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Electronic Supplementary Information

Acceptor-donor-acceptor type molecules for high performance organic photovoltaics – chemistry and mechanism

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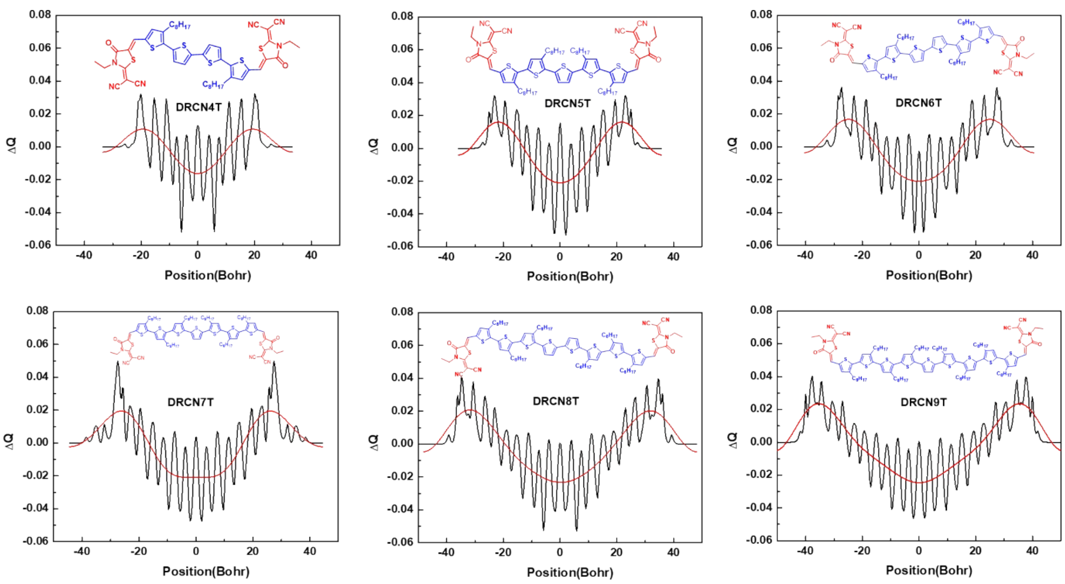
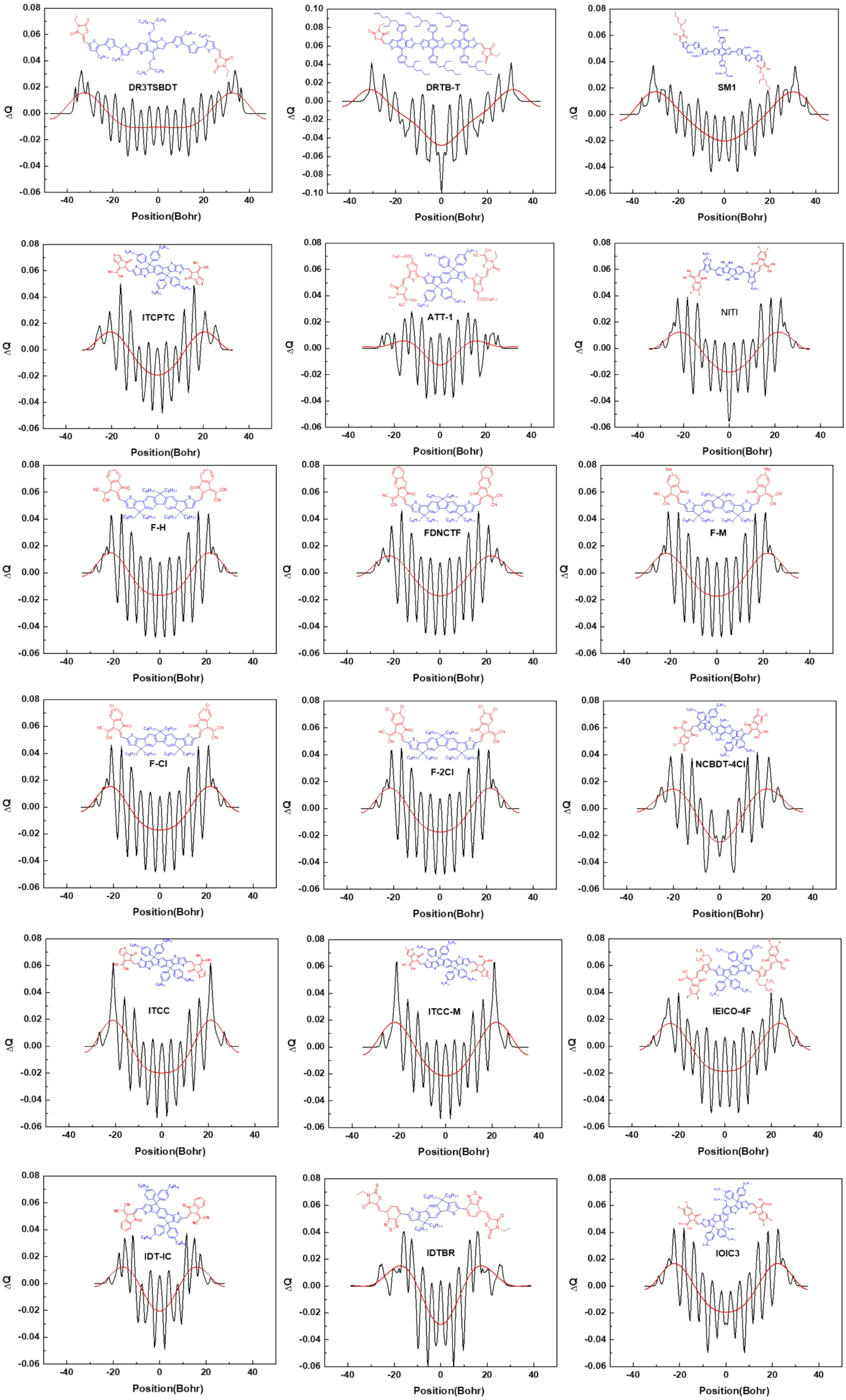
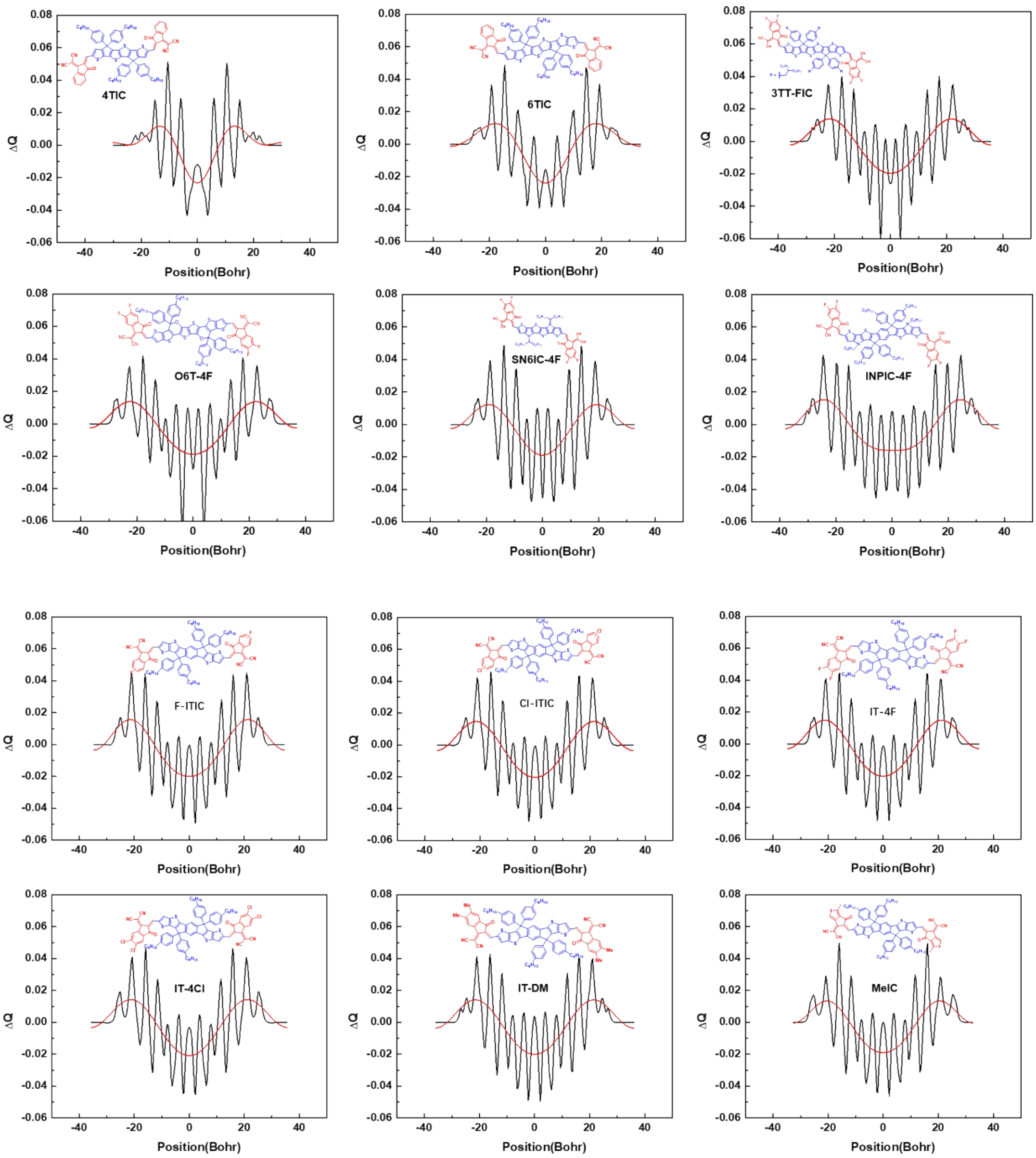
1. HOMO/LUMO calculation and analysis for some A-D-A molecules





