Exohedral functionalization of fullerene by substituents controlling of molecular organization for spontaneous C_{60} dimerization in liquid crystal solutions and in a bulk controlled by a potential

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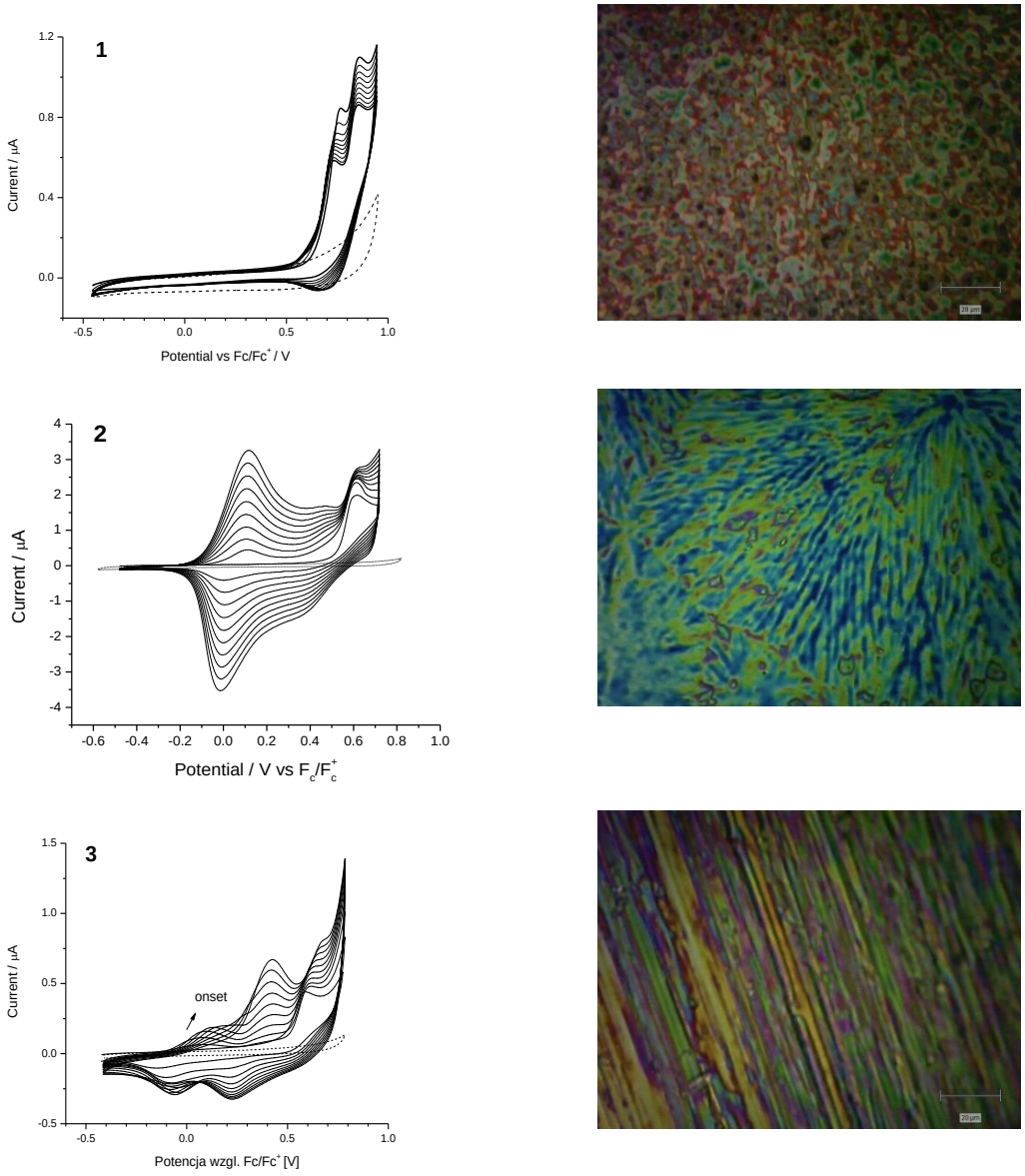


Figure S1. Voltammograms of oxidative electropolymerization of **1**, **2** and **3** in precursor solution (1 mM) in E1/dichloromethane (0.1 M) to produce polymers **p(1)**, **p(2)** and **p(3)** (left) and images of these polymers surfaces obtained using an optical Raman microscope under laser excitation of 514 nm (right).

