

# QTAIM-Based Scheme for Describing the Linear and Nonlinear Optical Susceptibilities of Molecular Crystals Composed of Molecules with Complex Shapes - Supplementary Information

Tomasz Seidler<sup>1,2,a),\*</sup>, Anna Krawczuk<sup>2</sup>, Benoît Champagne<sup>1,a),\*</sup>, and  
Katarzyna Stadnicka<sup>2</sup>

<sup>1</sup>Laboratoire de Chimie Théorique, Unité de Chimie Physique  
Théorique et Structurale, University of Namur, rue de Bruxelles, 61,  
B-5000, Namur, Belgium

<sup>2</sup>Faculty of Chemistry, Jagiellonian University, ul, Ingardena 3, 30-060,  
Kraków, Poland

---

a) Authors to whom correspondence should be addressed. Electronic addresses: [tomasz.seidler@unamur.be](mailto:tomasz.seidler@unamur.be) and [benoit.champagne@unamur.be](mailto:benoit.champagne@unamur.be).

## S.1 MNA - additional table

Tab. S1: QTAIM-atomic/group static ( $\lambda = \infty$ ) polarizability tensors (in a.u.) obtained at the MP2/6-311++G(d,p) level with polarizing electric field (experimental neutron diffraction coordinates) for MNA; The “FF” column gathers the finite field (FF) results performed on the whole molecule; for groups with hydrogen atoms the heavy atom contribution is given in parentheses.

group	C1	C2	C3H	C4	C5H	C6H	C7H <sub>3</sub>	N1H <sub>2</sub>	N2	O1	O2	SUM	FF
$\alpha_{xx}$	11.6	13.5	16.2(15.6)	17.3	12.7(10.8)	16.2(15.3)	13.6(7.1)	20.3(18.3)	21.0	11.5	4.6	158.6	158.6
$\alpha_{yy}$	11.5	12.6	9.3(5.8)	11.9	13.4(10.8)	12.2(7.6)	12.5(6.4)	9.6(7.1)	10.2	5.1	12.7	121.0	121.0
$\alpha_{zz}$	5.2	6.3	8.2(7.5)	7.6	7.7(6.7)	8.7(7.8)	12.0(5.2)	10.5(9.2)	6.8	4.7	4.1	81.8	81.7
$\alpha_{xy}$	-0.4	-1.9	0.6(0.8)	-2.3	-2.8(-4.4)	-0.2(0.3)	3.7(2.9)	-6.3(-6.1)	-4.8	4.6	-4.2	-14.1	-14.1
$\alpha_{xz}$	3.7	4.5	4.7(4.7)	5.9	3.3(2.8)	4.5(4.5)	0.9(1.1)	5.6(4.7)	8.7	3.4	0.8	46.0	45.9
$\alpha_{yz}$	-0.5	-1.2	0.2(0.4)	-1.3	-1.5(-2.2)	-0.4(0.0)	1.8(1.3)	-2.8(-2.3)	-2.7	2.0	-2.6	-9.0	-8.9
$\alpha_{iso}$	9.5	10.8	11.3(9.6)	12.2	11.3(9.4)	12.4(10.3)	12.7(6.2)	13.5(11.5)	12.7	7.1	7.1	120.5	120.4

## S.2 Additional polarizability calculations

The oxygen atomic polarizability in different *molecular* environments was assessed on the basis of several molecules: methanol, nitromethane, acetic acid, acetate anion, and MNA. The geometries were optimized with B3LYP/6-311++G(d,p) whereas the polarizabilities were calculated with MP2/6-311++G(d,p) method. The numbering scheme is presented in Figure S1 and the results of the calculations in Tables S2-S4.

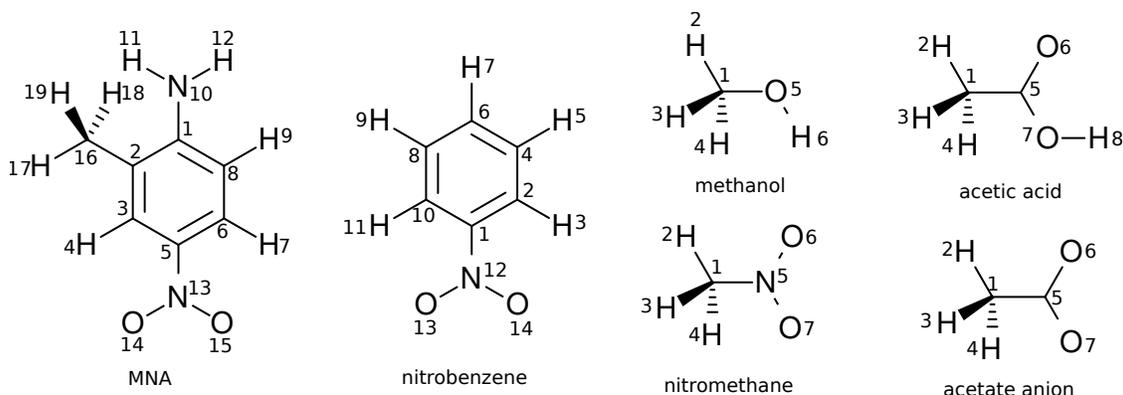


Fig. S1: Numbering scheme of the test molecules

Tab. S2: Eigenvalues of the atomic polarizability (in a.u.) tensors and isotropic polarizabilities for selected oxygen containing molecules - MNA and nitrobenzene.

MNA					nitrobenzene				
atom	$\alpha'_{xx}$	$\alpha'_{yy}$	$\alpha'_{zz}$	$\alpha_{iso}$	atom	$\alpha'_{xx}$	$\alpha'_{yy}$	$\alpha'_{zz}$	$\alpha_{iso}$
C(1)	3.48	10.90	11.63	8.67	C(1)	4.58	10.71	11.22	8.84
C(2)	4.05	11.53	14.53	10.04	C(2)	5.31	6.97	12.67	8.32
C(3)	5.15	6.18	15.67	9.00	H(3)	0.74	0.77	3.93	1.81
H(4)	0.71	0.79	3.73	1.74	C(4)	5.53	8.31	11.88	8.57
C(5)	4.65	10.99	14.86	10.17	H(5)	0.89	0.94	5.07	2.30
C(6)	5.29	7.01	14.28	8.86	C(6)	5.75	8.93	10.57	8.42
H(7)	0.70	0.79	4.12	1.87	H(7)	0.91	0.95	5.30	2.38
C(8)	5.59	7.85	15.22	9.55	C(8)	5.52	8.23	11.86	8.54
H(9)	0.91	0.93	5.10	2.31	H(9)	0.89	0.94	5.07	2.30
N(10)	4.80	7.43	19.81	10.68	C(10)	5.33	6.99	12.66	8.33
H(11)	0.30	0.46	2.58	1.11	H(11)	0.74	0.77	3.93	1.81
H(12)	0.30	0.47	2.84	1.20	N(12)	2.67	8.06	16.47	9.07
N(13)	2.67	8.22	20.53	10.47	O(13)	3.00	3.17	11.73	5.97
O(14)	2.96	3.27	12.71	6.31	O(14)	3.00	3.17	11.72	5.96
O(15)	2.91	3.24	13.10	6.42					
C(16)	3.98	4.83	10.32	6.38					
H(17)	1.00	1.03	4.34	2.12					
H(18)	0.90	1.13	4.84	2.29					
H(19)	0.94	1.17	4.99	2.37					

Tab. S3: Eigenvalues of the atomic polarizability (in a.u.) tensors and isotropic polarizabilities for selected oxygen containing molecules - nitromethane and methanol.

nitromethane					methanol				
atom	$\alpha'_{xx}$	$\alpha'_{yy}$	$\alpha'_{zz}$	$\alpha_{iso}$	atom	$\alpha'_{xx}$	$\alpha'_{yy}$	$\alpha'_{zz}$	$\alpha_{iso}$
C(1)	4.65	5.00	5.30	4.98	C(1)	4.12	4.94	5.03	4.70
H(2)	0.87	0.96	3.77	1.87	H(2)	0.85	1.00	4.29	2.04
H(3)	0.81	1.03	3.94	1.93	H(3)	0.90	1.04	4.66	2.20
H(4)	0.83	1.02	3.98	1.95	H(4)	0.90	1.04	4.66	2.20
N(5)	2.71	8.89	11.10	7.57	O(5)	4.36	4.62	10.45	6.48
O(6)	3.10	3.16	11.17	5.81	H(6)	0.24	0.28	1.82	0.78
O(7)	3.10	3.15	11.20	5.81					

Tab. S4: Eigenvalues of the atomic polarizability (in a.u.) tensors and isotropic polarizabilities for selected oxygen containing molecules - acetic acid and acetate anion.

