Electronic Supplementary Material for CrystEngComm

Molecular bricklaying II. Anion and chain length effects in bisbenzimidazolonium salts.

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Supplementary material

Details of X-ray crystal structure determinations.

2$Br_2.4H_2O$. The hydrogens of the cation were observed and refined isotropically. The hydrogens of water molecules were refined with restraints on bond lengths and bond angles.

2I_2. 4 EtOH. The hydrogens of the cation were observed and refined isotropically. The OH hydrogens of the ethanol molecules were observed and refined isotropically. The other ethanol hydrogens were calculated. The second ethanol molecule showed disorder for the central atom which was refined with two sites with occupation of 0.65 and 0.35 respectively.

2$(SO_4).5H_2O$. The N-H hydrogens were observed and refined. The hydrogens of water molecules were refined with restraints on bond lengths and bond angles. Other hydrogens were calculated.

2$[Au(CN)_2]_2$. The N-H hydrogens were observed and refined with restraints on bond distances. Other hydrogens were calculated.

4$Cl_2.2H_2O$ The hydrogens were observed and refined isotropically.

4$[CoCl_4]$.2EtOH The hydrogens of the cation and the OHb hydrogens were observed and refined with U_{iso} fixed at 0.05 $Å^2$. The two ethanol molecules are disordered. For molecule a a common position for C1 was fixed and occupation of 0.7 and 0.3 for the two sites. For molecule b a common position for the oxygen was fixed with equal populations of the two orientations. The hydrogens of the ethanol moleules were refined with restraints on bond lengths and bond angles and fixed at the end of the refinement.

5$Cl_2.2H_2O$. The hydrogens were observed and refined with U_{iso} fixed at 0.04 $Å^2$.

6Cl_2. The hydrogens were observed and refined with U_{iso} fixed at 0.04 $Å^2$.

6$SO_4.H_2O$. The hydrogens of water molecules were refined with restraints on bond lengths and bond angles. Other hydrogens were calculated.

7Cl_2.3MeOH. The hydrogens of the methyl groups and the non-disordered methanol were refined with restraints on bond lengths and bond angles and fixed at the end of the refinement. The second methanol molecule is disordered about a centre of inversion, and was refined with an occupation of 0.5, without hydrogens. Other hydrogens were calculated.

7$[CoCl_4].H_2O$. The hydrogens of the methyl groups were refined with restraints on bond lengths and bond angles and fixed at the end of the refinement. The N-H hydrogens were observed and refined with restraints on bond distances. The hydrogens of water molecules were refined with restraints on bond lengths and bond angles. Other hydrogens were calculated.