Table S1. Shape of frontier orbitals of **2** calculated at B3LYP/6-31G(d,p).
The isovalue is equal to 0.03 e^-/au^3 in each case.

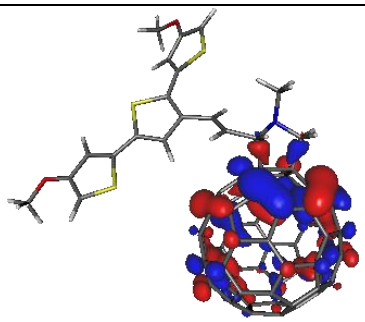
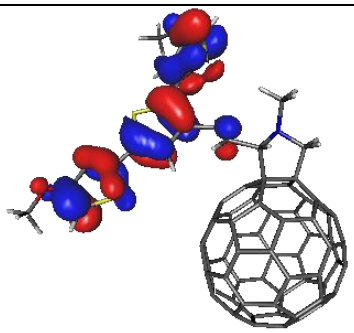
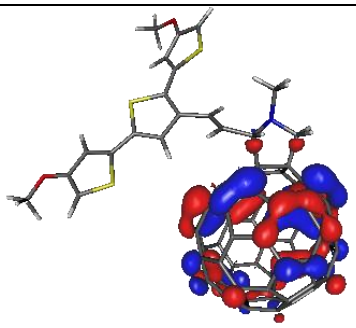
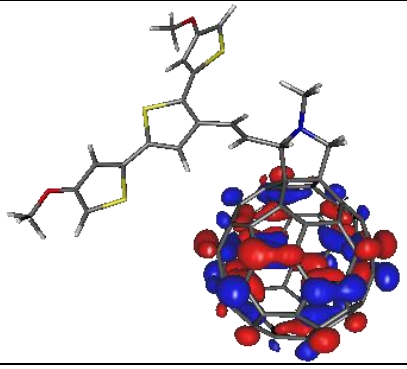
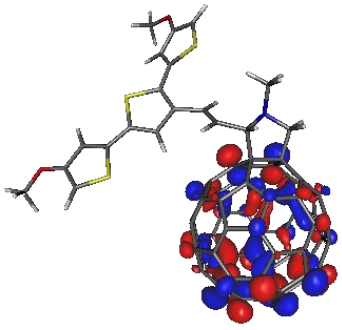
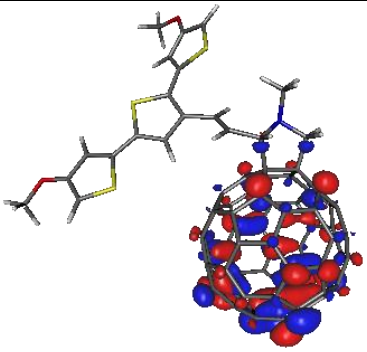
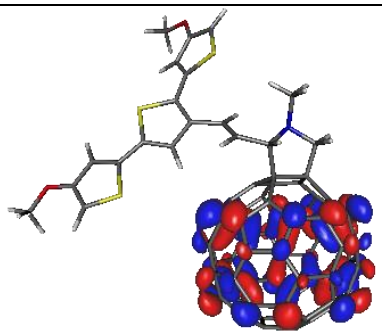
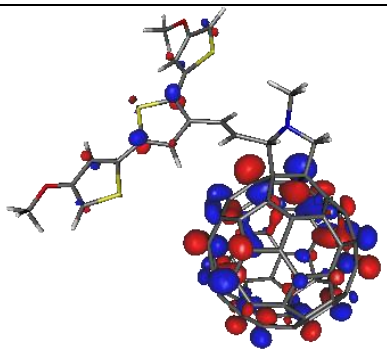
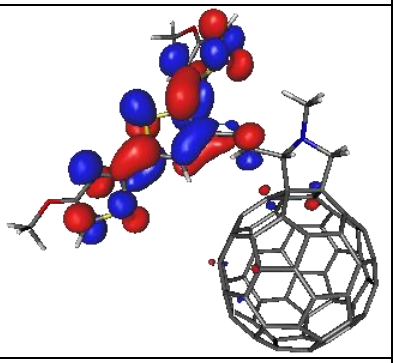
HOMO-1	**HOMO**	**LUMO**
		