acetic acid					acetate				
atom	$\alpha'_{xx}$	$\alpha'_{yy}$	$\alpha'_{zz}$	$\alpha_{iso}$	atom	$\alpha'_{xx}$	$\alpha'_{yy}$	$\alpha'_{zz}$	$\alpha_{iso}$
C(1)	5.00	5.12	7.94	6.02	C(1)	6.06	6.18	10.52	7.59
H(2)	0.95	1.01	4.18	2.05	H(2)	1.30	1.44	6.09	2.94
H(3)	0.94	1.09	4.41	2.15	H(3)	1.32	1.53	6.06	2.97
H(4)	0.94	1.10	4.41	2.15	H(4)	1.20	1.54	6.05	2.93
C(5)	3.00	4.71	7.51	5.07	C(5)	3.23	5.83	10.54	6.53
O(6)	3.66	3.82	11.83	6.44	O(6)	5.79	6.10	16.52	9.47
O(7)	4.38	4.50	11.30	6.72	O(7)	5.84	5.98	16.39	9.40
H(8)	0.20	0.25	1.83	0.76					

### S.3 MBANP - additional tables

Tab. S5: Neutron diffraction fractional coordinates and Mulliken populations (in e) of monoclinic MBANP as obtained from B3LYP/6-31G(d,p) calculations with periodic boundary conditions.

atom	x	y	z	Mulliken charge
N1	0.5712	0.7316	0.3026	-0.4573
C1	0.7922	0.8116	0.3324	0.4539
C2	0.9319	0.7158	0.3944	-0.1538
H2	1.1051	0.7868	0.4161	0.1374
C3	0.8457	0.5305	0.4240	-0.0470
H3	0.9485	0.4492	0.4702	0.1449
C4	0.6180	0.4461	0.3917	0.2142
C5	0.4876	0.5519	0.3326	0.0555
H5	0.3098	0.4870	0.3092	0.1285
N2	0.5221	0.2486	0.4197	0.4090
O1	0.6458	0.1528	0.4698	-0.4065
O2	0.3175	0.1837	0.3915	-0.4474
N3	0.8817	0.9896	0.3004	-0.5417
H3n	1.0504	1.0463	0.3223	0.3136
C6	0.7761	1.0788	0.2293	0.0031
H6	0.5722	1.1041	0.2343	0.1146
C7	0.9068	1.2913	0.2160	-0.3391
H7a	0.8215	1.3687	0.1664	0.1362
H7b	0.8927	1.3966	0.2642	0.1192
H7c	1.1034	1.2654	0.2071	0.1086
C8	0.8083	0.9335	0.1617	0.1339
C9	0.6266	0.9362	0.1012	-0.1214
H9	0.4594	1.0339	0.1057	0.0874
C10	0.6576	0.8155	0.0366	-0.1070
H10	0.5144	0.8191	-0.0089	0.0966
C11	0.8723	0.6900	0.0317	-0.1005
H11	0.8973	0.5943	-0.0180	0.1040
C12	1.0539	0.6858	0.0915	-0.1013
H12	1.2218	0.5898	0.0879	0.0944
C13	1.0243	0.8075	0.1567	-0.1497
H13	1.1690	0.8058	0.2031	0.1176

Tab. S6: Static polarizability and first hyperpolarizability of MBANP (in a.u.) for the cases with and without charge electric field. These are given in the crystal  $abc^*$  axes system for the first molecule of the unit cell.

	B3LYP/6-311++G(3df,3pd)		MP2/6-311++G(d,p)		MP2/6-311++G(2d,2p)	
	no field	charge field	no field	charge field	no field	charge field
$\alpha_{xx}$	163.9	164.3	163.7	164.4	166.6	167.3
$\alpha_{yy}$	204.2	212.2	198.5	207.0	203.8	212.1
$\alpha_{zz}$	204.0	206.7	200.2	203.1	205.3	208.0
$\alpha_{xy}$	21.0	23.9	19.3	22.3	20.1	23.1
$\alpha_{xz}$	4.4	1.8	6.7	4.2	5.5	3.1
$\alpha_{yz}$	-33.8	-39.9	-31.3	-37.6	-32.3	-38.3
$\alpha_{iso}$	190.7	194.4	187.5	191.5	191.9	195.8
$\beta_{xxx}$	-78.8	-29.4	-41.7	-12.2	-43.0	-11.0
$\beta_{yyy}$	1049.2	1354.2	1106.1	1636.3	1073.9	1574.8
$\beta_{zzz}$	-212.2	-162.4	-237.0	-270.6	-229.1	-259.7
$\beta_{xxy}$	112.4	161.3	141.1	200.9	136.9	195.8
$\beta_{xxz}$	-105.3	-108.2	-102.5	-129.2	-95.4	-119.9
$\beta_{xyy}$	505.5	636.1	495.6	701.4	479.4	676.4
$\beta_{xzz}$	217.3	278.6	196.8	294.3	191.4	283.9
$\beta_{yyz}$	-792.0	-968.3	-803.6	-1132.1	-783.0	-1096.1
$\beta_{yzz}$	414.0	533.3	435.4	650.3	432.3	637.1
$\beta_{xyz}$	-392.1	-469.4	-360.5	-496.9	-352.5	-481.7
$\beta_{tot}$	1219.0	1531.6	1281.4	1849.5	1247.2	1787.5

Tab. S7: Dynamic (at  $\lambda = 1064\text{nm}$ ) polarizability and first hyperpolarizability of MBANP (in a.u.) with and without charge electric field. These are given in the crystal  $abc^*$  axes system for the first molecule of the unit cell.

	B3LYP/6-311++G(3df,3pd)		MP2/6-311++G(d,p)		MP2/6-311++G(2d,2p)	
	no field	charge field	no field	charge field	no field	charge field
$\alpha_{xx}$	166.3	166.9	165.8	166.7	169.0	169.9
$\alpha_{yy}$	209.9	219.5	203.6	213.6	209.4	219.3
$\alpha_{zz}$	208.4	211.9	205.1	208.8	209.7	213.2
$\alpha_{xy}$	22.6	26.1	20.1	23.7	21.6	25.3
$\alpha_{xz}$	3.6	0.6	6.4	3.6	4.7	1.9
$\alpha_{yz}$	-36.6	-43.8	-34.1	-41.6	-35.0	-42.2
$\alpha_{iso}$	194.9	199.4	191.5	196.3	196.0	200.8
$\beta_{xxx}$	-54.1	24.7	-80.3	-26.1	-19.4	66.0
$\beta_{yyy}$	1969.8	2802.0	2130.9	3506.3	2021.3	3221.5
$\beta_{zzz}$	-481.2	-534.3	-456.5	-579.8	-498.4	-684.2
$\beta_{xxy}$	256.4	385.7	271.8	430.5	271.5	460.0
$\beta_{yxx}$	233.3	360.1	271.8	430.5	266.6	412.8
$\beta_{xxz}$	-211.2	-265.3	-197.5	-276.8	-183.0	-304.6
$\beta_{zxx}$	-193.4	-247.8	-197.5	-276.8	-173.5	-277.2
$\beta_{xyy}$	886.6	1202.5	954.8	1503.0	852.1	1368.6
$\beta_{yyx}$	914.1	1281.4	954.8	1503.0	880.5	1374.4
$\beta_{xzz}$	378.3	510.1	379.1	630.5	338.1	565.8
$\beta_{zxx}$	417.5	596.0	379.1	630.5	366.0	615.4
$\beta_{yyz}$	-1426.6	-1946.6	-1548.3	-2425.9	-1416.1	-2203.6
$\beta_{zyy}$	-1454.6	-2003.0	-1548.3	-2425.9	-1431.0	-2241.4
$\beta_{yzz}$	789.0	1106.2	838.9	1393.4	812.0	1319.5
$\beta_{zyz}$	837.8	1198.4	838.9	1393.4	851.7	1384.2
$\beta_{xyz}$	-657.0	-857.2	-694.5	-1064.8	2374.5	3778.1
$\beta_{yxz}$	-687.1	-930.0	-694.5	-1064.8	-781.1	-1165.7
$\beta_{zxy}$	-686.2	-932.2	-694.5	-1064.8	-781.3	-1166.0
$\beta_{tot}$	2348.6	3281.9	2468.8	3963.1	2737.0	4247.3

Tab. S8: QTAIM-atomic/group static ( $\lambda = \infty$ ) polarizability tensors (in a.u.) obtained with MP2/6-311++G(2d,2p) method without/*with* polarizing electric field (experimental crystal coordinates) for monoclinic MBANP in the  $abc^*$  axes system for the first molecule of the unit cell (experimental geometry), FF are the results of molecular calculations with finite field method; for groups with hydrogen atoms the heavy atom contribution is given in parentheses.