Table S1. Data for hydrogen bonds

Donor,D – H	Acceptor, A	D-H (Å)	H..A(Å)	D..A(Å)	D – H – A (°)	Symop.
2$Br_2.4H_2O$						
N1 -- H01	.. O2W	0.71(6)	1.98(6)	2.694(6)	179(10)	.
N2 -- H02	.. Br1	0.78(6)	2.57(6)	3.313(4)	159(6)	.
N3 -- H03	.. Br2	0.87(6)	2.39(6)	3.229(4)	164(5)	.
N4 -- H04	.. O1W	0.74(6)	1.97(6)	2.698(6)	169(7)	x,-1+y,z
O1W -- H11W	.. Br2	0.92(4)	2.46(5)	3.332(4)	159(4)	-1+x,y,z
O1W -- H12W	.. O3W	0.93(5)	1.84(5)	2.755(6)	170(4)	x,1+y,z
O2W -- H21W	.. O4W	0.94(4)	1.85(5)	2.754(6)	162(4)	.
O3W -- H31W	.. Br1	0.96(5)	2.47(4)	3.365(5)	155(4)	1-x,-y,1-z
O3W -- H32W	.. Br1	0.97(5)	2.36(5)	3.331(5)	178(6)	-1+x,y,z
O4W -- H41W	.. Br2	0.94(4)	2.43(4)	3.368(5)	179(5)	-1+x,y,z
O4W -- H42W	.. Br2	0.94(5)	2.54(4)	3.425(5)	156(4)	1-x,1-y,-z
2I_2. 4 EtOH						
N1 -- H01	.. O1B	0.80(6)	1.91(6)	2.697(6)	167(6)	.
N2 -- H02	.. O1A	0.82(5)	1.88(4)	2.696(5)	171(5)	2-x,1-y,1-z
O1A -- H01A	.. I	0.97(4)	2.52(4)	3.481(4)	171(4)	.
O1B -- H01B	.. I	0.98(3)	2.50(3)	3.465(5)	170(3)	.
2$(SO_4).5H_2O$						
N1A -- H01A	.. O3	0.90(3)	1.86(3)	2.754(3)	169(3)	.
N2A -- H02A	.. O1W	0.93(3)	1.80(3)	2.728(3)	176(3)	-x,-y,-z
N1B -- H01B	.. O2W	0.87(3)	1.84(3)	2.696(3)	168(3)	1-x,-y,1-z
N2B -- H02B	.. O1	0.90(3)	1.81(3)	2.705(3)	175(3)	.
O1W -- H11W	.. O3W	0.85(3)	1.89(3)	2.679(4)	153(3)	-1+x,y,z
O1W -- H12W	.. O2	0.85(2)	1.91(2)	2.752(3)	171(3)	.
O2W -- H21W	.. O2	0.86(3)	2.02(3)	2.849(3)	160(3)	.
O2W -- H22W	.. O5W	0.85(2)	1.83(2)	2.671(4)	172(2)	x,-1+y,z
O3W -- H31W	.. O3	0.81(2)	2.04(2)	2.816(4)	161(2)	1+x,y,z
O3W -- H32W	.. O4	0.84(3)	1.86(3)	2.690(4)	173(3)	.
O4W -- H41W	.. O1W	0.83(3)	2.08(3)	2.874(3)	159(3)	1+x,1+y,z
O4W -- H42W	.. O1	0.84(3)	2.00(3)	2.814(4)	163(3)	.
O5W -- H51W	.. O3	0.82(3)	2.18(3)	2.925(4)	151(3)	
O5W -- H52W	.. O4W	0.80(3)	1.98(3)	2.751(5)	162(3)	-1+x,y,z
C2B -- H2B	.. O3W	0.98	2.56	3.510(4)	164	1-x,-y,1-z
C5A -- H5A	.. O4	0.98	2.46	3.159(4)	128	1-x,1-y,-z
C5B -- H5B	.. O2	0.98	2.47	3.283(3)	140	.
2$[Au(CN)_2]_2$						
N1 -- H01	.. N01	0.94(6)	1.86(5)	2.798(7)	173(6)	1+x, y, z
N2 -- H02	.. N02	0.92(6)	1.92(6)	2.778(7)	153(6)	-1+x, -1+y, z
4$Cl_2.2H_2O$						
N1 -- H01	.. Cl	0.87(3)	2.29(3)	3.1250(17)	162(2)	1-x,-1-y,1-z
N2 -- H02	.. O1W	0.90(3)	1.90(3)	2.754(2)	159(3)	.
O1W -- H11W	.. Cl	0.80(3)	2.38(3)	3.1752(18)	173(3)	x,y,1+z
O1W -- H12W	.. Cl	0.80(3)	2.37(3)	3.170(2)	175(3)	-x,-y,1-z
4$[CoCl_4]$.2EtOH						
N1 -- H01	.. Cl2	0.82(4)	2.33(4)	3.145(3)	179(6)	.
N2 -- H02	.. O1A	0.76(4)	2.06(4)	2.796(6)	164(5)	x,3/2-y,-1/2+z
N3 -- H03	.. Cl3	0.86(4)	2.71(4)	3.332(3)	130(3)	-x,1/2+y,3/2-z
N3 -- H03	.. Cl4	0.86(4)	2.71(4)	3.421(3)	141(4)	-x,1/2+y,3/2-z
N4 -- H04	.. O1B	0.85(4)	1.85(4)	2.692(4)	173(5)	x,3/2-y,-1/2+z
O1B -- H01B	.. Cl4	0.79(4)	2.37(4)	3.148(4)	169(4)	-x,1/2+y,3/2-z
C1B -- H12B	.. Cl3	0.98	2.68	3.647(10)	169	.
C8 -- H82	.. Cl5	0.97(4)	2.70(4)	3.664(5)	173(3)	.

5$Cl_2.2H_2O$

N1 -- H01 .. O1W	0.82(3)	1.93(3)	2.754(2)	175(3)	.
O1W -- H1W .. Cl	0.85(3)	2.37(3)	3.2048(19)	170(3)	1-x,y,3/2-z
N2 -- H02 .. Cl	0.90(2)	2.18(2)	3.0662(18)	171.8(19)	-1/2+x,1/2+y,z
O1W -- H2W .. Cl	0.80(3)	2.43(3)	3.2190(19)	172(3)	.

6Cl_2

N1 -- H01 .. Cl	0.79(2)	2.39(2)	3.113(2)	153(2)	1/2+x,1/2-y,1-z
N2 -- H02 .. Cl	0.77(3)	2.31(3)	3.068(2)	173(3)	1/2+x,y,1/2-z

