

## **Electronic Supplementary Information:**

# **Triangulenium Dyes: the Comprehensive Photo-Absorption and Emission Story of a Versatile Family of Chromophores**

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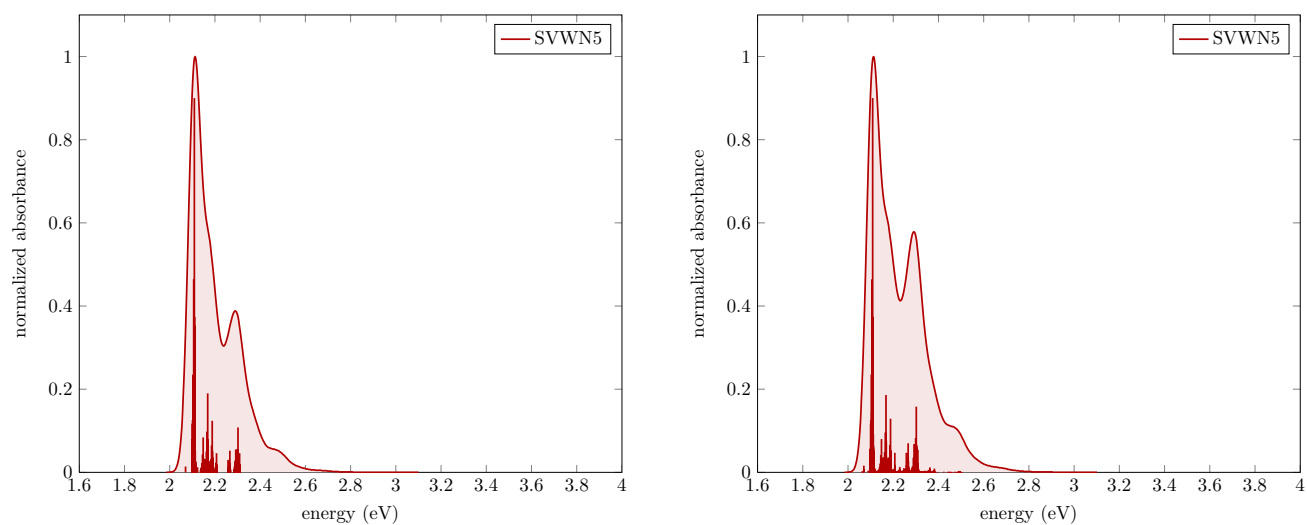
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# Density-Functional Benchmark

## 1 TATA

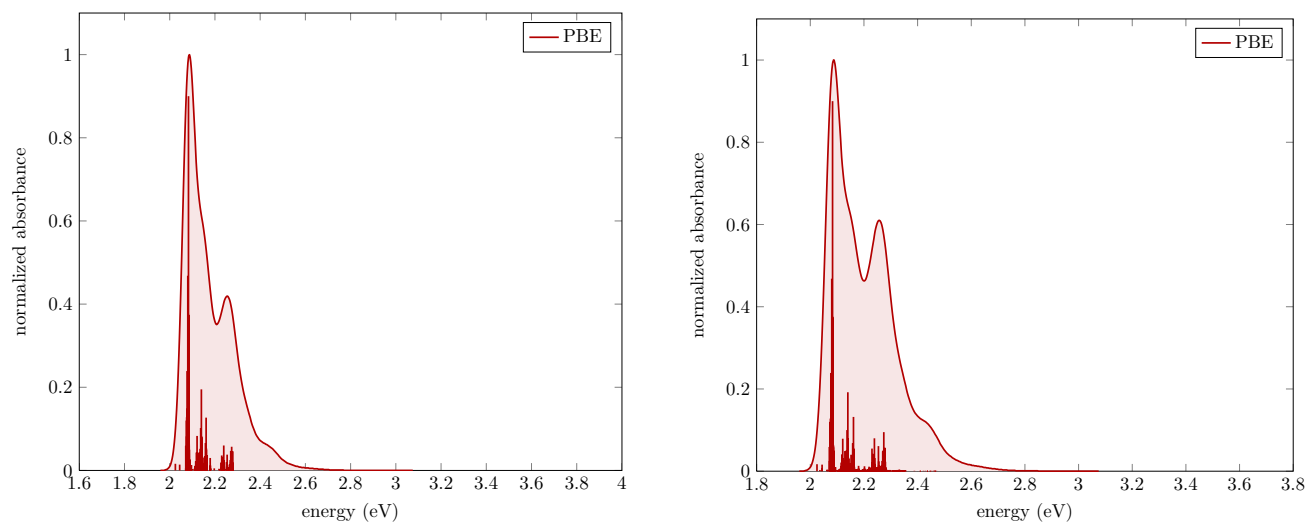
### 1.1 SVWN5



**Fig. S1** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at SVWN5/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

