

SUPPORTING INFORMATION

Multidecker Sandwich Complexes $V_n\text{Ben}_{n+1}$ ($n = 1, 2, 3$) as Stronger Electron Donor Relative to Ferrocene for Designing High-Performance Organometallic Second-Order NLO Chromophores: Evident Layer Effect on the First Hyperpolarizability and Two-Dimensional NLO Character

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TABLE S1. Bond dissociation energies BDE obtained with different theory methods for dissociation of FeCp₂ (ferrocene) into Fe and two Cp ligands.

FeCp ₂ →Fe(3d ⁶ 4s ²) ⁵ D+2Cp	D_e	D_o	Exp.
CAM-B3LYP	137.3	131.5	
B3LYP	130.3	124.8	
LC-BLYP	156.7	150.5	
			158±2 ^a

D_e is the bond dissociation energies BDE without ZPE corrections, D_o is the bond dissociation energies BDE with ZPE corrections. FeCp₂ (ferrocene) has an equilibrium geometry with D_{5h} symmetry, neutral fragments Fe has a (3d⁶4s²) ⁵D ground state. The bond dissociation energies were obtained at the different calculation level with the lanl2dz basis set including a corrected effective core potential (ECP) to take into account relativistic effects for Fe and 6-31G* basis set for C and H. ^a Ref 50.

TABLE S2. AEF (Applied Electric Field, a.u.) dependence of the first hyperpolarizability β_0 at the ROCAM-B3LYP/gen level of second-order NLO organometallic complex V₂Bz₃-mid-(C₂H₂)₃-NO₂.

Fields (a.u.)	β_0
0.0005	10795
0.0007	10820
0.0010	10857
0.0012	10887
0.0015	10939