6$SO_4.H_2O$

N1 -- H01 .. O4	0.98	1.74	2.692(3)	164	3/2-x,1/2+y,3/2-z
N2 -- H02 .. O1	0.98	2.59	3.316(3)	131	.
N2 -- H02 .. O2	0.98	1.74	2.698(3)	166	.
N3 -- H03 .. O1	0.98	1.70	2.670(3)	170	-1/2+x,3/2-y,-1/2+z
N4 -- H04 .. O3	0.98	1.74	2.684(3)	162	2-x,1-y,1-z
O1W -- H01W .. O4	1.00(2)	1.82(2)	2.811(4)	170(2)	.
O1W -- H02W .. O1	0.996(17)	2.38(2)	3.114(5)	130(3)	2-x,1-y,1-z
O1W -- H02W .. O4	0.996(17)	2.12(2)	3.081(4)	162(3)	2-x,1-y,1-z
C16 -- H16 .. O1W	0.98	2.41	3.274(5)	146	3/2-x,1/2+y,1/2-z
C8 -- H82 .. O1	0.98	2.58	3.234(4)	124	.
C13 -- H132 .. O1W	0.98	2.27	3.092(6)	141	2-x,1-y,1-z

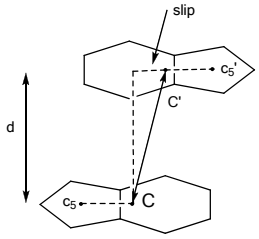
7$Cl_2.3MeOH$

N2 -- H02 .. O101	0.98	1.77	2.729(3)	166	1-x,1-y,1-z
O101 -- H0101 .. Cl	0.95	2.08	3.015(2)	166	x,3/2-y,-1/2+z
C11 -- H111 .. Cl	0.98	2.74	3.688(4)	165	1-x,1/2+y,3/2-z
C11 -- H112 .. O201	0.98	2.49	3.264(7)	136	1-x,2-y,1-z

7$[CoCl_4].H_2O$

N2A -- H02A .. Cl1	0.94(2)	2.26(2)	3.139(2)	156(2)	1-x,1-y,1-z
N2B -- H02B .. O1W	0.93(3)	1.83(3)	2.755(3)	176(2)	1-x,1-y,1-z
O1W -- H12W .. Cl3	0.86(3)	2.34(3)	3.198(3)	174(2)	1-x,-y,1-z
C8B -- H81B .. Cl2	0.98	2.77	3.697(3)	158	1-x,-y,1-z
C8A -- H82A .. Cl4	0.98	2.78	3.705(3)	158	x,1+y,-1+z

Table S2. Details of stacking interactions.



d - interplane distance

C, C' centroids of benzimidazole

c_5, c_5' centroids of 5-membered rings

The torsion is defined as the angle c_5 - C -C' -c_5'

	Interplane distance, d (Å)	Interplane angle (°)	C-C' (Å)	Slip (Å)	torsion (°)
2$Br_2.4H_2O$					
	3.433	0	3.550	0.904	180
	3.350	0	3.648	1.444	180
	3.442	0	3.540	0.827	180
	3.419	0	3.631	1.223	180
2I_2.4EtOH					
	3.333	0.94	3.708	1.625	145
2$(SO_4).5H_2O$					
	3.456	0	3.659	1.202	180
	3.423	0	3.607	1.137	180
	3.454	0	3.667	1.232	180
	3.347	0	3.643	1.438	180
4$Cl_2.2H_2O$					
	3.482	0	3.656	1.114	180
	3.352	0	3.889	1.972	180
4$(CoCl_4)$.2EtOH					
	3.367	0	4.450	2.910	180
	3.500	5.75	3.621	0.928	180
	3.400	0	3.513	0.884	180
	3.560	5.75	3.621	0.662	180
5$Cl_2.2H_2O$					
	3.392	0	3.602	1.212	180
	3.33	2.79	3.485	1.028	180
6$(SO_4).H_2O$					
	3.363	4.96	3.404	0.527	167
	3.421	0	3.565	1.003	180
7$(CoCl_4).H_2O$					
	3.274	0	3.866	2.056	180

The interplane distance for stacks where the planes are not parallel is taken as the average value of the distances of atoms of one benzimidazole to the least squares plane through the other

Thermal ellipsoid plots of crystal structures showing atomic numbering. All ellipsoids are drawn at 50% probability.