atom	N1	C1	C2H	C3H	C4	C5H	N2	O1	O2	N3H	C6H	C7H <sub>3</sub>	C8	C9H	C10H	C11H	C12H	C13H	2A5NP	phenyl	rest	SUM	FF
$\alpha_{xx}$	11.4	6.0	10.1(6.6)	10.6(8.9)	8.8	8.1(5.1)	8.6	4.3	10.0	7.3(5.4)	6.8(3.6)	11.2(4.8)	8.1	10.5(6.9)	11.1(8.4)	10.5(9.3)	12.0(8.0)	11.1(8.4)	85.2	63.3	18.0	166.5	166.6
	11.7	5.9	9.8(6.6)	10.4(8.9)	8.9	8.2(5.1)	8.9	4.3	10.8	7.0(5.6)	6.9(3.6)	11.4(4.9)	8.0	10.5(6.9)	11.1(8.4)	10.5(9.3)	11.9(8.0)	10.9(8.4)	85.9	62.9	18.3	167.2	167.3
$\alpha_{yy}$	14.2	10.2	13.8(12.3)	12.5(11.2)	13.7	10.0(9.0)	16.2	7.5	6.2	18.9(18.0)	11.1(9.5)	18.6(10.3)	6.4	9.0(6.6)	8.8(7.7)	9.4(7.1)	9.2(6.8)	8.4(7.4)	123.3	51.0	29.7	204.1	203.8
	14.8	10.7	14.1(12.7)	12.7(11.5)	15.4	10.6(9.6)	18.3	8.2	6.8	19.8(19.1)	11.3(9.6)	18.5(10.3)	6.3	9.0(6.6)	8.8(7.7)	9.4(7.2)	9.3(6.9)	8.3(7.4)	131.5	51.0	29.8	212.4	212.1
$\alpha_{zz}$	7.1	9.4	13.7(11.9)	9.8(6.9)	9.5	10.4(9.1)	7.8	8.8	3.7	17.5(16.7)	11.9(10.4)	12.7(5.3)	13.4	14.5(13.4)	14.6(10.7)	14.2(9.8)	13.2(12.0)	13.1(10.2)	97.8	83.0	24.6	205.4	205.3
	7.4	9.8	13.7(12.1)	9.3(6.5)	9.8	11.0(9.6)	8.3	9.6	3.6	17.5(16.9)	12.2(10.7)	12.8(5.3)	13.5	14.6(13.5)	14.8(10.8)	14.1(9.8)	13.2(12.1)	12.8(10.1)	100.1	83.1	25.0	208.2	208.0
$\alpha_{xy}$	6.6	2.5	2.6(1.4)	3.6(4.3)	3.7	0.9(0.2)	5.6	-1.9	4.2	5.4(4.5)	-0.3(0.8)	2.0(1.9)	-2.6	-2.7(-1.0)	-2.1(-2.1)	-2.2(-1.8)	-2.9(-1.1)	-2.4(-2.3)	33.2	-15.0	1.7	19.9	20.1
	7.0	2.6	2.4(1.4)	4.0(4.7)	4.4	0.8(0.1)	6.5	-2.1	4.9	5.5(4.8)	-0.3(0.8)	2.2(1.9)	-2.6	-2.8(-1.0)	-2.0(-2.0)	-2.1(-1.8)	-2.9(-1.0)	-2.2(-2.2)	35.9	-14.7	1.9	23.1	23.1
$\alpha_{xz}$	-1.6	0.3	1.7(0.4)	0.7(-0.6)	0.4	1.4(0.4)	-0.1	2.0	1.2	-2.1(-2.6)	-1.5(-1.2)	-0.6(-0.4)	0.5	1.7(1.6)	1.7(-0.5)	0.2(0.9)	0.0(0.5)	-0.3(-1.8)	4.0	3.6	-2.1	5.5	5.5
	-1.7	0.1	1.4(0.2)	0.4(-0.7)	0.2	1.4(0.4)	-0.5	2.1	1.1	-2.5(-2.8)	-1.5(-1.3)	-0.6(-0.5)	0.4	1.7(1.6)	1.6(-0.5)	0.2(0.9)	-0.1(0.4)	-0.6(-2.0)	2.0	3.2	-2.2	3.1	3.1
$\alpha_{yz}$	-3.6	-5.7	-4.9(-5.5)	-3.1(-2.2)	-5.0	-4.3(-4.7)	-6.7	-4.6	1.0	-9.8(-10.1)	0.5(0.5)	-0.1(-0.4)	2.9	1.7(1.9)	1.9(2.1)	2.8(0.8)	2.3(2.0)	2.4(2.6)	-46.7	14.0	0.4	-32.3	-32.3
	-3.9	-6.3	-5.4(-6.1)	-3.3(-2.4)	-6.1	-4.7(-5.2)	-7.9	-5.3	1.1	-10.5(-10.7)	0.3(0.4)	0.0(-0.4)	2.9	1.6(1.8)	1.8(2.0)	2.7(0.8)	2.2(1.9)	2.3(2.5)	-52.3	13.5	0.4	-38.5	-38.3
$\alpha_{tso}$	10.9	8.5	12.6(10.3)	11.0(9.0)	10.7	9.5(7.7)	10.9	6.9	6.6	14.6(13.4)	9.9(7.8)	14.2(6.8)	9.3	11.3(9.0)	11.5(8.9)	11.3(8.7)	11.4(8.9)	10.9(8.6)	102.1	65.8	24.1	192.0	191.9
	11.3	8.8	12.5(10.5)	10.8(9.0)	11.4	10.0(8.1)	11.9	7.3	7.1	14.8(13.8)	10.1(8.0)	14.3(6.8)	9.3	11.4(9.0)	11.6(9.0)	11.3(8.8)	11.5(9.0)	10.7(8.6)	105.9	65.7	24.4	195.9	195.8

## S.4 ( $\pm$ )-MBANP & 3MMBANP - additional tables

Tab. S9: Optimized fractional coordinates and Mulliken populations (in e) of ( $\pm$ )-MBANP (*S* form) as obtained from B3LYP/6-31G(d,p) calculations with periodic boundary conditions.

atom	x	y	z	Mulliken charge
N1	0.401788	0.032664	0.363460	-0.5043
C1	0.378832	-0.029941	0.437040	0.4549
C2	0.298818	-0.044437	0.470277	-0.1289
H2	0.280882	-0.092955	0.533711	0.1208
C3	0.243520	0.004111	0.418041	-0.0661
H3	0.181950	-0.005870	0.437506	0.1481
C4	0.268198	0.066950	0.336807	0.1814
C5	0.347888	0.079168	0.314511	0.0941
H5	0.368092	0.128286	0.255866	0.1444
N2	0.212279	0.117975	0.275945	0.3895
O1	0.142374	0.106432	0.302294	-0.4015
O2	0.236756	0.170947	0.197662	-0.4215
N3	0.436588	-0.076537	0.471967	-0.5252
H3n	0.491271	-0.062145	0.431761	0.3225
C6	0.426183	-0.147632	0.546101	-0.0165
H6	0.376208	-0.173456	0.484348	0.1047
C7	0.499078	-0.192534	0.509767	-0.3413
H7a	0.509132	-0.196881	0.374051	0.1148
H7b	0.550560	-0.169316	0.570091	0.1376
H7c	0.491644	-0.246659	0.559211	0.1270
C8	0.408808	-0.145444	0.734146	0.1207
C9	0.353159	-0.192307	0.800591	-0.1463
H9	0.320606	-0.227666	0.717652	0.1118
C10	0.339253	-0.195006	0.973721	-0.1080
H10	0.297839	-0.233335	1.023769	0.1164
C11	0.380553	-0.149870	1.081777	-0.0922
H11	0.369715	-0.151867	1.216154	0.0847
C12	0.436400	-0.102806	1.016557	-0.1238
H12	0.469717	-0.068460	1.100423	0.1069
C13	0.450642	-0.100826	0.843397	-0.1097
H13	0.495188	-0.065370	0.793775	0.1051

Tab. S10: Optimized fractional coordinates and Mulliken populations (in e) of 3MMBANP as obtained from B3LYP/6-31G(d,p) calculations with periodic boundary conditions.

