1.4001	0.0000
$_{77}$Ir	36.810	2.4437	2.4437	2.4437	0.0000	$_{10}$Ne	8.788	1.5159	1.5159	1.5159	0.0000
10						$_{18}$Ar	18.706	1.9501	1.9501	1.9501	0.0000
$_{28}$Ni	28.084	2.2329	2.2433	2.2225	0.1359	$_{36}$Kr	20.386	2.0068	2.0068	2.0068	0.0000
$_{46}$Pd	24.492	2.1333	2.1333	2.1333	0.0000	$_{54}$Xe	28.820	2.2523	2.2523	2.2523	0.0000
$_{78}$Pt	32.810	2.3517	2.3547	2.3487	0.0713	$_{86}$Rn	31.976	2.3316	2.3316	2.3316	0.0000

Table S14 All-electron relativistic polarizability components (α/a.u.) and the tensor polarizabilities (α_2/a.u.) for the Au atom computed using different functionals and DZP-DKH basis set.

	Configuration	α_{xx}	α_{yy}	α_{zz}	α_2
B3LYP	$[Xe]4f^{14}5d^{9.26}6s^{1.74}$	32.178	32.178	33.826	1.648
CAM-B3LYP	$[Xe]4f^{14}5d^{9.27}6s^{1.73}$	33.075	33.075	34.851	1.777
TPSSh	$[Xe]4f^{14}5d^{9.47}6s^{1.53}$	31.069	31.069	32.985	1.916
TPSS	$[Xe]4f^{14}5d^{9.42}6s^{1.58}$	30.674	30.675	32.532	1.858
BHandHLYP	$[Xe]4f^{14}5d^{9.21}6s^{1.79}$	34.249	34.249	36.155	1.906
BP86	$[Xe]4f^{14}5d^{9.35}6s^{1.65}$	30.763	30.763	32.260	1.497
B3PW	$[Xe]4f^{14}5d^{9.48}6s^{1.52}$	31.147	31.147	32.888	1.741
PBE	$[Xe]4f^{14}5d^{9.36}6s^{1.64}$	31.018	31.018	32.694	1.676
PW91	$[Xe]4f^{14}5d^{9.37}6s^{1.63}$	30.741	30.741	32.377	1.636
BLYP	$[Xe]4f^{14}5d^{9.24}6s^{1.76}$	31.492	31.492	32.881	1.389
LC-BLYP	$[Xe]4f^{14}5d^{9.18}6s^{1.82}$	31.832	31.832	33.235	1.403
B1LYP	$[Xe]4f^{14}5d^{9.23}6s^{1.77}$	32.794	32.794	34.451	1.657
X3LYP	$[Xe]4f^{14}5d^{9.24}6s^{1.76}$	32.493	32.493	34.139	1.646
M06L	$[Xe]4f^{14}5d^{9.45}6s^{1.55}$	38.944	38.951	41.308	2.361
mPW1PW	$[Xe]4f^{14}5d^{9.54}6s^{1.46}$	31.526	31.526	33.205	1.679
wB97X	$[Xe]4f^{14}5d^{9.27}6s^{1.73}$	35.658	35.658	36.902	1.244

The B3LYP hybrid functional is the Gaussian version as implemented in the ORCA quantum chemistry package (B3LYP/G).[121]

Table S15 Spin multiplicities (2S+1), relativistic anisotropies ($\alpha_{2,R}$/a.u.) calculated at the all-electron relativistic DKH2-B3LYP/DZP-DKH level, non-relativistic anisotropies ($\alpha_{2,nR}$/a.u.) calculated at the all-electron non-relativistic B3LYP/DZP level, and relativistic effect ($\alpha_{2,R} - \alpha_{2,nR}$) on elements Hf-Rn.

	2S + 1	$\alpha_{2,R}$	$\alpha_{2,nR}$	$\alpha_{2,R} - \alpha_{2,nR}$
$_{72}$Hf	3	1.9068	-5.0424	6.9492
$_{73}$Ta	4	0.0015	39.942	-39.941
$_{73}$Ta	6	-36.762	54.707	-91.469
$_{74}$W	5	2.6899	-4.4125	7.1024
$_{74}$W	7	0.0000	0.0000	0.0000
$_{75}$Re	6	0.0000	0.0000	0.0000
$_{76}$Os	5	1.5047	4.2787	-2.7740
$_{77}$Ir	4	0.0000	3.3609	-3.3609
$_{78}$Pt	3	-0.0003	0.0000	-0.0003
$_{79}$Au	2	1.6479	0.0000	1.6479
$_{80}$Hg	1	0.0000	0.0000	0.0000
$_{81}$Tl	2	25.306	7.2583	18.048
$_{82}$Pb	3	-8.7362	-1.2207	-7.5155
$_{83}$Bi	4	0.0000	0.0000	0.0000
$_{84}$Po	3	3.4095	-3.3536	6.7631
$_{85}$At	2	-0.9557	3.3661	-4.3218
$_{86}$Rn	1	0.0000	0.0000	0.0000

All-electron relativistic anisotropies for Hf and Ta calculated using ANO-RCC-DZP basis set. Nonrelativistic Ir is triaxial ellipsoidal atom.