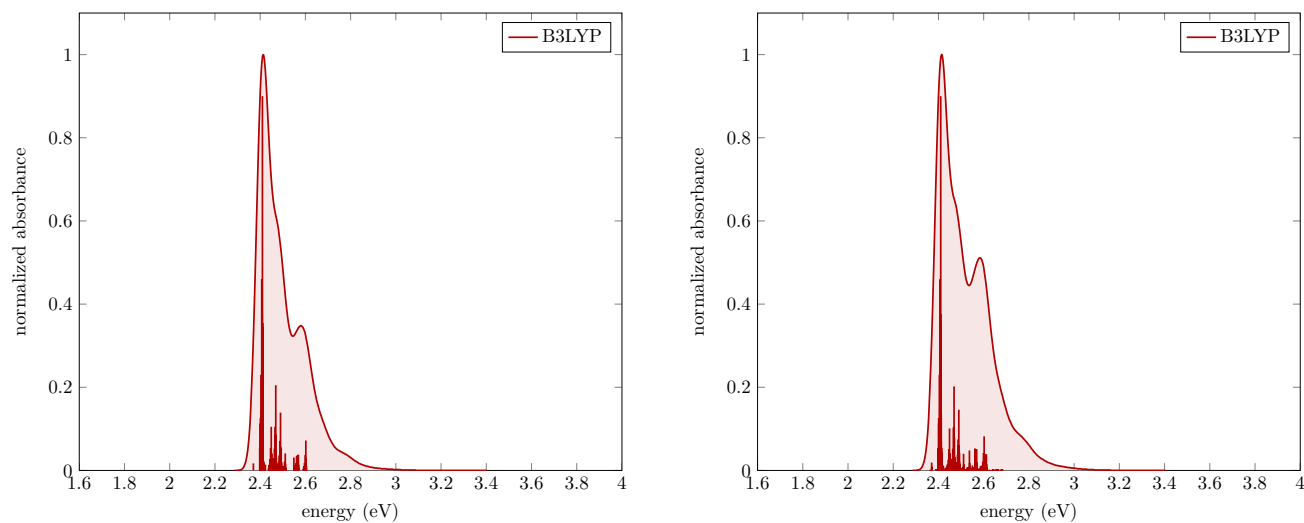


## 1.2 PBE



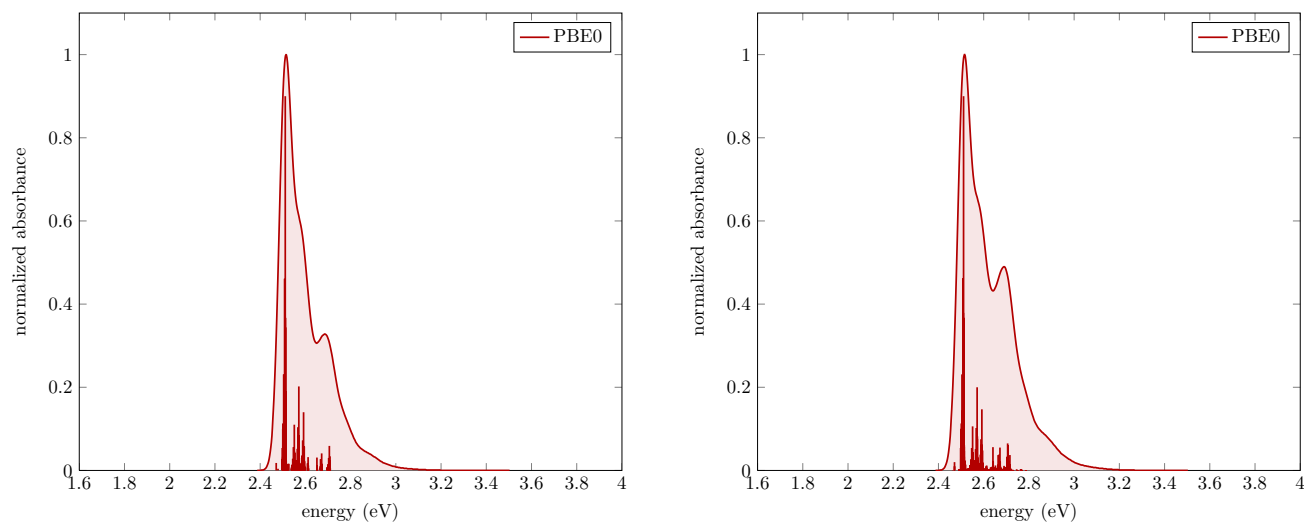
**Fig. S2** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