atom	x	y	z	Mulliken charge
N1	0.496142	0.321544	0.110708	-0.4463
C1	0.688808	0.245057	0.091916	0.4205
C2	0.781496	0.330560	0.058133	0.0699
C2a	0.977867	0.229694	0.035992	-0.4139
H2a	0.924710	0.078489	0.029150	0.1349
H2b	1.149854	0.220219	0.051395	0.1345
H2c	1.010583	0.309198	0.010056	0.1414
C3	0.682917	0.507830	0.047130	-0.1056
H3	0.750182	0.581001	0.022310	0.1510
C4	0.492891	0.591826	0.068226	0.1989
C5	0.401606	0.490140	0.099047	0.0537
H5	0.246554	0.548279	0.114292	0.1372
N2	0.390643	0.777875	0.058327	0.3894
O1	0.472292	0.865560	0.030714	-0.4049
O2	0.224018	0.844741	0.078205	-0.4440
N3	0.793311	0.082578	0.106649	-0.5377
H3n	0.942829	0.024694	0.094009	0.3172
C6	0.726908	0.006167	0.143550	0.0030
H6	0.529288	-0.017100	0.143457	0.1237
C7	0.853501	-0.191912	0.150176	-0.3411
H7a	1.051461	-0.173790	0.151978	0.1157
H7b	0.790294	-0.254792	0.176466	0.1296
H7c	0.811828	-0.296543	0.127920	0.1192
C8	0.785540	0.151673	0.174687	0.1423
C9	0.621434	0.176537	0.204095	-0.1273
H9	0.449055	0.097733	0.203724	0.0908
C10	0.671859	0.306719	0.233207	-0.1075
H10	0.538986	0.326991	0.255317	0.0928
C11	0.888103	0.415408	0.233112	-0.1026
H11	0.928474	0.519378	0.255256	0.1068
C12	1.053659	0.390437	0.203882	-0.1047
H12	1.223500	0.472692	0.203721	0.0981
C13	1.003971	0.258575	0.174994	-0.1518
H13	1.136634	0.238957	0.152778	0.1168

Tab. S11: Static polarizability and first hyperpolarizability of ( $\pm$ )-MBANP and 3MMBANP (in a.u.) for the case without and with the electric field originating from the point charges. These are given in the crystal  $abc^*$  axes system fitted to the first molecule of the unit cell of monoclinic MBANP (PBC-optimized crystal coordinates).

	( $\pm$ )-MBANP				3MMBANP			
	B3LYP		MP2		B3LYP		MP2	
	no field	charge field	no field	charge field	no field	charge field	no field	charge field
$\alpha_{xx}$	179.3	178.9	178.8	178.8	182.7	182.8	181.8	182.3
$\alpha_{yy}$	233.3	241.3	226.5	235.6	218.6	227.6	213.3	224.1
$\alpha_{zz}$	172.6	173.7	171.3	172.9	212.7	215.9	209.5	213.7
$\alpha_{xy}$	43.5	47.8	41.0	45.7	28.8	32.4	26.7	31.0
$\alpha_{xz}$	13.9	11.7	15.6	13.2	13.5	10.3	15.1	11.7
$\alpha_{yz}$	-10.0	-12.1	-9.4	-12.1	-27.4	-33.8	-26.0	-33.6
$\alpha_{iso}$	195.1	198.0	192.2	195.8	204.7	208.8	201.5	206.7
$\beta_{xxx}$	-63.8	28.4	-13.8	50.0	-103.9	-16.2	-56.1	11.5
$\beta_{yyy}$	1668.4	2074.1	1781.3	2359.2	980.5	1285.3	1153.2	1781.7
$\beta_{zzz}$	60.8	-59.6	3.7	-112.4	-126.3	-124.2	-208.9	-276.1
$\beta_{xxy}$	262.4	367.2	300.5	406.6	95.8	156.6	136.5	221.7
$\beta_{xxz}$	-128.1	-154.7	-130.7	-180.7	-94.6	-98.7	-102.8	-141.9
$\beta_{xyy}$	821.6	1008.0	839.6	1087.5	491.1	618.6	522.1	771.8
$\beta_{xzz}$	65.8	96.2	80.5	121.3	182.3	270.4	188.9	324.6
$\beta_{yyz}$	-565.5	-665.1	-640.8	-837.4	-751.5	-920.2	-847.5	-1222.3
$\beta_{yzz}$	65.0	115.0	137.3	208.2	366.6	481.8	457.7	685.0
$\beta_{xyz}$	-334.8	-370.8	-334.6	-422.8	-382.0	-456.2	-379.0	-543.0
$\beta_{tot}$	1350.0	1758.6	1510.2	2053.0	1098.5	1441.1	1318.1	2003.1

Tab. S12: Dynamic (at  $\lambda = 1064\text{ nm}$ ) polarizability and first hyperpolarizability of ( $\pm$ )-MBANP and 3MMBANP (in a.u.) for the case without and with the electric field originating from the point charges. These are given in the crystal  $abc^*$  axes system fitted to the first molecule of the unit cell of monoclinic MBANP (PBC-optimized crystal coordinates).

	( $\pm$ )-MBANP				3MMBANP			
	B3LYP		MP2		B3LYP		MP2	
	no field	charge field	no field	charge field	no field	charge field	no field	charge field
$\alpha_{xx}$	182.4	182.3	181.9	182.3	185.4	185.7	184.3	185.1
$\alpha_{yy}$	241.2	250.7	234.1	244.8	224.7	235.6	219.1	231.9
$\alpha_{zz}$	175.0	176.3	173.7	175.5	217.2	221.3	214.1	219.1
$\alpha_{xy}$	46.5	51.5	43.9	49.4	30.7	35.0	28.4	33.3
$\alpha_{xz}$	13.6	11.2	15.3	12.7	12.8	9.1	14.6	10.7
$\alpha_{yz}$	-11.3	-13.8	-10.7	-13.8	-30.2	-37.8	-28.8	-37.6
$\alpha_{iso}$	199.6	203.1	196.6	200.8	209.1	214.2	205.9	212.0
$\beta_{xxx}$	8.7	173.2	77.7	210.9	-96.7	32.9	-31.5	91.9
$\beta_{yyy}$	3129.3	4134.4	3327.8	4695.7	1920.2	2779.2	2242.3	3832.1
$\beta_{zzz}$	50.6	-106.3	-30.2	-188.3	-348.1	-450.7	-480.6	-735.7
$\beta_{xxy}$	557.2	798.6	611.2	873.1	233.9	383.3	289.8	532.4
$\beta_{yyx}$	517.2	757.6	584.4	847.0	201.7	345.3	271.2	476.2
$\beta_{xxz}$	-231.1	-295.2	-241.2	-345.5	-200.2	-262.8	-207.6	-353.4
$\beta_{zxx}$	-211.1	-276.8	-228.2	-335.5	-173.3	-233.3	-195.4	-308.0
$\beta_{xyy}$	1495.0	1954.2	1542.0	2127.8	871.0	1188.1	936.8	1580.4
$\beta_{yyx}$	1520.1	2000.1	1563.3	2181.5	916.1	1302.7	984.4	1624.1
$\beta_{xzz}$	109.8	155.8	141.6	208.3	318.5	481.0	343.5	627.1
$\beta_{zzx}$	132.7	190.7	159.0	241.9	384.6	613.8	389.6	714.7
$\beta_{yyz}$	-1018.4	-1259.5	-1156.4	-1581.1	-1384.4	-1894.9	-1579.7	-2542.4
$\beta_{zyy}$	-1014.4	-1278.8	-1154.6	-1608.4	-1431.8	-1991.5	-1608.0	-2597.7
$\beta_{yzz}$	148.1	228.2	266.4	391.6	700.7	997.1	873.1	1426.2
$\beta_{zyz}$	181.0	277.0	290.4	437.3	777.9	1137.6	931.6	1537.5
$\beta_{xyz}$	-553.8	-651.3	-575.6	-760.8	-641.9	-838.6	-658.1	-1069.6
$\beta_{yxz}$	-577.8	-688.3	-592.0	-797.7	-696.7	-956.0	-695.7	-1139.0
$\beta_{zxy}$	-558.8	-673.0	-581.0	-790.1	-691.1	-952.3	-700.7	-1119.9
$\beta_{tot}$	2608.2	3552.4	2875.2	4116.5	2205.2	3196.9	2595.1	4371.7

Tab. S13: QTAIM-atomic/group static ( $\lambda = \infty$ ) polarizability tensors (in a.u.) obtained with MP2/6-311++G(d,p) method without/with polarizing electric field (optimized crystal coordinates) for ( $\pm$ )-MBANP (*S* form) for the first molecule of the unit cell; for groups with hydrogen atoms the heavy atom contribution is given in parentheses.