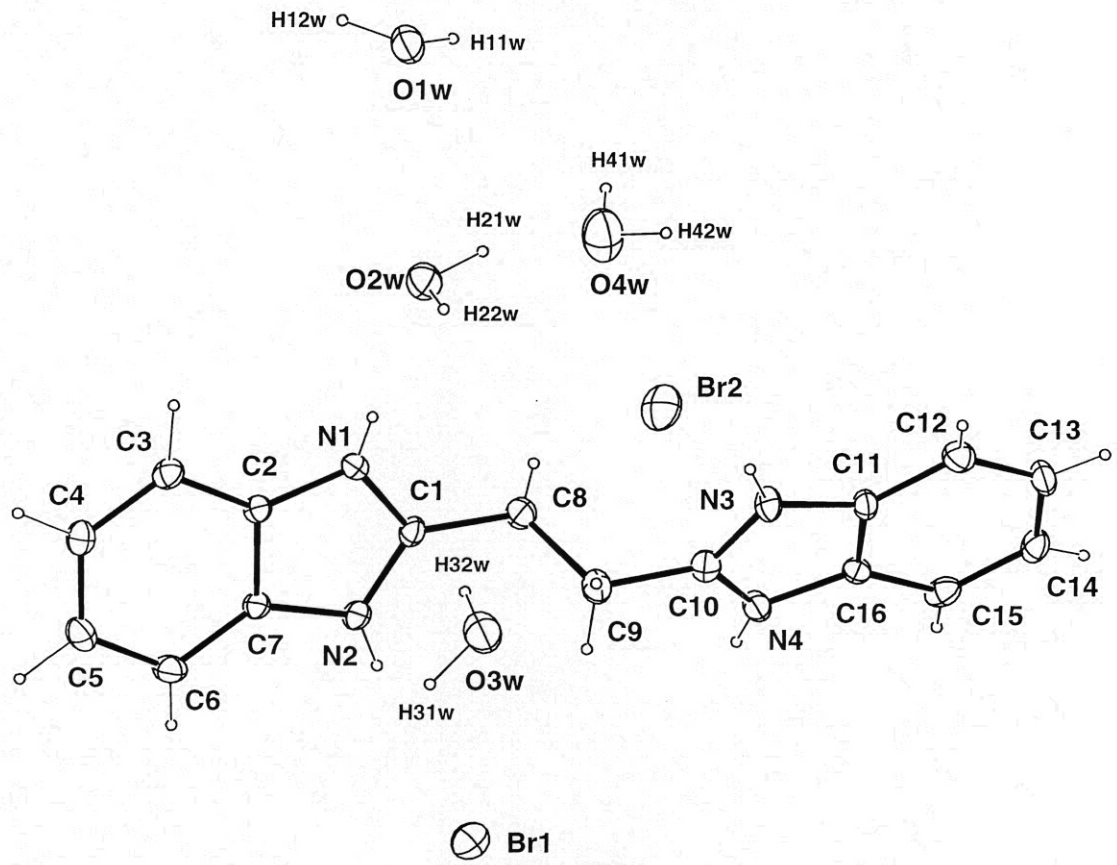


Fig. S1 **2**$Br_2.4H_2O$

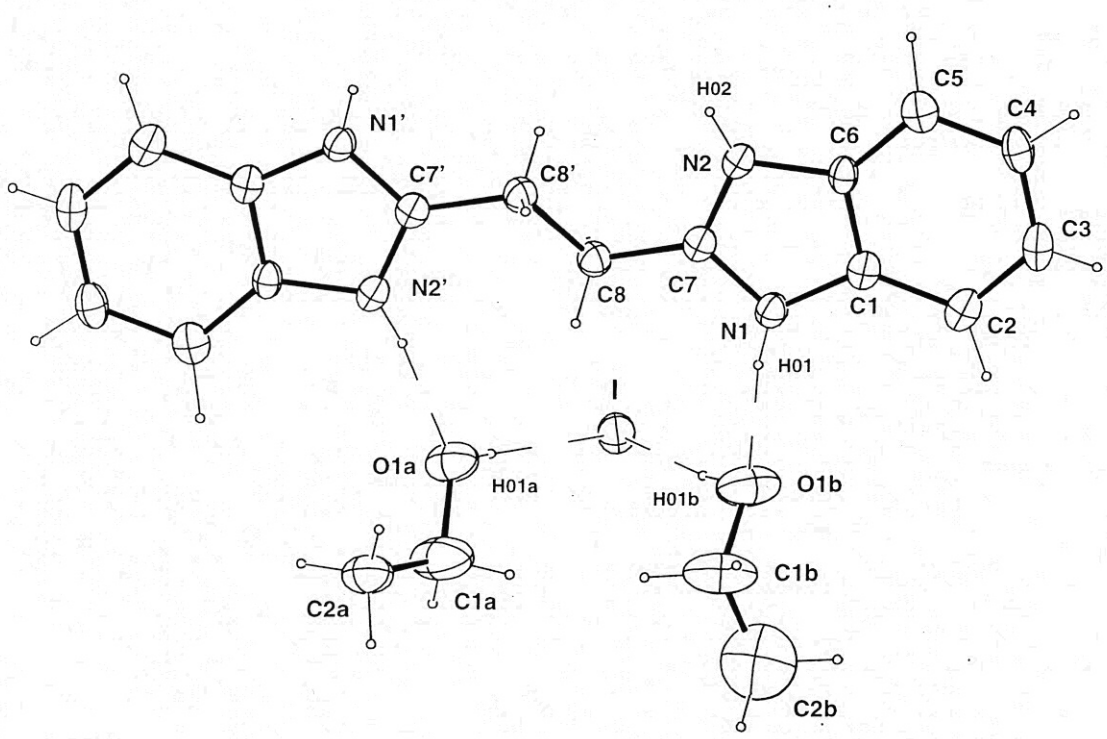


Fig. S2 **2**I_2. 4 EtOH. The primed and unprimed atoms are related by a centre of inversion between C8 and C8'