### 1.3 B3LYP



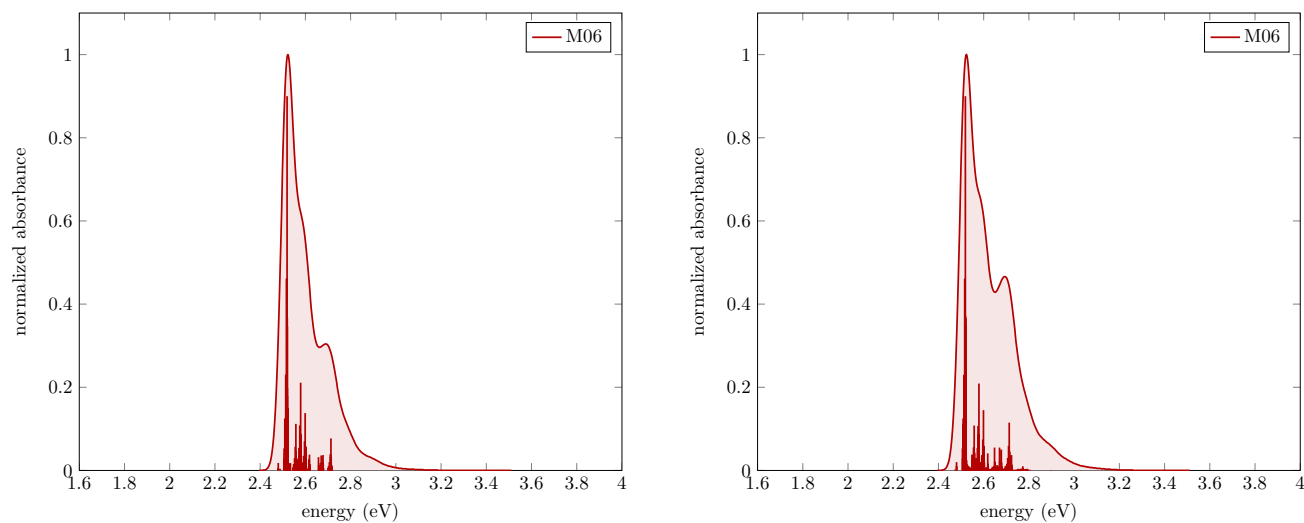
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## 1.4 PBE0



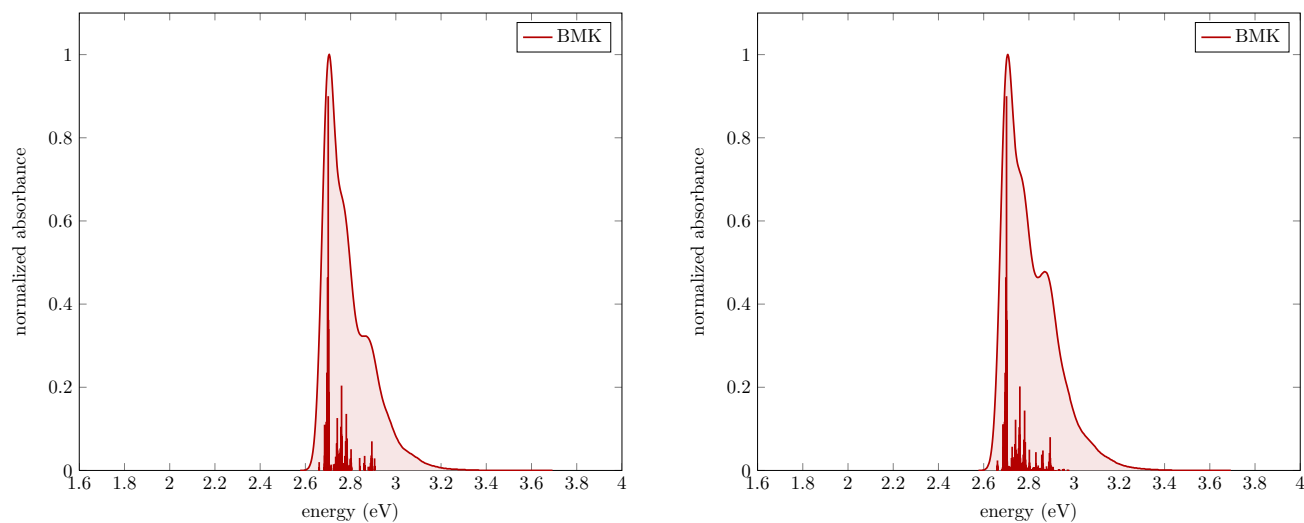
**Fig. S4** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 1.5 M06