atom	N1	C1	C2H	C3H	C4	C5H	N2	O1	O2	N3H	C6H	C7H <sub>3</sub>	C8	C9H	C10H	C11H	C12H	C13H	2A5NP	phenyl	rest	SUM	FF
$\alpha_{xx}$	10.7	11.2	14.8(13.6)	11.1(7.6)	13.0	13.1(11.9)	14.7	2.9	13.5	15.1(12.7)	7.4(5.6)	14.2(7.6)	6.4	8.7(6.8)	9.5(6.8)	8.9(7.9)	9.4(7.3)	9.0(6.3)	120.1	51.9	21.6	193.6	193.6
	11.2	11.7	15.3(14.2)	11.0(7.7)	14.0	13.6(12.4)	15.7	2.8	14.1	15.1(13.2)	7.5(5.7)	13.9(7.4)	6.4	8.5(6.7)	9.7(7.2)	9.3(8.2)	9.5(7.4)	8.5(6.1)	124.5	51.9	21.3	197.8	197.4
$\alpha_{yy}$	19.0	9.0	11.5(8.5)	13.4(12.5)	12.2	9.0(6.0)	13.9	11.2	3.0	20.1(19.6)	9.0(6.8)	14.1(7.0)	7.4	10.6(7.9)	10.3(7.3)	9.2(8.3)	9.5(7.3)	8.5(6.2)	122.2	55.5	23.1	200.8	200.9
	19.6	9.3	11.5(8.7)	13.7(12.9)	13.0	8.8(5.9)	14.9	12.0	2.9	20.9(20.5)	9.0(6.8)	14.0(7.0)	7.5	10.6(8.1)	9.9(7.2)	9.5(8.5)	9.8(7.6)	8.5(6.3)	126.6	55.8	22.9	205.3	205.3
$\alpha_{zz}$	9.0	4.6	7.9(6.3)	8.0(7.2)	7.0	6.0(4.7)	6.2	6.3	3.2	9.6(9.0)	12.6(10.6)	13.2(5.7)	14.7	14.3(12.1)	14.3(12.5)	15.1(9.6)	15.1(12.1)	14.9(13.5)	67.8	88.4	25.8	182.0	182.2
	9.2	4.6	8.0(6.5)	8.4(7.5)	7.4	5.9(4.6)	6.5	6.7	3.2	9.7(9.2)	12.7(10.7)	13.3(5.7)	14.8	14.3(12.1)	14.2(12.6)	15.5(9.8)	15.2(12.3)	14.9(13.6)	69.6	88.9	26.0	184.6	184.5
$\alpha_{xy}$	-4.3	-3.7	-3.7(-4.5)	-2.5(-3.2)	-3.6	-1.4(-2.5)	-6.8	0.9	-0.6	-9.8(-10.0)	-2.0(-2.8)	-3.5(-2.9)	2.9	3.3(2.0)	3.3(1.5)	2.5(2.4)	2.9(1.7)	2.6(1.1)	-35.4	17.5	-5.5	-23.3	-23.2
	-4.5	-4.3	-4.2(-4.9)	-2.9(-3.6)	-4.6	-1.7(-2.8)	-7.7	0.8	-0.8	-10.8(-10.9)	-2.3(-3.0)	-3.4(-2.9)	2.9	3.0(1.9)	3.0(1.3)	2.2(2.2)	2.8(1.6)	2.5(1.1)	-40.6	16.5	-5.7	-29.9	-29.7
$\alpha_{xz}$	1.9	1.5	1.9(2.3)	1.3(1.8)	1.8	0.6(1.1)	3.2	-0.6	-0.3	3.6(4.1)	-0.4(-1.3)	-1.5(-1.2)	-0.3	-1.0(-2.0)	-0.9(0.2)	-0.6(0.0)	0.6(-1.0)	0.7(1.3)	14.9	-1.6	-2.0	11.3	11.3
	2.0	1.8	2.1(2.4)	1.5(1.9)	2.4	0.7(1.3)	3.7	-0.5	-0.2	4.1(4.5)	-0.4(-1.3)	-1.7(-1.2)	-0.2	-1.1(-2.0)	-0.9(0.2)	-0.9(-0.1)	0.6(-0.9)	0.8(1.4)	17.6	-1.6	-2.1	13.9	13.8
$\alpha_{yz}$	-7.5	-3.2	-3.0(-1.8)	-4.3(-4.2)	-4.1	-2.1(-0.9)	-6.3	-4.9	0.2	-8.2(-8.1)	-0.2(-0.9)	0.8(0.7)	-1.1	-1.1(-2.4)	-1.1(0.2)	-0.3(0.0)	0.3(-1.3)	0.1(0.8)	-43.5	-3.3	0.5	-46.3	-46.4
	-7.9	-3.4	-3.1(-1.8)	-4.4(-4.4)	-4.7	-2.1(-1.0)	-6.9	-5.3	0.2	-8.7(-8.6)	-0.4(-1.0)	0.6(0.6)	-1.2	-1.2(-2.4)	-0.8(0.3)	-0.2(0.1)	0.3(-1.2)	0.0(0.7)	-46.3	-3.2	0.2	-49.2	-49.3
$\alpha_{iso}$	12.9	8.3	11.4(9.5)	10.8(9.1)	10.7	9.4(7.5)	11.6	6.8	6.5	14.9(13.8)	9.7(7.7)	13.9(6.7)	9.5	11.2(8.9)	11.4(8.9)	11.1(8.6)	11.3(8.9)	10.8(8.7)	103.4	65.2	23.5	192.1	192.2
	13.3	8.6	11.6(9.8)	11.0(9.4)	11.5	9.4(7.6)	12.4	7.2	6.7	15.2(14.3)	9.7(7.8)	13.7(6.7)	9.6	11.1(9.0)	11.3(9.0)	11.4(8.8)	11.5(9.1)	10.7(8.6)	106.9	65.5	23.4	195.9	195.8

Tab. S14: QTAIM-atomic/group static ( $\lambda = \infty$ ) polarizability tensors (in a.u.) obtained with MP2/6-311++G(d,p) method without/*with* polarizing electric field (optimized crystal coordinates) for 3MMBANP for the first molecule of the unit cell; for groups with hydrogen atoms the heavy atom contribution is given in parentheses.

atom	N1	C1	C2	C3H	C4	C5H	C2aH3	N2	O1	O2	N3H	C6H	C7H3	C8	C9H	C10H	C11H	C12H	C13H	2A5NP	phenyl*	rest	SUM	FF
$\alpha_{xx}$	12.0	5.7	7.7	10.2(9.1)	8.5	7.6(4.9)	14.1(7.2)	8.3	3.4	8.9	7.2(5.8)	6.7(3.6)	11.2(4.9)	8.3	10.3(6.5)	11.1(8.4)	10.5(9.3)	12.0(7.8)	11.4(8.6)	79.8	63.5	32.0	175.3	175.4
	12.5	5.7	7.9	10.2(9.1)	9.0	7.6(4.9)	13.7(7.1)	8.8	3.4	9.7	7.0(5.9)	6.6(3.6)	11.3(4.9)	8.4	10.1(6.5)	11.1(8.5)	10.5(9.4)	12.0(7.9)	11.3(8.8)	81.6	63.4	31.7	176.7	176.8
$\alpha_{yy}$	14.9	9.2	12.4	13.0(11.8)	14.3	10.0(9.1)	11.4(5.2)	18.2	7.4	6.2	18.5(17.5)	11.1(9.6)	18.3(10.2)	6.5	8.9(6.9)	8.8(7.8)	9.5(6.9)	9.1(7.0)	8.4(7.5)	124.1	51.1	40.8	216.0	215.9
	15.8	10.1	13.1	13.5(12.4)	16.9	10.6(9.8)	10.9(5.1)	20.9	8.2	7.0	19.7(18.9)	11.1(9.6)	18.4(10.3)	6.3	8.9(6.9)	8.8(7.8)	9.5(7.0)	9.1(7.1)	8.3(7.5)	135.9	50.8	40.4	227.2	227.2
$\alpha_{zz}$	6.3	10.5	12.6	9.0(6.0)	9.7	10.7(9.1)	13.5(6.5)	8.0	9.7	4.3	18.1(17.3)	10.6(9.4)	12.6(5.3)	12.4	13.8(12.7)	13.9(10.1)	13.0(9.3)	12.4(11.3)	11.9(9.1)	98.9	77.4	36.7	213.0	213.3
	6.4	10.9	12.9	8.4(5.7)	10.2	11.3(9.6)	13.2(6.4)	8.5	10.5	4.1	18.2(17.6)	10.8(9.5)	12.6(5.3)	12.5	13.9(12.8)	14.1(10.2)	12.9(9.3)	12.5(11.4)	11.8(9.3)	101.3	77.8	36.6	215.6	216.1
$\alpha_{xy}$	-7.7	-2.6	-3.8	-4.6(-5.1)	-4.2	-1.3(-0.6)	-1.7(-1.6)	-7.1	1.4	-4.3	-5.8(-5.0)	0.3(-0.7)	-1.7(-1.7)	2.4	2.5(0.8)	1.8(2.1)	2.1(1.5)	2.7(0.8)	2.1(2.4)	-40.0	13.7	-3.1	-29.4	-29.2
	-8.3	-2.9	-4.0	-5.2(-5.6)	-5.3	-1.2(-0.6)	-1.5(-1.5)	-8.2	1.4	-5.1	-6.3(-5.7)	0.4(-0.7)	-1.8(-1.8)	2.5	2.5(0.8)	1.8(2.1)	2.0(1.5)	2.7(0.8)	2.0(2.3)	-45.1	13.5	-2.9	-34.5	-34.3
$\alpha_{xz}$	0.5	-0.5	-2.2	-1.0(-0.1)	-1.0	-1.3(0.0)	-2.6(-2.3)	-0.4	-1.4	-2.7	3.1(3.5)	1.9(1.8)	0.7(0.5)	-0.4	-1.8(-1.5)	-1.6(0.5)	-0.3(-1.0)	-0.1(-0.4)	0.4(1.9)	-7.0	-3.8	-0.1	-10.9	-10.8
	0.7	-0.2	-2.0	-0.6(0.3)	-0.5	-1.2(0.1)	-2.6(-2.3)	0.1	-1.3	-2.7	3.6(3.9)	1.8(1.8)	0.8(0.5)	-0.3	-1.8(-1.5)	-1.6(0.6)	-0.3(-1.0)	0.0(-0.3)	0.6(2.0)	-4.0	-3.4	0.1	-7.3	-7.3
$\alpha_{yz}$	-2.3	-5.1	-4.1	-1.8(-0.7)	-4.0	-4.3(-4.7)	2.3(1.7)	-5.6	-5.3	2.3	-9.9(-10.2)	0.7(0.8)	-0.5(-0.7)	3.4	2.2(2.4)	2.6(2.2)	3.2(1.1)	2.6(2.5)	2.8(2.7)	-40.1	16.9	2.5	-20.7	-20.8
	-2.5	-5.8	-4.7	-1.8(-0.8)	-5.2	-4.8(-5.3)	2.5(1.8)	-6.8	-6.1	2.6	-10.9(-11.1)	0.4(0.6)	-0.4(-0.7)	3.3	2.1(2.3)	2.5(2.1)	3.0(1.0)	2.5(2.4)	2.7(2.5)	-46.1	16.1	2.5	-27.6	-27.6
$\alpha_{iso}$	11.1	8.5	10.9	10.8(8.9)	10.8	9.4(7.7)	13.0(6.3)	11.5	6.8	6.5	14.6(13.5)	9.5(7.5)	14.0(6.8)	9.1	11.0(8.7)	11.2(8.8)	11.0(8.5)	11.1(8.7)	10.6(8.4)	100.9	64.0	36.5	201.4	201.5
	11.5	8.9	11.3	10.7(9.1)	12.0	9.8(8.1)	12.6(6.2)	12.7	7.4	6.9	15.0(14.1)	9.5(7.6)	14.1(6.8)	9.1	11.0(8.7)	11.3(8.8)	11.0(8.6)	11.2(8.8)	10.5(8.5)	106.3	64.0	36.2	206.5	206.7