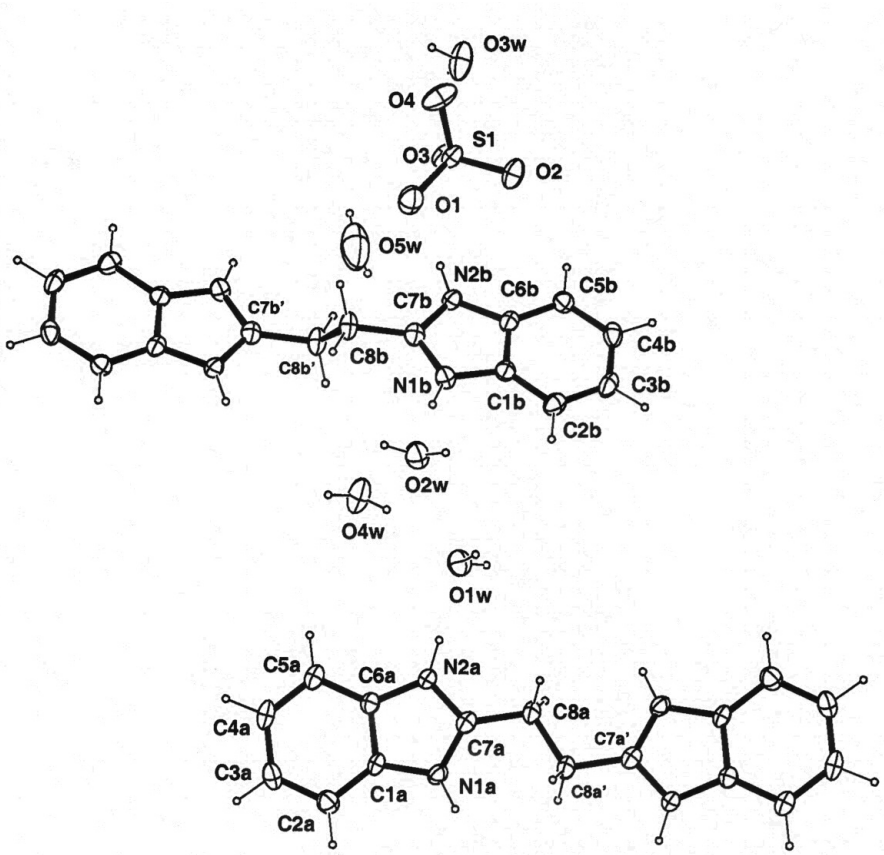


Fig. S3 **2**(SO_4).5H_2O. The primed and unprimed atoms are related by a centre of inversion between C8 and C8'

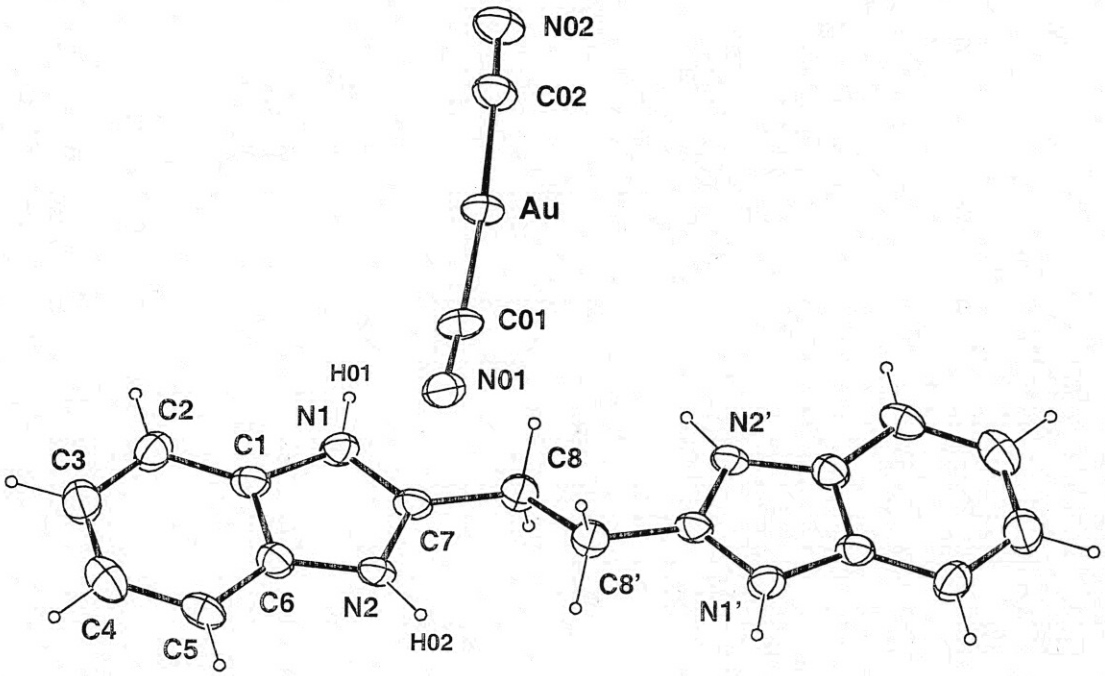


Fig. S4 **2**$[Au(CN)_2]_2$. The primed and unprimed atoms are related by a centre of inversion between C8 and C8'