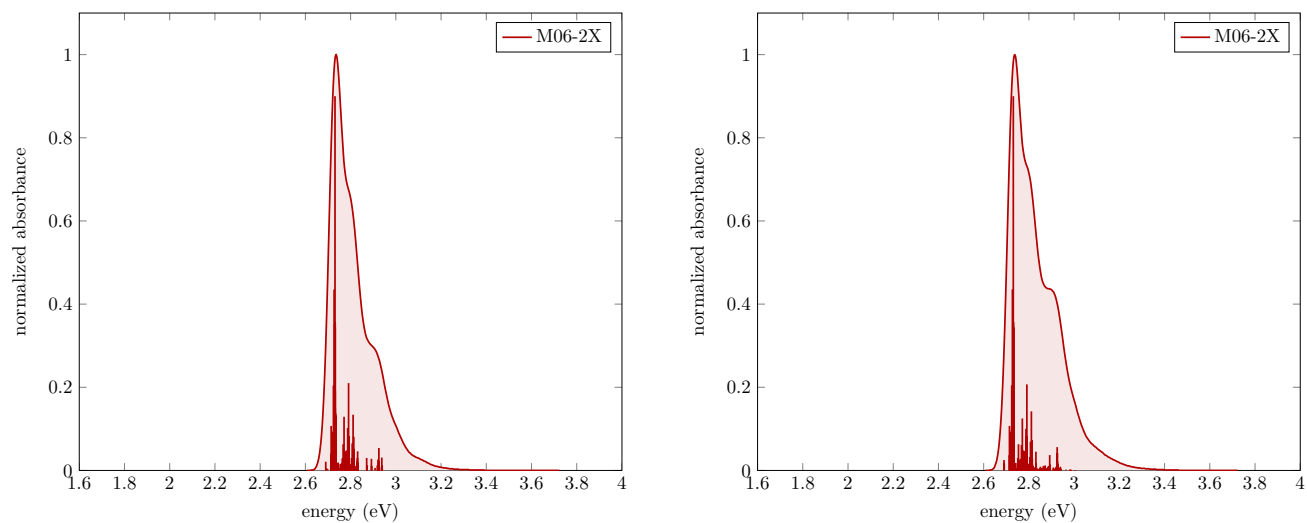
**Fig. S5** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 1.6 BMK



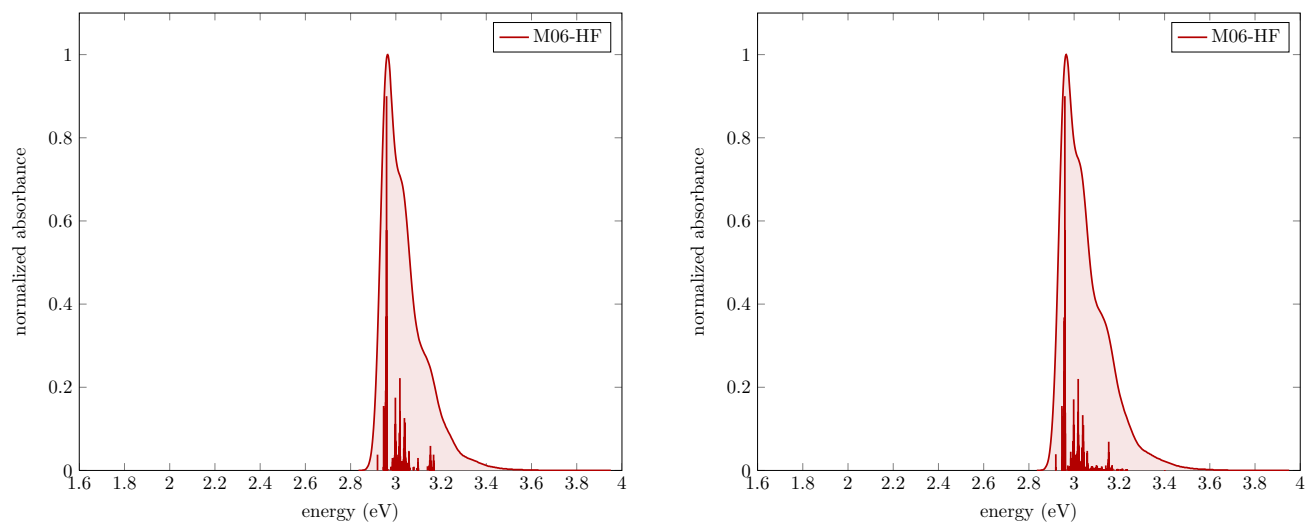
**Fig. S6** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at BMK/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 1.7 M06-2X



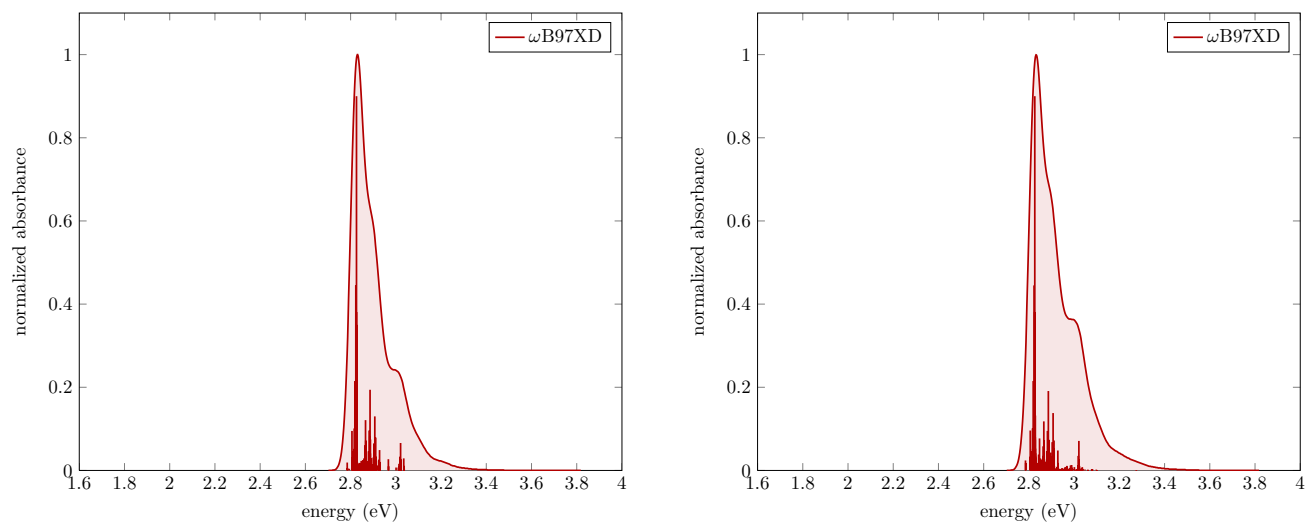
**Fig. S7** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06-2X/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM =  $250\text{ cm}^{-1}$ ) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 1.8 M06-HF