-5.32	**-5.15**	**-3.33**
LUMO+1	**LUMO+2**	**LUMO+3**
		
-2.99	**-2.77**	**-2.20**
LUMO+4	**LUMO+5**	**LUMO+6**
		
-1.81	**-1.59**	**-1.56**

Table S2. Shape of frontier orbitals of **2 (without alkoxy group)** calculated at B3LYP/6-31G(d,p). The isovalue is equal to 0.03 e^-/au^3 in each case.

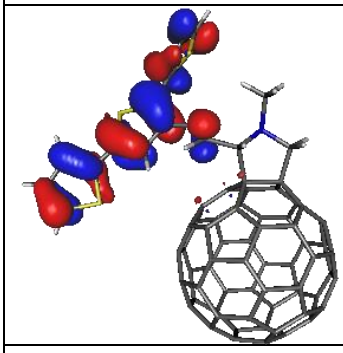
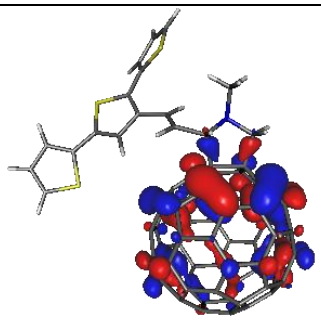
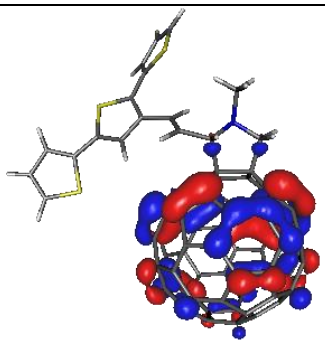
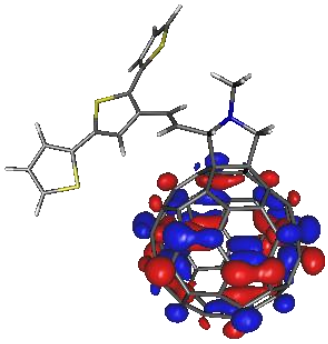
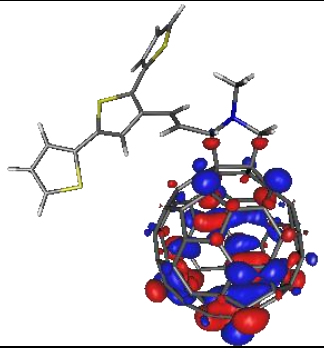
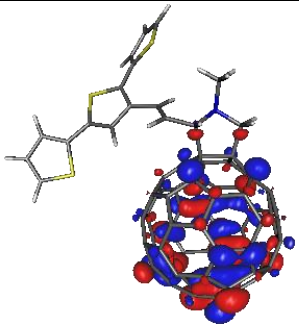
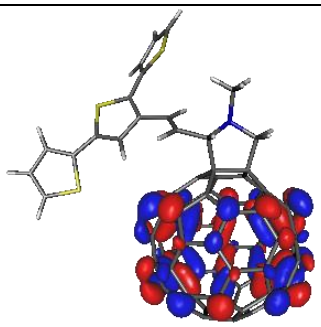
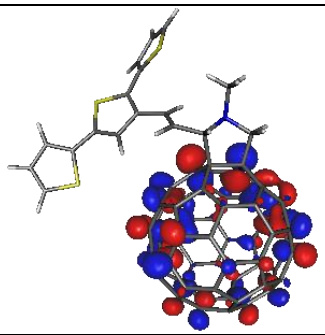
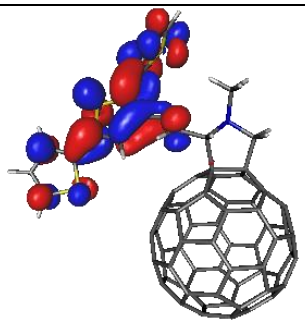
HOMO-1	**HOMO**	**LUMO**
		
-5.38	**-5.32**	**-3.33**
LUMO+1	**LUMO+2**	**LUMO+3**
		
-3.00	**-2.77**	**-2.20**
LUMO+4	**LUMO+5**	**LUMO+6**
		
-1.81	**-1.59**	**-1.50**

Table S3. Shape of frontier orbitals of **3** calculated at B3LYP/6-31G(d,p).
The isovalue is equal to 0.03 e^-/au^3 in each case.