Table S16 Spin multiplicities (2S+1), relativistic anisotropies ($\alpha_{2,R}$/a.u.) calculated at the all-electron relativistic B3LYP/SARC-DKH-TZVPP level, non-relativistic anisotropies ($\alpha_{2,nR}$/a.u.) calculated at the all-electron non-relativistic B3LYP/DZP level, and relativistic effect ($\alpha_{2,R} - \alpha_{2,nR}$) on lanthanides and actinides.

	2S + 1	$\alpha_{2,R}$	$\alpha_{2,nR}$	$\alpha_{2,R} - \alpha_{2,nR}$
Lanthanoids				
$_{57}$La	2	11.174	6.6673	4.5067
$_{58}$Ce	3	-1.4340	-2.3820	0.9480
$_{59}$Pr[a]	4	0.0000	0.0000	0.0000
$_{60}$Nd	5	-7.0741	0.0000	-7.0741
$_{61}$Pm	6	4.3758	0.0009	4.3749
$_{62}$Sm	7	-0.0003	0.0000	-0.0003
$_{63}$Eu	8	0.0001	0.0000	0.0001
$_{64}$Gd	9	7.5692	0.0503	7.5189
$_{65}$Tb	6	1.1829	0.0185	1.1644
$_{66}$Dy	5	0.0003	0.0000	0.0003
$_{67}$Ho	4	0.8104	0.0000	0.8104
$_{68}$Er	3	3.2122	-0.0370	3.2492
$_{69}$Tm[a]	2	0.0000	0.0000	0.0000
$_{70}$Yb	1	0.0000	0.0000	0.0000
$_{71}$Lu	2	36.381	-0.2227	36.604
Actinoids				
$_{89}$Ac	2	-9.3252	0.3896	-9.7148
$_{90}$Th	3	4.0336	0.9234	3.1102
$_{91}$Pa	4	2.4974	0.0001	2.4973
$_{92}$U[a]	5	1.8116	0.0000	1.8116
$_{93}$Np	6	1.4012	0.0598	1.3414
$_{94}$Pu	7	0.0004	0.0002	0.0002
$_{95}$Am	8	0.0000	0.0000	0.0000
$_{96}$Cm	9	11.093	0.0000	11.093
$_{97}$Bk	6	-1.3837	0.0329	-1.4166
$_{98}$Cf	5	0.0009	-0.0415	0.0424
$_{99}$Es	4	0.0001	-0.0001	0.0002
$_{100}$Fm	3	3.7922	-0.3324	4.1246
$_{101}$Md	2	0.0000	0.0000	0.0000
$_{102}$No	1	0.0000	0.0000	0.0000
$_{103}$Lr	2	-60.649	0.6022	-61.251

[a] Relativistic anisotropies for $_{92}$U, $_{69}$Tm, $_{59}$Pr, and Yb performed using DKH2-B3LYP/DZP-DKH method. Non-relativistic anisotropies for $_{58}$Ce and $_{101}$Md calculated using Gaussian09 package. Non-relativistic $_{93}$Np and $_{94}$Pu are triaxial ellipsoidal atoms. The available theoretical computations of the relativistic effects on the static dipole polarizabilities for the alkaline-earth elements 18,160, the groups 11^{41} and 12^{18} atoms have shown that the dipole polarizability is overestimated when the non-relativistic wavefunctions are used. As can be seen in Tables S15 and S16, the relativistic effects may decrease or increase the polarizability anisotropy of the elements of the Periodic Table, depending on how the relativity influences the energy of the orbitals and also the electron charge distributions around the nucleus of the isolated atoms. Especially, the contraction and stabilization of the outer shells has significant contributions on the electron charge distributions. Furthermore, the orbital-orbital charge transfer as a result of the change in the energy of the relativistic orbitals may predict different ground-configurations, spin-multiplicities and electric anisotropies for the isolated atoms.

Table S17 Spin multiplicities (2S+1), relativistic anisotropies ($\alpha_{2,R}$/a.u.) calculated at the all-electron relativistic DKH2-B3LYP/DZP-DKH level, non-relativistic anisotropies ($\alpha_{2,nR}$/a.u.) calculated at the all-electron non-relativistic B3LYP/DZP level, and relativistic effect ($\alpha_{2,R} - \alpha_{2,nR}$) on inert-gas atoms.