**Fig. S8** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06-HF/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

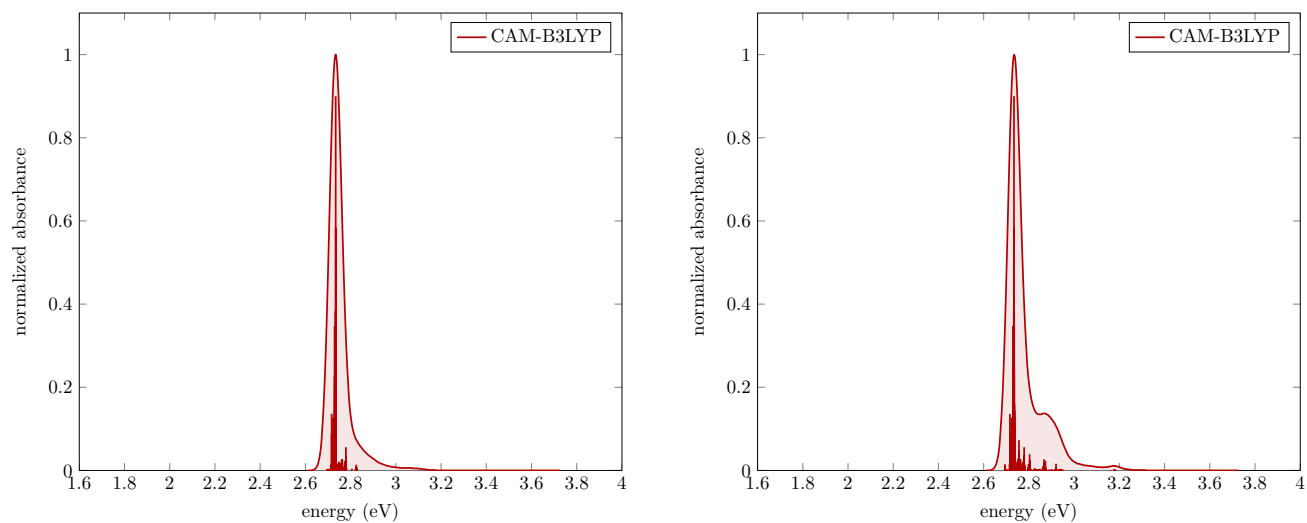
## 1.9 $\omega$ B97XD



**Fig. S9** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at  $\omega$ B97XD/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.