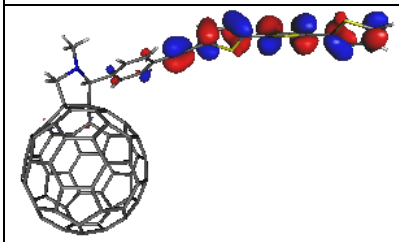
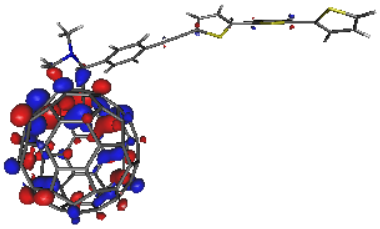
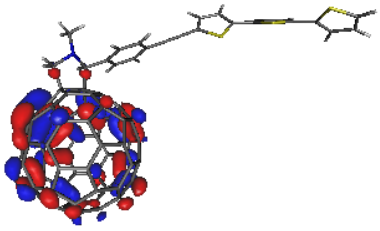
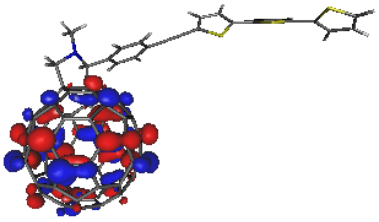
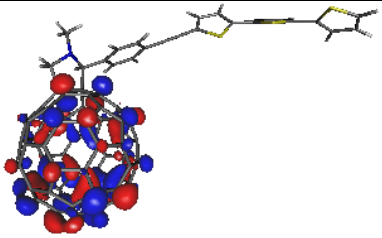
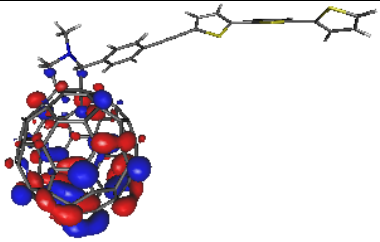
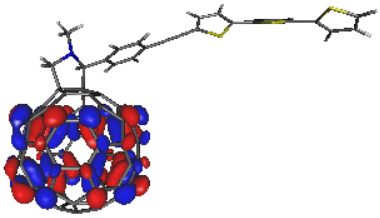
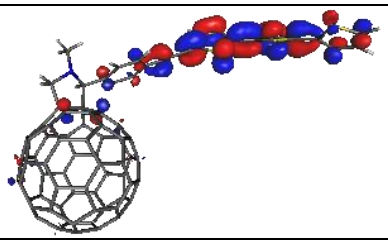
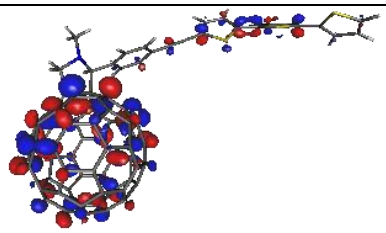
HOMO-1	**HOMO**	**LUMO**
		
-5.36	**-5.34**	**-3.35**
LUMO+1	**LUMO+2**	**LUMO+3**
		
-3.01	**-2.79**	**-2.22**
LUMO+4	**LUMO+5**	**LUMO+6**
		
-1.82	**-1.63**	**-1.61**

Table S4. Spin density of reduced form of monomers calculated at B3LYP/6-31G(d), isovalue is equal to 0.005 e-/au^3.