	2S + 1	$\alpha_{2,R}$	$\alpha_{2,nR}$	$\alpha_{2,R} - \alpha_{2,nR}$
$_{2}$He	1	0.0000	0.0000	0.0000
$_{10}$Ne	1	0.0000	0.0000	0.0000
$_{18}$Ar	1	0.0000	0.0000	0.0000
$_{36}$Kr	1	0.0000	0.0000	0.0000
$_{54}$Xe	1	0.0000	0.0000	0.0000
$_{86}$Rn	1	0.0000	0.0000	0.0000

It has been found that the relativistic effects do not play any significant role on the dipole polarizabilities of inert-gas atoms Kr, Xe and Rn.[18] The present computations (Table 17) reveals that the relativistic effects do not alter the sphericity of the closed-shell inert-gas atoms. Furthermore, the all-electron relativistic calculations including 4th-order Douglas-Kroll-Hess Hamiltonian and the spin-orbit terms at the DKHSO-GHF/DZP-DKH level is also performed for the inert-gas atoms (using Gaussian 09 package); and it is found that the inclusion of the spin-orbit coupling (SOC) do not alter the sphericity of the atoms He, Ne, Ar, Kr, and Xe ($\alpha_{2,R,SO} = 0$); however, a very small polarizability anisotropy value of 3.88×10^{-5} a.u. is predicted for the heavy element Rn at this level of theory.

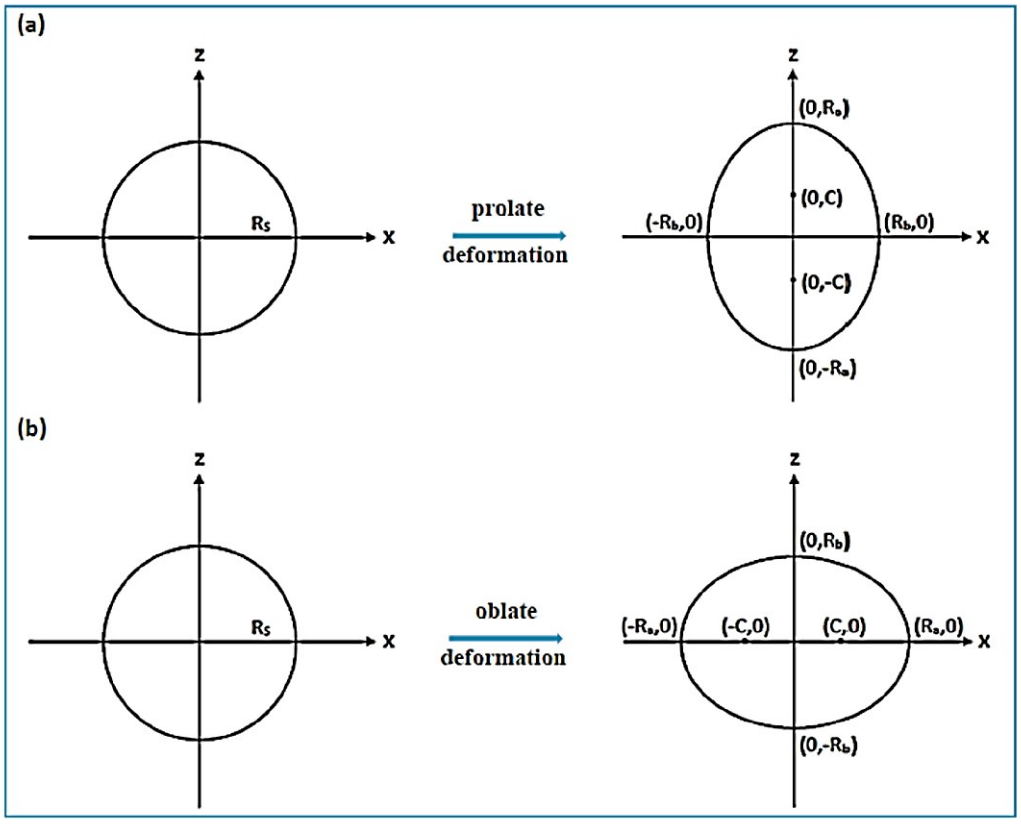

Fig. S1 Intersections of sphere and ellipsoids of rotation with xz plane: deformation of a hypothetical circle with radius R_S to (a) vertical and (b) horizontal ellipses with semimajor axis R_a and semiminor axis R_b. ($R_a > R_b$)