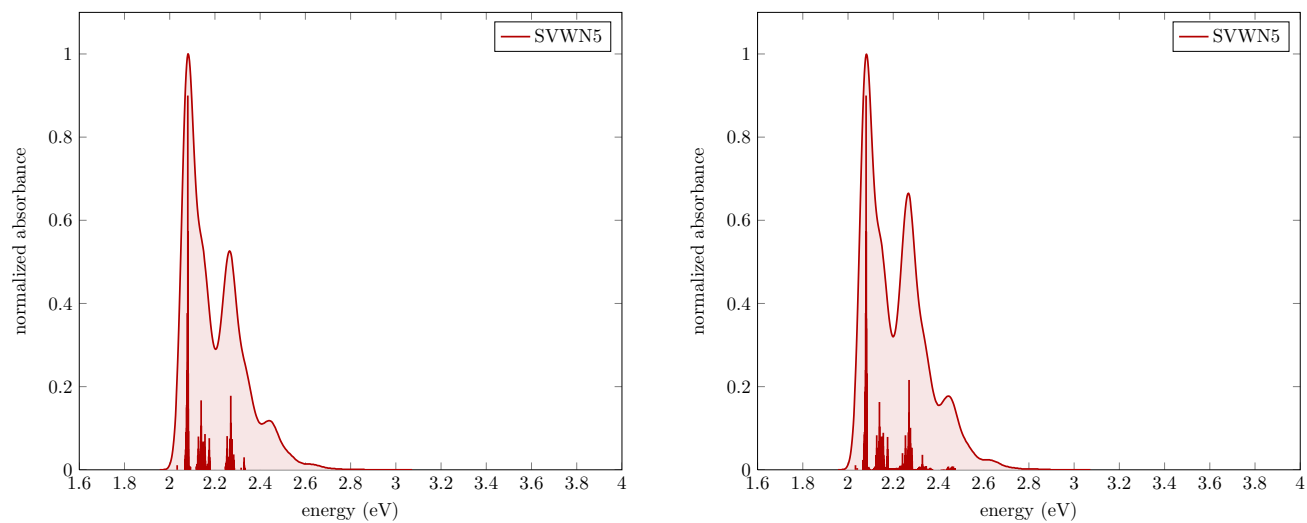
## 1.10 CAM-B3LYP



**Fig. S10** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at CAM-B3LYP/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

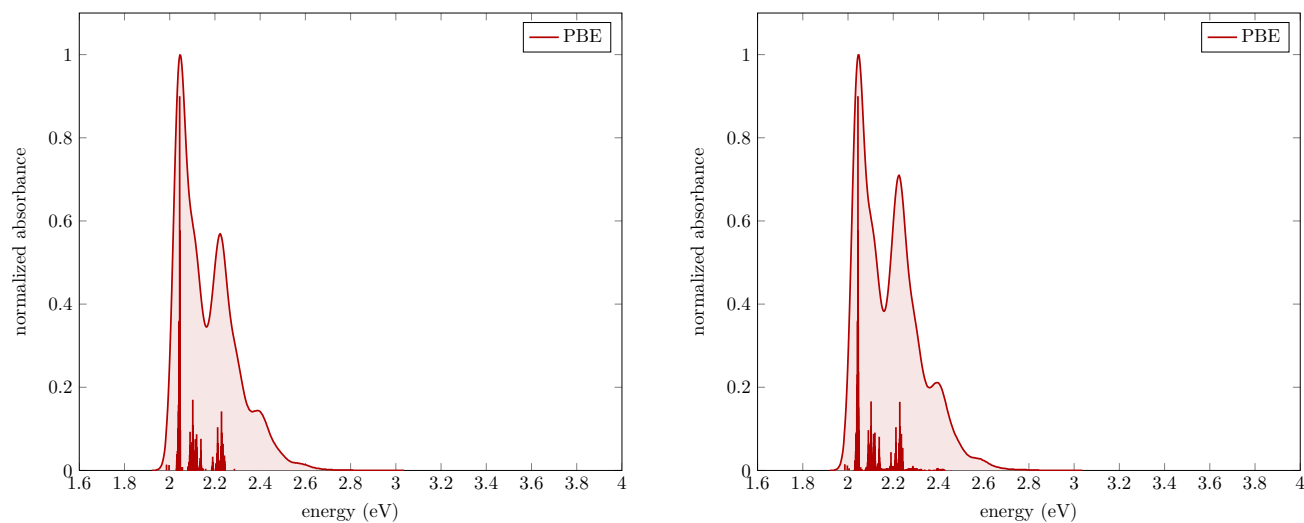
## 2 ADOTA

### 2.1 SVWN5



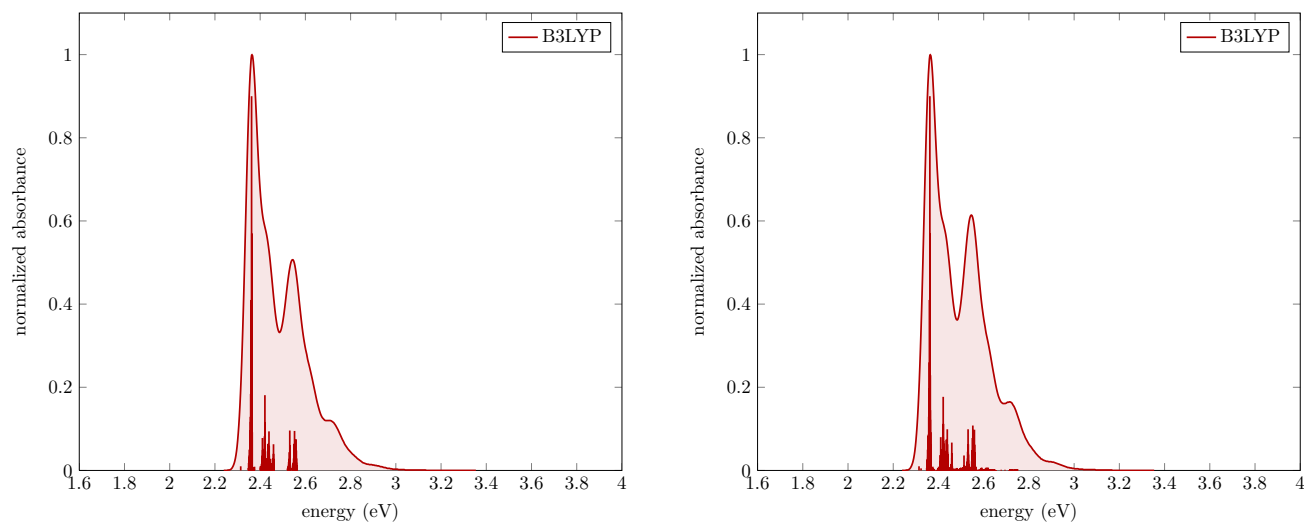
**Fig. S11** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at SVWN5/6-31+G<sup>\*</sup> level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 2.2 PBE