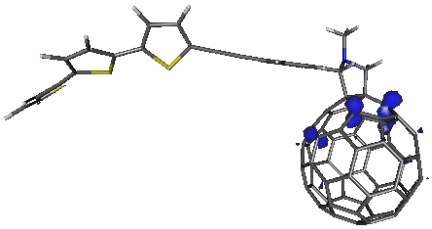
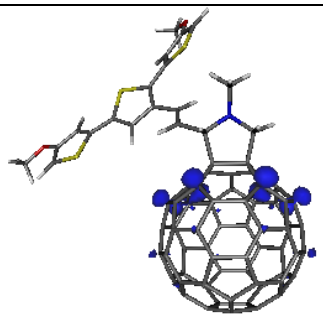
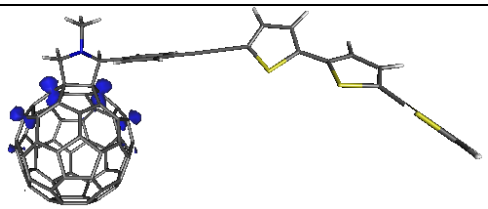
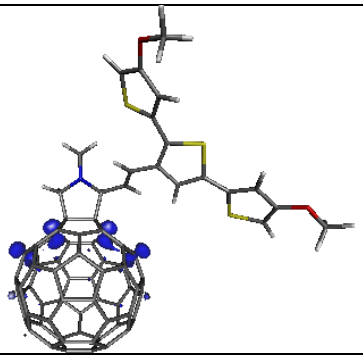
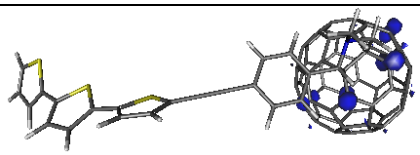
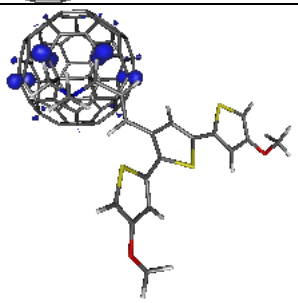
2	3
	
	
	

Table S5. Shape of frontier orbitals of **(2)**$_2$ dimer in neutral state, calculated at B3LYP/6-31G(d,p). The isovalue is equal to 0.03 e^-/au^3 in each case.

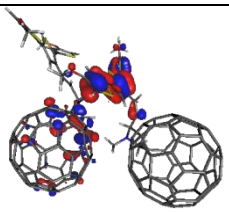
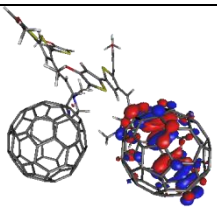
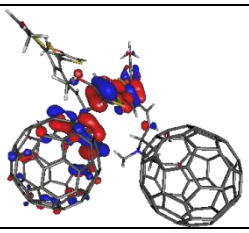
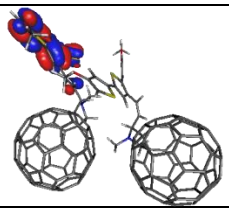
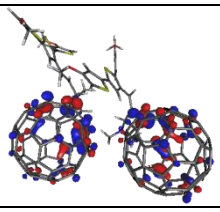
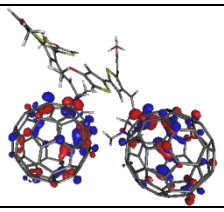
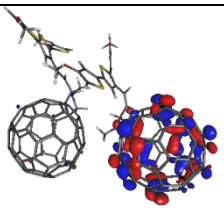
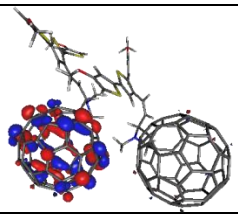
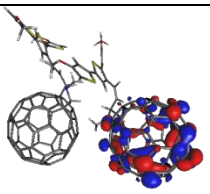
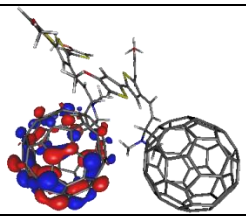
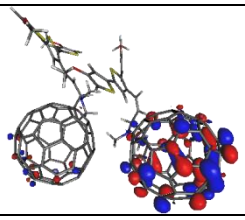
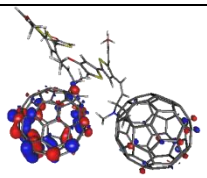
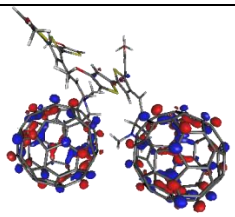
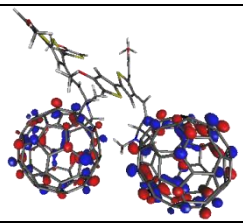
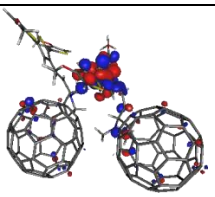
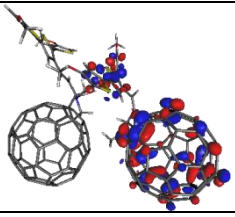
HOMO-3	HOMO-2	HOMO-1
		