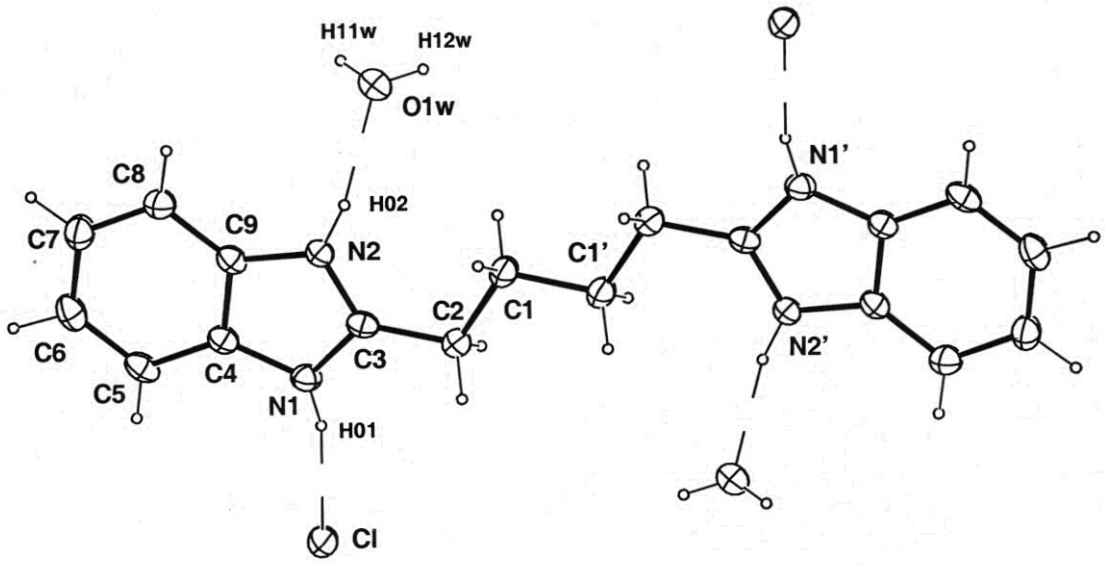


Fig. S5 **4**$Cl_2.2H_2O$. The primed and unprimed atoms are related by a centre of inversion between C1 and C1'

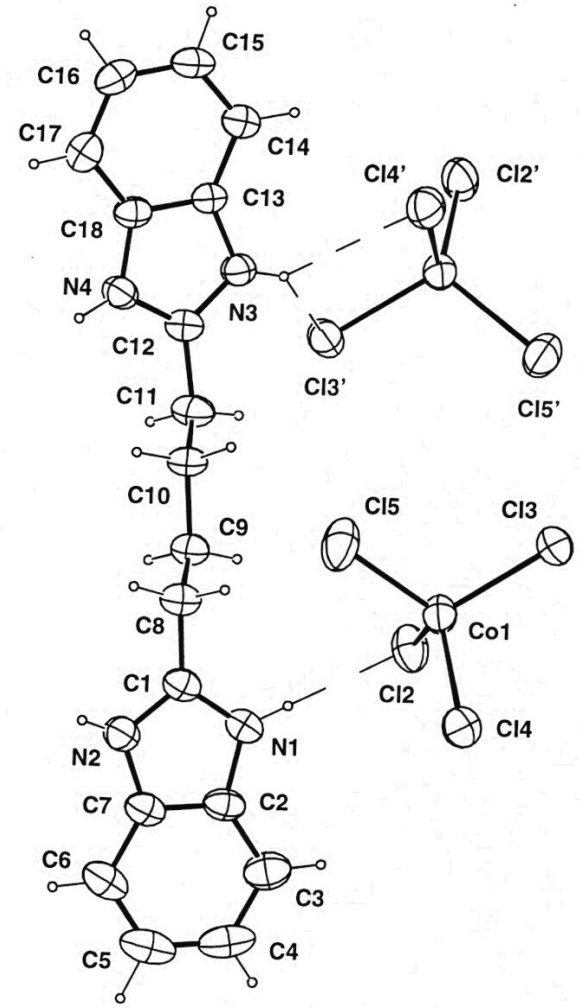


Fig. S6 **4**[$CoCl_4$].2EtOH. The primed and unprimed atoms are related by the operation –x, ½ + y, 3/2-z