\* C2aH3 group not taken into account

## S.5 NMMBANP - additional tables

Tab. S15: Optimized fractional coordinates and Mulliken populations (in e) of NMMBANP as obtained from B3LYP/6-31G(d,p) calculations with periodic boundary conditions.

atom	molecule 1				molecule 2			
	x	y	z	Mulliken charge	x	y	z	Mulliken charge
N1	0.438972	-0.030334	0.651645	-0.4678	0.624508	0.337002	-0.130683	-0.4690
C1	0.401529	0.052299	0.643533	0.4583	0.770232	0.323960	-0.077781	0.4611
C2	0.434120	0.102510	0.524966	-0.1553	0.809421	0.297762	0.067267	-0.1517
H2	0.398274	0.166931	0.513112	0.1442	0.925206	0.291059	0.111479	0.1285
C3	0.512770	0.067584	0.422842	-0.0703	0.697667	0.281872	0.151742	-0.0629
H3	0.541958	0.104599	0.333826	0.1426	0.723807	0.261996	0.261806	0.1523
C4	0.555076	-0.016966	0.436138	0.1931	0.547663	0.292466	0.092683	0.1900
C5	0.512344	-0.063501	0.550884	0.0425	0.517841	0.321882	-0.047943	0.0694
H5	0.539710	-0.129596	0.561198	0.1608	0.403633	0.334641	-0.093336	0.1543
N2	0.642746	-0.054872	0.335865	0.3702	0.427842	0.274871	0.175218	0.3697
O1	0.686180	-0.010683	0.240261	-0.4104	0.459142	0.253961	0.302420	-0.4092
O2	0.674413	-0.130353	0.350095	-0.4088	0.296499	0.279825	0.117259	-0.4172
N3	0.333313	0.085331	0.752003	-0.4160	0.876608	0.337190	-0.166121	-0.4126
C3n	0.290844	0.173551	0.748806	-0.2066	1.035210	0.329956	-0.111783	-0.2089
H3na	0.259889	0.191360	0.853050	0.1240	1.103209	0.343491	-0.197619	0.1451
H3nb	0.195495	0.185855	0.668452	0.1514	1.061677	0.266678	-0.072825	0.1314
H3nc	0.384015	0.213417	0.726133	0.1450	1.065949	0.374567	-0.025392	0.1507
C6	0.292159	0.033342	0.872268	-0.0395	0.838283	0.361505	-0.317776	-0.0340
H6	0.324963	-0.030009	0.846691	0.1540	0.716242	0.359543	-0.335153	0.1536
C7	0.383032	0.058603	1.013412	-0.3390	0.887441	0.451768	-0.341877	-0.3390
C7a	0.502822	0.054788	1.003478	0.1274	0.830002	0.493527	-0.274051	0.1161
C7b	0.360058	0.014993	1.097670	0.1280	0.855983	0.469754	-0.453307	0.1186
C7c	0.357975	0.122051	1.046963	0.1039	1.008317	0.461102	-0.316527	0.1266
C8	0.122364	0.032917	0.875021	0.1223	0.896744	0.296127	-0.416358	0.1046
C9	0.035323	-0.017107	0.775082	-0.1285	0.834714	0.215400	-0.419569	-0.1155
CH9	0.091116	-0.054785	0.700973	0.0989	0.749752	0.200553	-0.349981	0.0961
C10	-0.119965	-0.020740	0.772801	-0.0990	0.881673	0.154047	-0.509813	-0.0865
H10	-0.185993	-0.060642	0.696405	0.0922	0.831550	0.092267	-0.511008	0.0886
C11	-0.191271	0.025616	0.871291	-0.0918	0.992003	0.172251	-0.598826	-0.1129
H11	-0.311977	0.021601	0.870333	0.0991	1.027246	0.124506	-0.669889	0.1010
C12	-0.106153	0.075521	0.971426	-0.0932	1.056217	0.251851	-0.595537	-0.1218
H12	-0.160629	0.111228	1.048917	0.0993	1.143458	0.265853	-0.663037	0.1141
C13	0.049676	0.079251	0.972988	-0.1386	1.008251	0.313209	-0.504697	-0.1094
H13	0.114487	0.118106	1.052031	0.0988	1.060924	0.374268	-0.501883	0.0871

Tab. S16: Static polarizability and first hyperpolarizability of NMMBANP (first and second molecule of the asymmetric unit) (in a.u.) for the case without and with the electric field originating from the point charges. These are given in the crystal  $abc^*$  axes system fitted to the first molecule of the unit cell of monoclinic MBANP (PBC-optimized crystal coordinates).

	molecule 1				molecule 2			
	B3LYP		MP2		B3LYP		MP2	
	no field	charge field	no field	charge field	no field	charge field	no field	charge field
$\alpha_{xx}$	177.7	177.6	177.5	177.7	176.5	178.1	176.0	178.1
$\alpha_{yy}$	293.4	305.6	284.1	297.7	254.6	268.0	249.0	265.9
$\alpha_{zz}$	170.4	172.1	169.9	172.0	210.9	214.2	207.7	212.1
$\alpha_{xy}$	31.6	36.0	30.4	35.3	47.2	52.9	45.0	52.0
$\alpha_{xz}$	-12.7	-15.7	-10.5	-13.6	-30.3	-34.3	-26.6	-31.2
$\alpha_{yz}$	-52.6	-58.4	-50.4	-56.8	-62.6	-71.0	-59.8	-70.0
$\alpha_{iso}$	213.8	218.4	210.5	215.8	214.0	220.1	210.9	218.7
$\beta_{xxxx}$	-89.2	-2.7	-50.4	19.7	-96.6	-3.6	-58.5	37.0
$\beta_{yyyy}$	2280.9	2931.9	2530.6	3507.1	1816.3	2360.0	2089.0	3196.7
$\beta_{zzz}$	-57.0	-96.3	-135.7	-191.0	-136.1	-170.7	-241.8	-280.3
$\beta_{xxy}$	180.0	256.7	228.5	327.6	291.3	405.7	349.2	539.5
$\beta_{xxz}$	-120.3	-132.0	-134.1	-170.2	-194.6	-239.5	-219.0	-317.3
$\beta_{xyy}$	890.0	1065.4	934.5	1258.9	908.0	1171.0	990.3	1505.8
$\beta_{xzz}$	157.3	208.9	171.4	257.1	265.8	388.0	302.0	486.8
$\beta_{yyz}$	-1047.6	-1280.0	-1175.1	-1576.4	-1069.2	-1297.6	-1210.9	-1730.2
$\beta_{yzz}$	266.5	367.9	357.7	524.5	432.8	611.8	541.5	825.2
$\beta_{xyz}$	-467.8	-529.8	-483.8	-628.2	-572.3	-697.6	-603.0	-876.1
$\beta_{tot}$	1883.7	2440.2	2156.4	3006.9	1856.5	2455.1	2179.6	3305.2