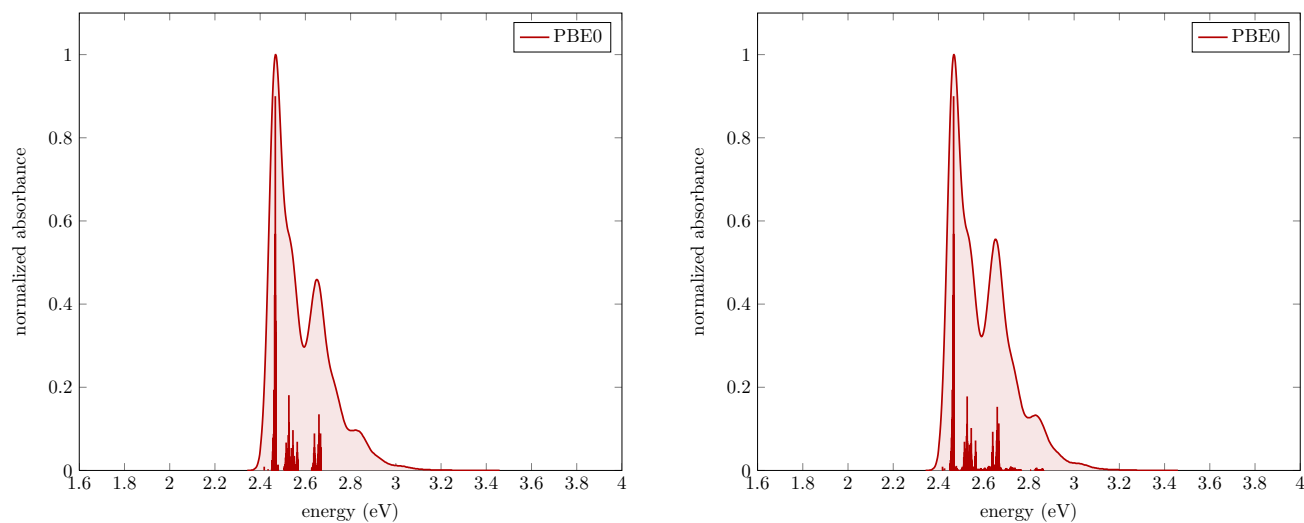
**Fig. S12** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 2.3 B3LYP



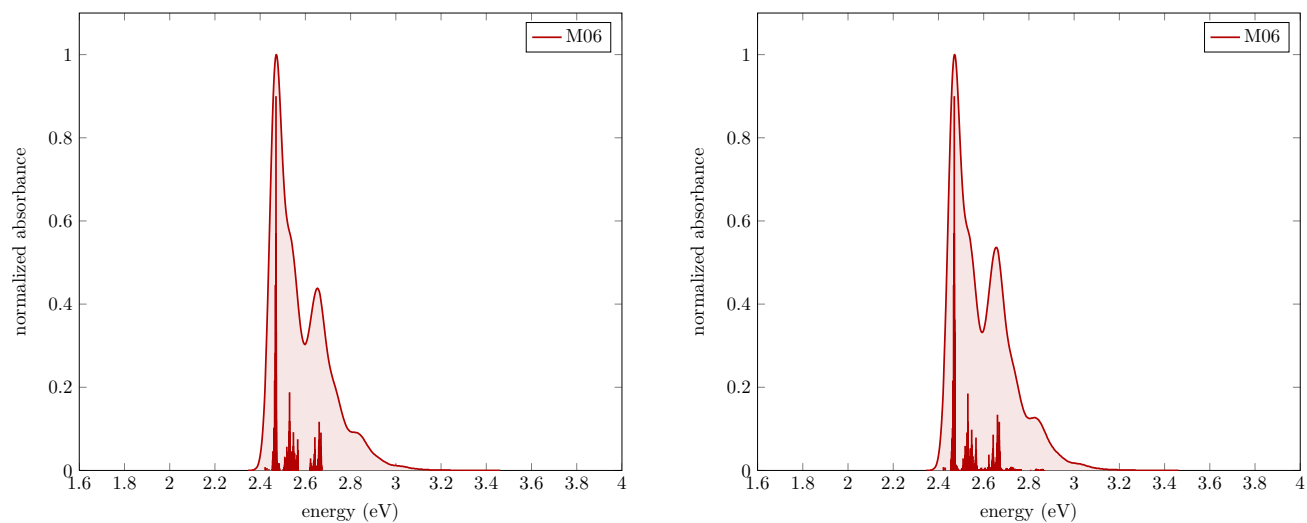
**Fig. S13** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at B3LYP/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 2.4 PBE0