-5.37	-5.31	-5.24
HOMO	**LUMO**	**LUMO+1**
		
-4.99	-3.33	-3.30
LUMO+2	**LUMO+3**	**LUMO+4**
		
-3.04	-3.00	-2.81
LUMO+5	**LUMO+6**	**LUMO+7**
		
-2.78	-2.21	-2.19
LUMO+8	**LUMO+9**	**LUMO+10**
		
-1.84	-1.81	-1.73
LUMO+11		
		
-1.62		

Table S6. Shape of frontier orbitals of **(3)**$_2$ dimer in neutral state, calculated at B3LYP/6-31G(d,p). The isovalue is equal to 0.03 e^-/au^3 in each case.

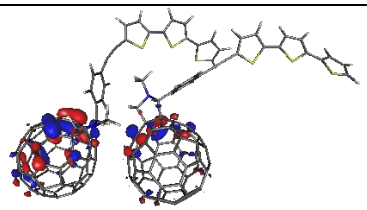
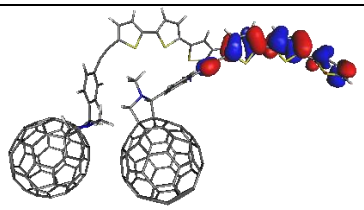
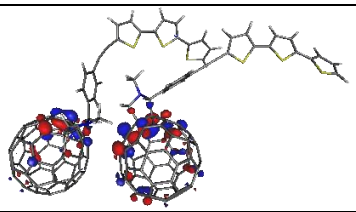
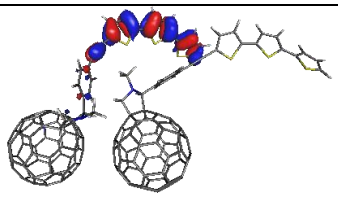
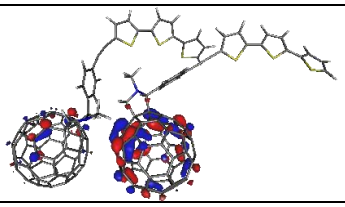
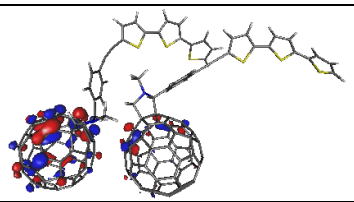
