Electronic Supplementary Information

Computational design of donor-bridge-acceptor systems exhibiting pronounced quantum interference effects

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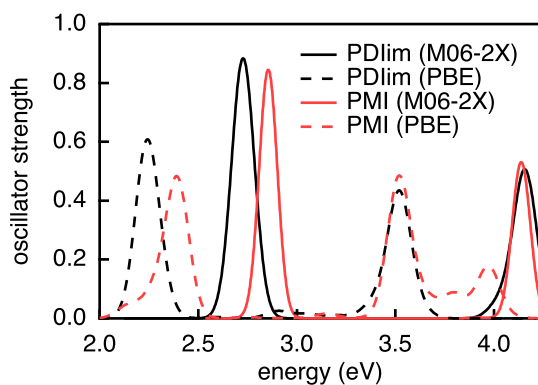[2] *Present address: Novaled GmbH, Dresden, Germany*

Figure 1: Comparison of optical excitation spectra of **PDIim** and **PMI** using TD-DFT with DZP basis set and M06-2X functional (solid) and PBE functional (dashed).

Table 1: Contribution of the HOFO of the hole donor to the initial state with the direct coupling $V_{DB_{\mathrm{HOFO}}}$ between donor and bridge

	$V_{DB_{\mathrm{HOFO}}}$	HOFO contribution
PDIim (M06-2X)	0.00 eV	100 %
PDIim (PBE)	0.00 eV	83 %
PMI (M06-2X)	-0.11 eV	94 %
PMI (PBE)	0.08 eV	67 %

Table 2: Contribution of the HOFO of the hole acceptor to the final state with the effective electronic couplings for hole transfer J_{eff}. Note, that a coupling of 1 meV is regarded as the limit of accuracy that can be obtained with DFT.

	HOFO contribution	J_{eff}
SNS (M06-2X)	98 %	0.1 meV
SNS (PBE)	98 %	-1.6 meV
lin_carbeth (M06-2X)	83 %	39 meV
lin_carbeth (PBE)	82 %	65 meV
cross_carbeth (M06-2X)	88 %	-3.8 meV
cross_carbeth (PBE)	86 %	2.8 meV