Tab. S17: Dynamic (at  $\lambda = 1064\text{ nm}$ ) polarizability and first hyperpolarizability of NMMBANP (first and second molecule of the asymmetric unit) (in a.u.) for the case without and with the electric field originating from the point charges. These are given in the crystal  $abc^*$  axes system fitted to the first molecule of the unit cell of monoclinic MBANP (PBC-optimized crystal coordinates).

	molecule 1				molecule 2			
	B3LYP		MP2		B3LYP		MP2	
	no field	charge field	no field	charge field	no field	charge field	no field	charge field
$\alpha_{xx}$	180.4	180.5	180.2	180.7	179.7	181.8	178.9	181.9
$\alpha_{yy}$	304.8	319.7	295.0	311.4	263.6	280.2	257.2	278.0
$\alpha_{zz}$	173.4	175.6	172.8	175.5	215.7	220.0	213.3	217.8
$\alpha_{xy}$	34.2	39.5	33.0	38.9	50.7	57.7	48.0	56.9
$\alpha_{xz}$	-13.8	-17.2	-11.6	-15.1	-32.5	-37.4	-28.8	-34.2
$\alpha_{yz}$	-56.4	-63.4	-54.1	-61.7	-67.1	-77.3	-64.5	-76.2
$\alpha_{iso}$	219.5	225.3	216.0	222.5	219.6	227.4	216.4	225.9
$\beta_{xxx}$	-58.0	86.4	-11.0	163.6	-10.6	202.1	45.6	372.8
$\beta_{yyy}$	4790.3	6848.6	5306.2	8071.7	3701.0	5426.6	4224.1	7269.4
$\beta_{zzz}$	-194.7	-308.2	-324.5	-499.6	-436.6	-713.8	-623.9	-956.9
$\beta_{xxy}$	427.8	632.3	480.7	855.5	663.4	1029.1	755.7	1526.9
$\beta_{yyx}$	393.3	603.9	472.2	805.7	614.6	978.2	715.3	1181.8
$\beta_{xxz}$	-243.0	-307.4	-266.5	-416.8	-422.9	-622.4	-467.3	-883.5
$\beta_{zxx}$	-220.0	-288.5	-234.6	-394.2	-384.8	-580.0	-445.8	-699.8
$\beta_{xyy}$	1703.0	2258.4	1785.4	2808.8	1766.3	2555.7	1934.7	3852.2
$\beta_{yyx}$	1757.7	2342.4	1848.5	2826.2	1797.8	2639.2	1966.8	3277.9
$\beta_{xzz}$	280.1	391.4	311.0	520.7	522.5	842.1	601.4	1180.8
$\beta_{zzx}$	322.5	457.4	349.8	582.5	570.3	918.6	642.6	1103.5
$\beta_{yyz}$	-2111.9	-2866.4	-2383.0	-3497.5	-2152.4	-3027.2	-2450.8	-3946.8
$\beta_{zyy}$	-2122.0	-2930.1	-2394.1	-3613.5	-2179.8	-3084.6	-2487.1	-4063.1
$\beta_{yzz}$	608.8	895.0	787.9	1224.7	977.5	1559.2	1199.7	2009.6
$\beta_{zyz}$	670.9	993.0	841.1	1340.3	1029.2	1616.7	1246.6	2097.6
$\beta_{xyz}$	-825.1	-1033.6	-858.0	-1303.7	-1076.5	-1500.6	-1157.9	-2157.1
$\beta_{yxz}$	-882.7	-1119.5	-915.4	-1359.2	-1119.6	-1590.6	-1196.5	-1919.6
$\beta_{zxy}$	-853.5	-1092.9	-900.6	-1352.7	-1106.0	-1570.2	-1188.5	-1915.8
$\beta_{tot}$	4013.3	5735.2	4533.8	7013.2	3944.3	5921.6	4565.8	7903.1

Tab. S18: QTAIM-atomic/group static ( $\lambda = \infty$ ) polarizability tensors (in a.u.) obtained with MP2/6-311++G(d,p) method without/with polarizing electric field (optimized crystal coordinates) for NMMBANP (first molecule of the asymmetric unit); for groups with hydrogen atoms the heavy atom contribution is given in parenthesis.

atom	N1	C1	C2H	C3H	C4	C5H	N2	O1	O2	N3	C6H3	C6H	C7H3	C8	C9H	C10H	C11H	C12H	C13H	2A5NP	phenyl	rest	SUM	FF
$\alpha_{xx}$	12.7	7.8	11.3(10.3)	9.8(8.8)	10.8	8.8(7.9)	12.0	6.2	4.7	12.8	11.7(4.9)	14.9(12.5)	13.1(6.0)	16.4	15.4(13.6)	15.5(13.2)	15.9(10.0)	15.5(12.9)	15.3(13.7)	96.9	93.9	39.7	230.5	230.7
	13.3	8.9	11.4(10.5)	10.2(9.2)	12.3	9.1(8.3)	13.3	6.7	5.1	13.4	11.3(4.9)	15.3(12.7)	12.9(5.9)	16.7	15.7(13.8)	15.7(13.3)	15.7(10.0)	15.6(13.0)	15.4(13.9)	103.1	94.8	39.5	237.4	237.5
$\alpha_{yy}$	14.0	6.5	10.6(6.8)	11.9(10.0)	10.3	8.8(5.0)	11.1	4.3	12.5	12.9	12.4(5.8)	6.0(3.4)	10.4(4.2)	6.0	8.2(6.3)	9.1(6.7)	8.6(7.6)	9.1(7.0)	8.1(6.0)	102.9	49.1	28.8	180.8	180.9
	14.4	6.4	10.1(6.6)	11.9(10.1)	10.6	8.2(4.8)	11.6	4.2	13.3	13.1	11.8(5.7)	6.2(3.5)	10.3(4.2)	6.0	8.5(6.5)	9.1(6.8)	8.5(7.6)	9.1(6.9)	8.0(6.0)	103.8	49.2	28.3	181.3	181.3
$\alpha_{zz}$	13.8	12.0	15.6(14.5)	11.3(8.6)	13.4	12.6(11.8)	14.1	10.4	3.1	19.1	9.0(3.7)	11.4(8.6)	16.9(9.2)	7.8	8.6(6.3)	10.0(7.5)	9.6(8.7)	10.7(7.6)	10.9(8.1)	125.3	57.6	37.3	220.3	219.9
	14.6	12.9	16.0(15.2)	11.5(9.0)	15.0	13.2(12.5)	15.6	11.3	3.2	20.0	8.7(3.6)	11.7(8.6)	17.1(9.4)	7.9	8.6(6.3)	10.2(7.6)	9.5(8.6)	10.5(7.4)	10.9(8.1)	133.3	57.6	37.5	228.4	228.7
$\alpha_{xy}$	-7.1	-1.5	-1.2(-0.5)	-3.2(-3.9)	-3.1	-0.6(0.0)	-5.4	2.3	-3.9	-4.2	-0.6(-0.6)	-0.4(0.5)	0.9(0.6)	0.5	1.3(2.1)	1.0(-0.4)	0.3(0.0)	-0.5(0.8)	-0.6(-1.4)	-28.0	2.0	-0.1	-26.2	-26.0
	-7.5	-1.6	-1.3(-0.7)	-3.6(-4.2)	-3.8	-0.4(0.1)	-6.2	2.4	-4.6	-4.6	-0.7(-0.6)	-0.5(0.4)	1.0(0.6)	0.4	1.2(2.0)	0.9(-0.5)	0.1(-0.1)	-0.7(0.7)	-0.8(-1.5)	-31.1	1.2	-0.1	-30.1	-30.1
$\alpha_{yz}$	-8.4	-6.4	-6.2(-6.4)	-4.3(-3.6)	-6.8	-5.3(-5.4)	-10.0	-4.6	-0.3	-12.3	0.1(-0.1)	-2.6(-1.0)	1.6(1.7)	-2.1	-0.2(0.9)	-0.1(-1.5)	-0.7(-0.6)	-1.9(0.0)	-2.3(-3.1)	-64.6	-7.4	-0.9	-72.9	-72.9
	-8.9	-7.0	-6.8(-6.9)	-4.7(-4.0)	-8.3	-5.8(-5.9)	-11.4	-5.2	-0.5	-13.1	-0.1(-0.2)	-3.0(-1.2)	1.4(1.6)	-2.3	-0.3(0.8)	-0.1(-1.5)	-0.7(-0.5)	-1.8(0.0)	-2.5(-3.2)	-71.7	-7.7	-1.7	-81.0	-81.0
$\alpha_{yz}$	5.6	0.5	-0.4(0.3)	1.4(2.9)	1.5	-0.8(-0.4)	3.7	-3.2	1.4	3.2	0.7(0.4)	1.3(0.8)	0.1(0.5)	2.7	2.3(1.1)	2.9(1.4)	2.3(2.3)	3.1(1.5)	3.2(1.7)	12.8	16.5	2.1	31.4	31.2
	5.9	0.8	-0.1(0.5)	1.9(3.4)	2.3	-0.7(-0.4)	4.6	-3.3	2.0	3.8	1.0(0.5)	1.3(0.9)	0.3(0.6)	2.8	2.2(1.0)	2.8(1.3)	2.3(2.3)	3.2(1.6)	3.3(1.8)	17.1	16.6	2.6	36.3	26.3
$\alpha_{iso}$	13.5	8.8	12.5(10.5)	11.0(9.2)	11.5	10.0(8.3)	12.4	7.0	6.8	14.9	11.0(4.8)	10.8(8.2)	13.5(6.5)	10.1	10.8(8.7)	11.5(9.1)	11.4(8.8)	11.8(9.2)	11.4(9.3)	108.4	66.9	35.3	210.5	210.5
	14.1	9.2	12.5(10.8)	11.2(9.4)	12.6	10.2(8.5)	13.5	7.4	7.2	15.5	10.6(4.7)	11.1(8.3)	13.4(6.5)	10.2	10.9(8.8)	11.7(9.3)	11.3(8.7)	11.7(9.1)	11.4(9.3)	113.4	67.2	35.1	215.7	215.8