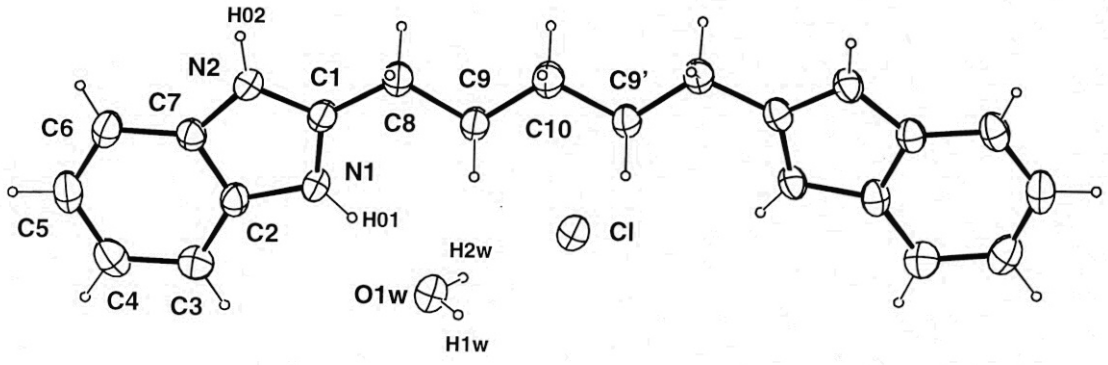


Fig. S7 **5**Cl_2.2H_2O. The primed and unprimed atoms are related by a twofold axis passing through C10.

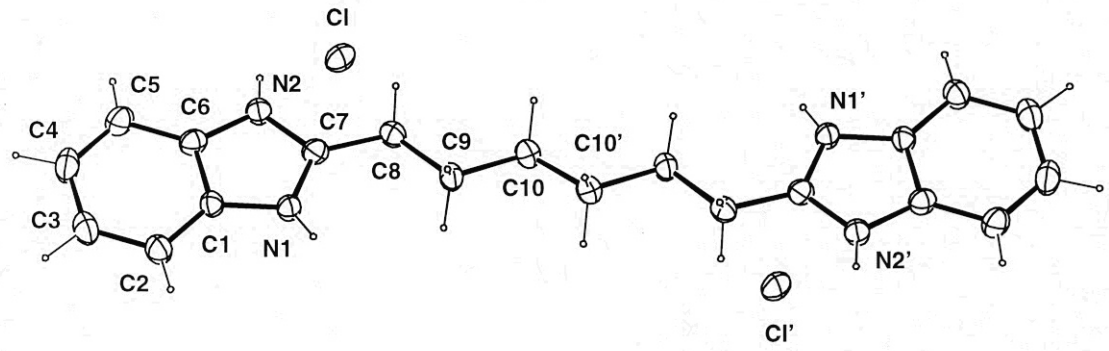


Fig. S8 **6**Cl_2. The primed and unprimed atoms are related by a centre of inversion between C10 and C10'