DR3TSBDT
DRTB-T
SM1
ITCPTC
ATT-1
NITI
F-H
FDNCTF
F-M
F-Cl
F-2Cl
NCBDT-4Cl
ITCC
ITCC-M
IEICO-4F
IDT-IC
IDTBR
IOIC3
ΔQ
Position(Bohr)



2. A semi-empirical model analysis for PCE prediction

2.1 For the semi-expirical analysis based on a single cell, the fundamental assumptions are made as follows:

1) An internal quantum efficiency (IQE) of 100% is considered for the whole absorption wavelengths.[1]

2) EQE is assumed to be same in the whole absorption range with a given value of 80% and *FF* is assumed to be 0.8.

3) In the discussion as follows, the optical gap of the subcell E_g (= $1240/\lambda_{onset}$) is defined as the narrower optical gaps of the donor-acceptor couples . Note, for the

fullerene based devices, the absorption onset is considered to be that of the donor materials. For the fullerene-free based devices, it referred to be that of material, either donor or acceptor, whichever has a narrower bandgap.[2]

Based on above assumptions, the three photovoltaic parameters V_{oc}, J_{sc} and FF of a single cell is obtained as follows:

1) For a single cell with absorption onset λ, the J_{sc} of the cell could be obtained from equation 1 (Eq. 1).

$$J_{SC} = \int_{300}^{\lambda} \frac{q\lambda}{hc} \cdot E(\lambda) \cdot EQE(\lambda) \cdot d\lambda \tag{1}$$

where E(λ) is the spectral irradiance in AM 1.5G, λ is the absorption onset of the cell, h is Planck's constant, c is the speed of light and q is the elementary charge.

2) The V_{oc} of the cell is determined by the following Eq. 2.

$$V_{\mathrm{OC}} = \frac{1}{q}(E_g - E_{loss}) = \frac{1}{q}(\frac{1240}{\lambda} - E_{loss}) \tag{2}$$

The E_{loss} are assumed to be 0.4-0.8 eV according to overall reported values.[3]

So for the single cell, the PCE can be calculated from the Eq. 3 under AM 1.5G light illumination.

$$\begin{aligned} PCE(\%) &= V_{OC} \cdot J_{SC} \cdot FF / P_{in} \\ &= \frac{1}{q}(\frac{1240}{\lambda} - E_{loss})\int_{300}^{\lambda} \frac{q\lambda}{hc} \cdot E(\lambda) \cdot EQE(\lambda) \cdot d\lambda \cdot FF / P_{in} \end{aligned} \tag{3}$$

2.2 For the semi-expirical analysis based on a 2-terminal monolithic tandem cell with two subcells connected in series, the detailed description has been reported in literature.[4] The Figure 7b in the ms is obtained under the given EQE of 80% and FF%.

References:

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