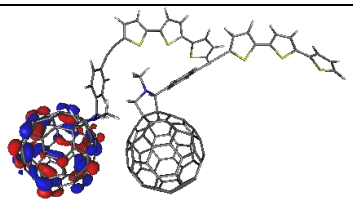
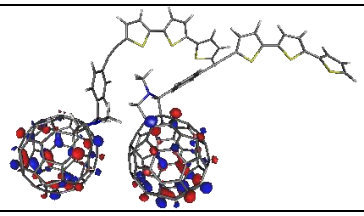
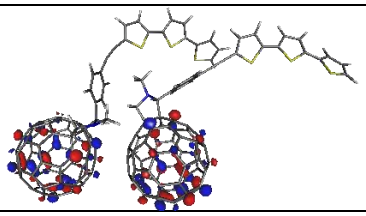
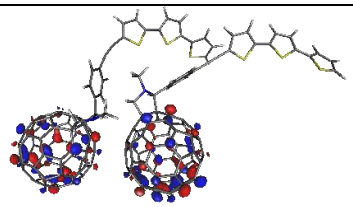
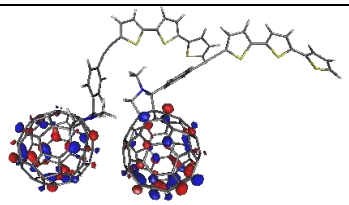
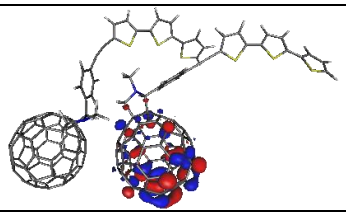
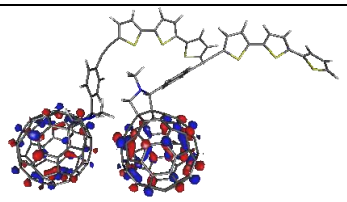
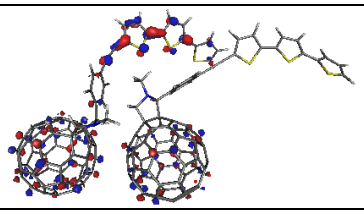
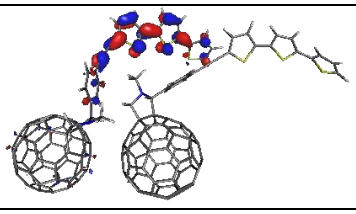
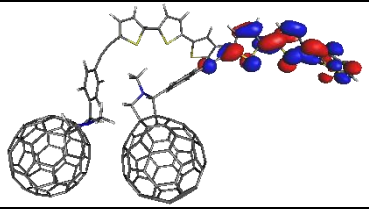
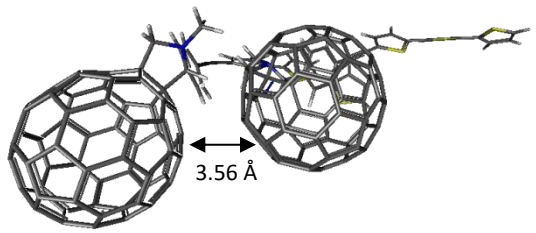
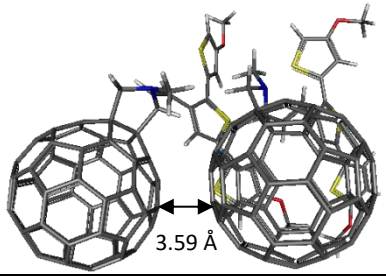
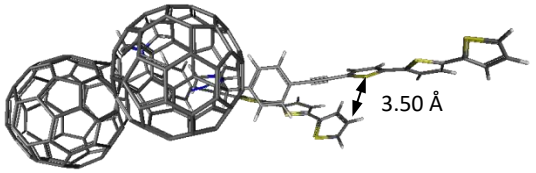
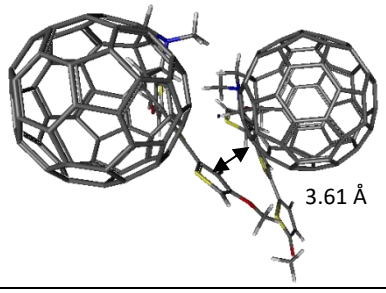
HOMO-3	**HOMO-2**	**HOMO-1**
		