Tab. S19: QTAIM-atomic/group static ( $\lambda = \infty$ ) polarizability tensors (in a.u.) obtained with MP2/6-311++G(d,p) method without/with polarizing electric field (optimized crystal coordinates) for NMMBANP (second molecule of the asymmetric unit); for groups with hydrogen atoms the heavy atom contribution is given in parentheses.

atom	N1	C1	C2H	C3H	C4	C5H	N2	O1	O2	N3	CH <sub>3</sub>	C6H	C7H <sub>3</sub>	C8	C9H	C10H	C11H	C12H	C13H	2A5NP	phenyl	rest	SUM	FF
$\alpha_{xx}$	23.8	10.1	13.4(10.2)	15.7(14.9)	15.4	10.4(7.2)	19.9	3.0	14.1	21.1	12.8(6.2)	9.7(5.8)	10.7(4.5)	8.5	9.0(6.2)	10.2(8.7)	10.4(8.7)	11.7(7.8)	11.1(9.3)	147.0	60.9	33.1	241.0	240.9
	25.2	10.9	13.8(10.7)	16.8(16.1)	18.3	10.2(7.9)	23.1	3.0	16.0	22.8	12.5(6.2)	10.0(6.0)	10.5(4.5)	8.7	9.0(6.2)	10.4(8.9)	10.6(8.8)	11.4(7.9)	11.1(9.4)	160.1	61.2	33.0	254.3	254.3
$\alpha_{yy}$	5.0	3.5	7.1(6.2)	6.1(5.0)	5.5	5.7(4.8)	3.6	4.1	3.2	3.4	10.5(4.1)	10.7(9.7)	16.5(9.3)	10.8	12.6(11.4)	13.5(8.9)	12.6(9.4)	11.8(10.7)	11.3(7.8)	47.2	72.7	37.7	157.6	157.4
	5.2	3.5	7.0(6.2)	6.0(5.1)	5.7	5.6(4.8)	3.7	4.4	3.3	3.5	10.0(4.0)	10.7(9.7)	16.6(9.4)	10.6	12.5(11.3)	13.4(8.8)	12.6(9.5)	11.8(10.7)	11.3(7.8)	48.0	72.2	37.4	157.6	157.6
$\alpha_{zz}$	11.9	12.7	17.1(15.3)	11.2(7.5)	13.6	14.0(12.8)	14.0	14.0	3.1	20.2	9.7(4.1)	12.1(9.0)	13.0(5.5)	11.0	10.6(8.4)	10.9(9.8)	11.3(8.3)	11.8(9.0)	11.9(10.9)	131.9	67.4	34.9	234.2	234.3
	12.5	13.6	17.8(16.2)	11.0(7.4)	15.4	14.9(13.7)	15.8	15.7	2.7	21.3	9.1(3.8)	12.5(9.2)	13.1(5.6)	11.2	10.6(8.4)	11.0(10.0)	11.7(8.7)	11.9(9.3)	12.0(11.0)	140.8	68.4	34.7	243.9	244.2
$\alpha_{xy}$	0.8	0.8	1.1(1.4)	0.5(0.7)	1.5	0.3(1.0)	2.7	0.5	-0.1	2.0	0.0(0.0)	-0.6(-0.6)	1.1(1.1)	-0.4	0.1(-0.6)	0.5(-0.8)	-0.2(1.1)	0.4(-0.2)	0.5(-1.0)	10.0	0.8	0.5	11.4	11.5
	1.0	1.1	1.3(1.7)	0.7(0.9)	2.3	0.7(1.3)	3.4	0.7	0.0	2.3	-0.2(0.0)	-0.6(-0.6)	1.2(1.2)	-0.6	-0.1(-0.7)	0.6(-0.8)	-0.3(1.0)	0.2(-0.4)	0.4(-1.1)	13.6	0.1	0.4	14.2	14.2
$\alpha_{yz}$	-7.7	-4.7	-3.8(-5.2)	-2.6(-3.2)	-5.0	-2.9(-4.1)	-7.8	-1.6	2.0	-9.0	0.6(0.3)	-4.7(-3.6)	-0.9(-0.7)	-5.2	-3.1(-1.5)	-3.2(-3.3)	-3.5(-2.3)	-4.5(-2.2)	-5.1(-4.8)	-43.2	-24.6	-5.1	-72.9	-73.1
	-8.7	-5.9	-4.8(-6.1)	-3.4(-3.9)	-7.4	-3.8(-4.9)	-10.2	-2.4	1.7	-10.5	0.2(0.2)	-5.1(-3.9)	-1.1(-0.7)	-5.4	-3.1(-1.6)	-3.1(-3.3)	-3.5(-2.2)	-4.2(-2.1)	-5.4(-5.0)	-55.3	-24.8	-6.0	-86.1	-86.3
$\alpha_{yz}$	-1.6	-2.3	-2.5(-2.3)	-1.5(-0.6)	-2.8	-2.8(-2.5)	-3.2	-2.8	0.2	-3.0	1.1(0.5)	1.6(1.9)	-1.5(-1.1)	3.9	3.5(3.8)	2.8(2.4)	2.9(0.7)	2.0(2.5)	1.8(2.0)	-22.3	16.8	1.3	-4.1	-4.4
	-1.8	-2.5	-2.8(-2.6)	-1.4(-0.6)	-3.4	-3.0(-2.8)	-3.7	-3.2	0.3	-3.2	1.1(0.5)	1.4(1.8)	-1.6(-1.2)	3.9	3.6(3.9)	3.0(2.5)	2.9(0.8)	2.0(2.4)	1.6(1.8)	-24.9	16.9	1.0	-7.0	-7.1
$\alpha_{iso}$	13.6	8.8	12.5(10.6)	11.0(9.2)	11.5	10.0(8.3)	12.5	7.0	6.8	14.9	11.0(4.8)	10.8(8.2)	13.4(6.5)	10.1	10.7(8.7)	11.5(9.2)	11.4(8.8)	11.8(9.2)	11.5(9.3)	108.7	67.0	35.2	210.9	210.9
	14.3	9.3	12.8(11.0)	11.3(9.5)	13.2	10.2(8.6)	14.2	7.7	7.3	15.9	10.5(4.7)	11.1(8.3)	13.4(6.5)	10.2	10.7(8.6)	11.6(9.2)	11.6(9.0)	11.7(9.3)	11.5(9.4)	116.3	67.2	35.0	218.6	218.7