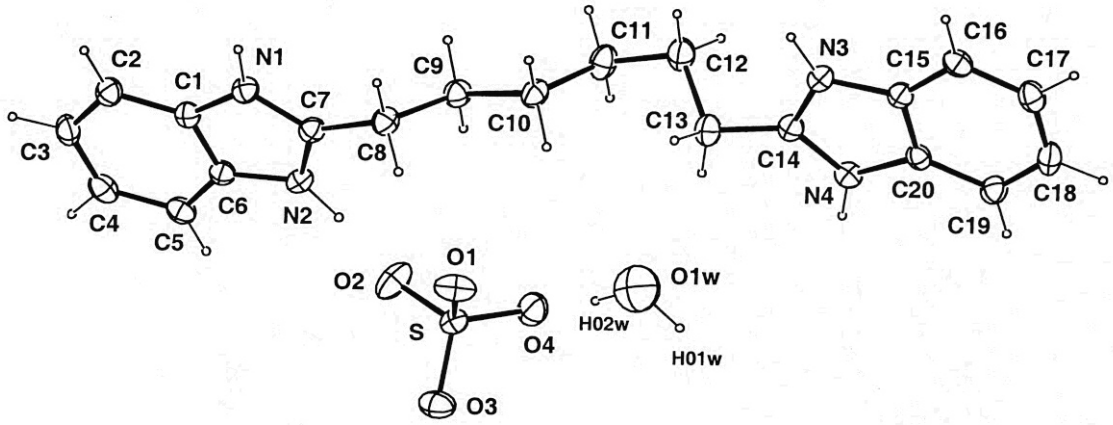


Fig. S9 **6**$SO_4.H_2O$

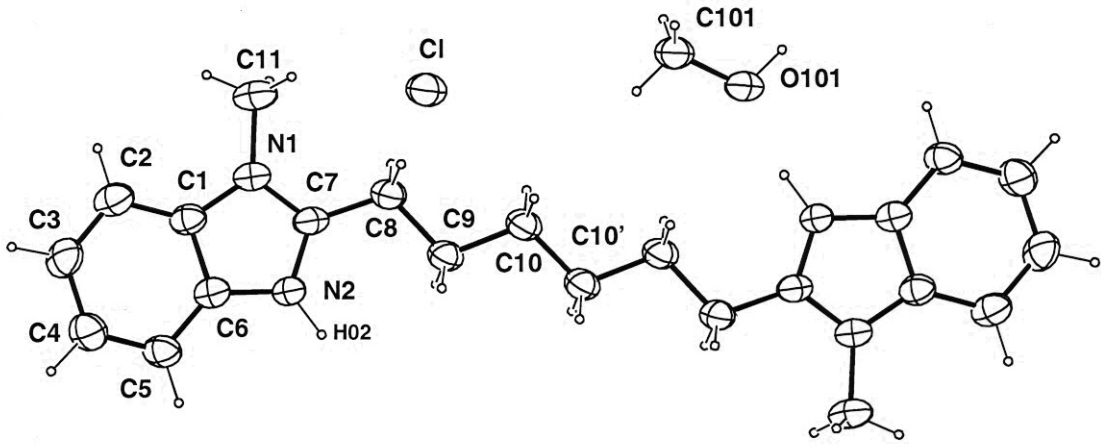


Fig. S10 **7**Cl_2.3MeOH. The primed and unprimed atoms are related by a centre of inversion between C10 and C10'

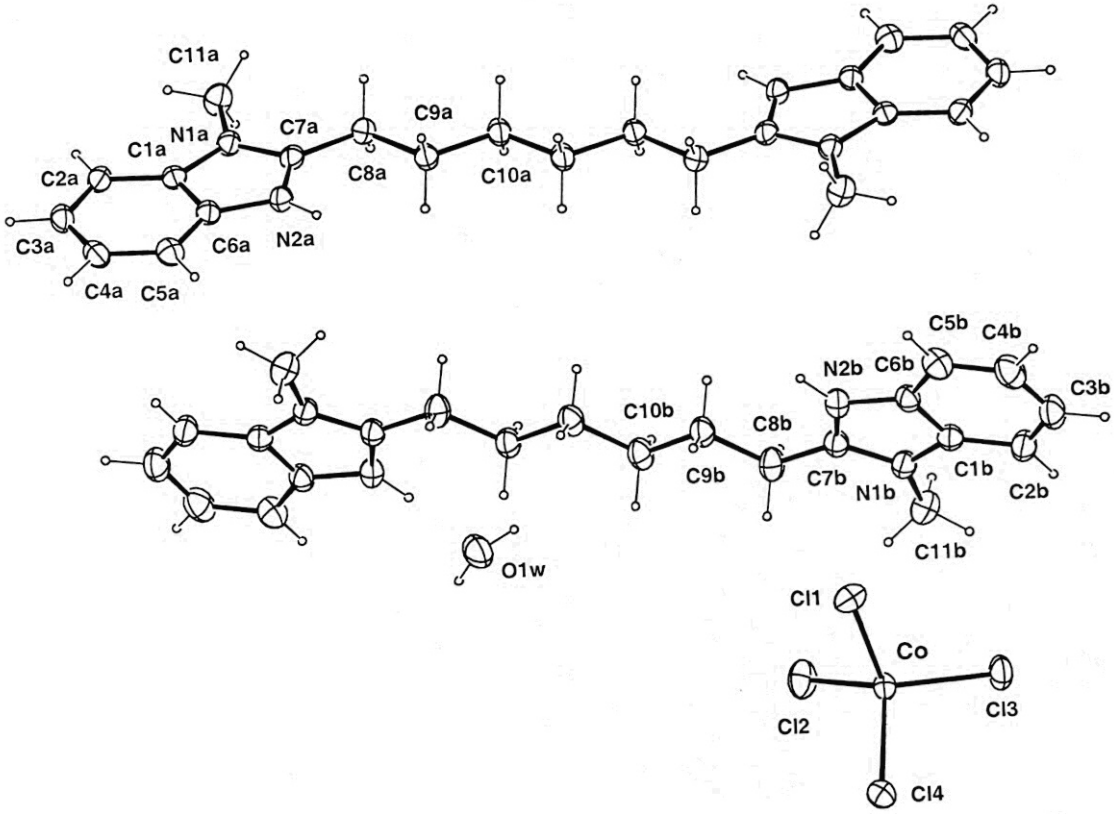


Fig. S11 **7**[$CoCl_4$].H_2O. The two halves of each cation are related by a centre of inversion.