-5.35	**-5.31**	**-5.30**
HOMO	**LUMO**	**LUMO+1**
		
-5.26	**-3.37**	**-3.37**
LUMO+2	**LUMO+3**	**LUMO+4**
		
-3.03	**-3.02**	**-2.84**
LUMO+5	**LUMO+6**	**LUMO+7**
		
-2.78	**-2.24**	**-2.23**
LUMO+8	**LUMO+9**	**LUMO+10**
		
-1.85	**-1.83**	**-1.82**
LUMO+11		
		
-1.73		

Table S7. Optimized structures of **(2)**$_2$ and **(3)**$_2$ dimers in neutal state, calculated at ωB97X-D functional combined with 6-31G(d,p) basis set.

(3)$_2$	**(2)**$_2$
	
	

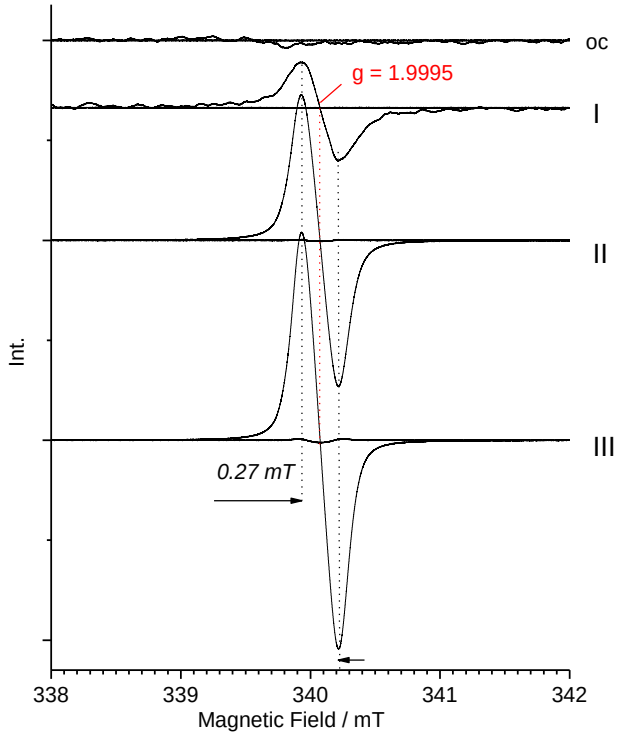


Figure S2. The change in ESR signal at I (first), II (second) and third (III) reduction peak in C_{60} solution in E1/DCM, 0.6 mT (modulation width); 200 (amplitude) (oc – open cell, without polarization).

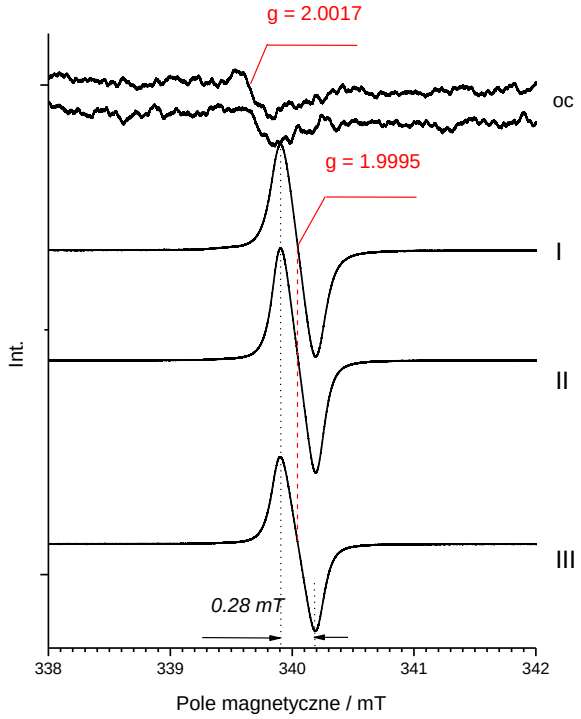


Figure S3. The change in ESR signal at I (first), II (second) and third (III) reduction peak in **1** solution in E1/DCM, 0.6 mT (modulation width); 200 (amplitude) (oc – open cell, without polarization).

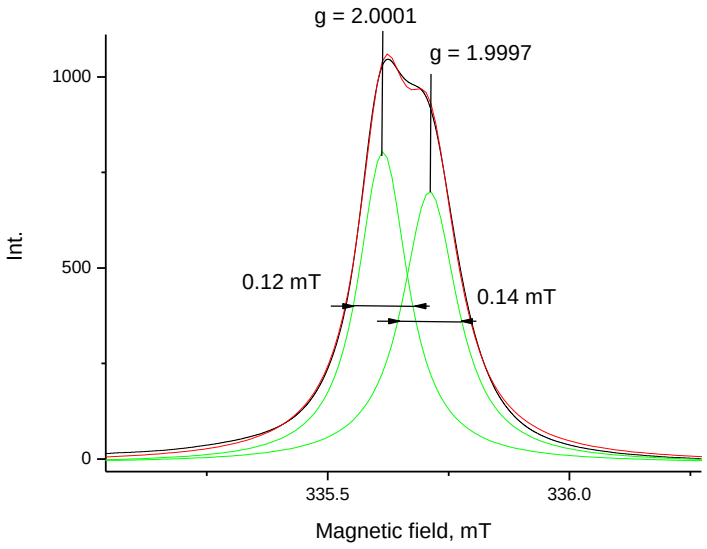


Figure S4. Graphical method showing the decomposition of absorption ESR signal into two components under potential of B peak in **2** solution in E1/DCM, 0.6 mT (modulation width); 200 (amplitude).