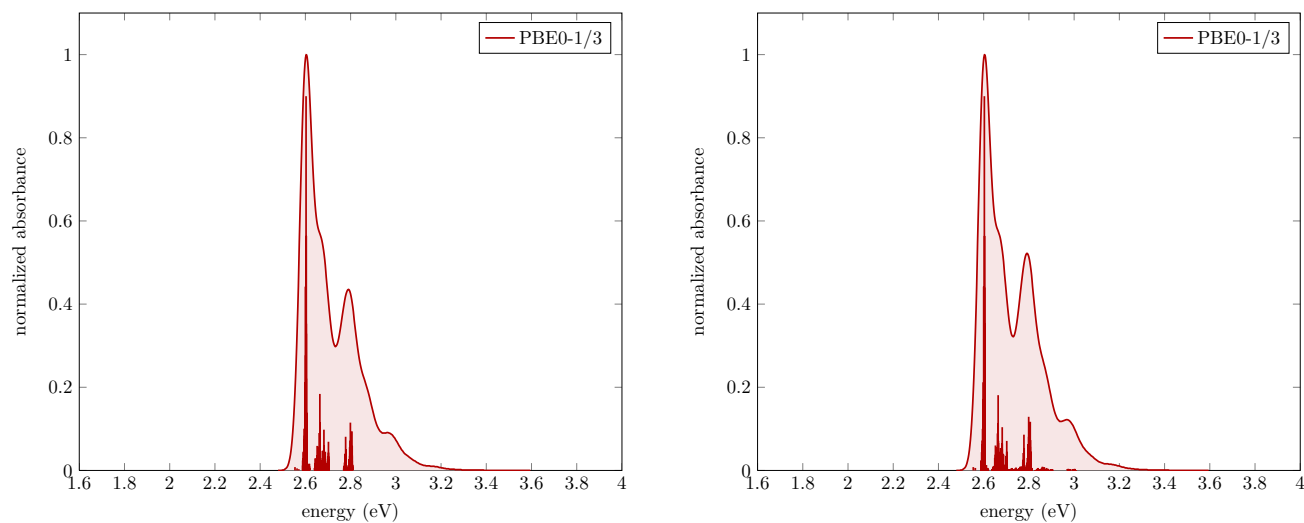
**Fig. S14** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 2.5 M06



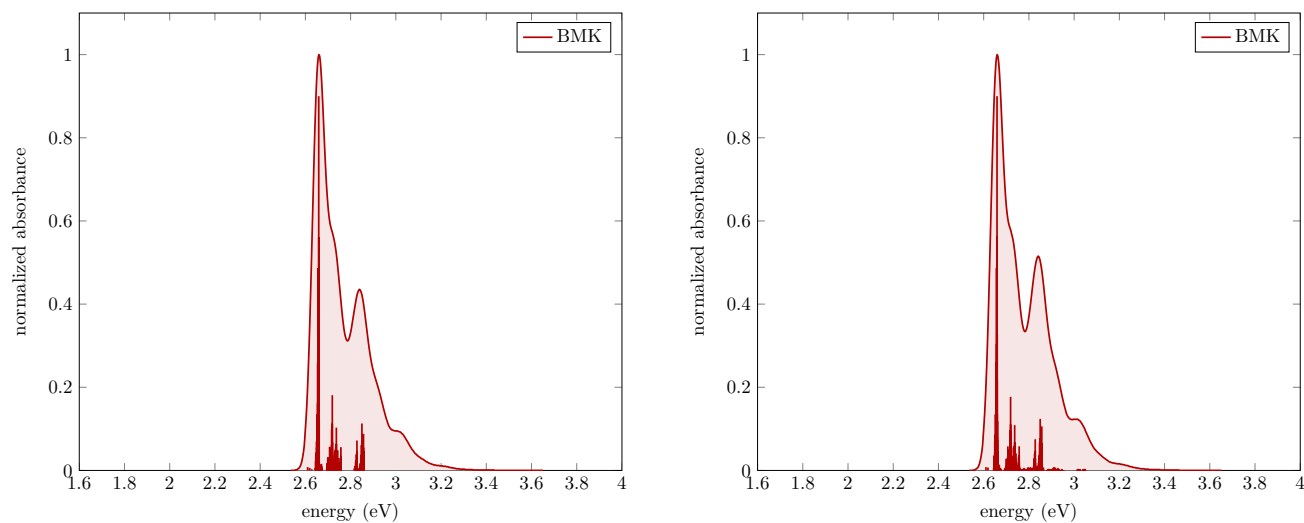
**Fig. S15** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 2.6 PBE0-1/3



**Fig. S16** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0-1/3/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

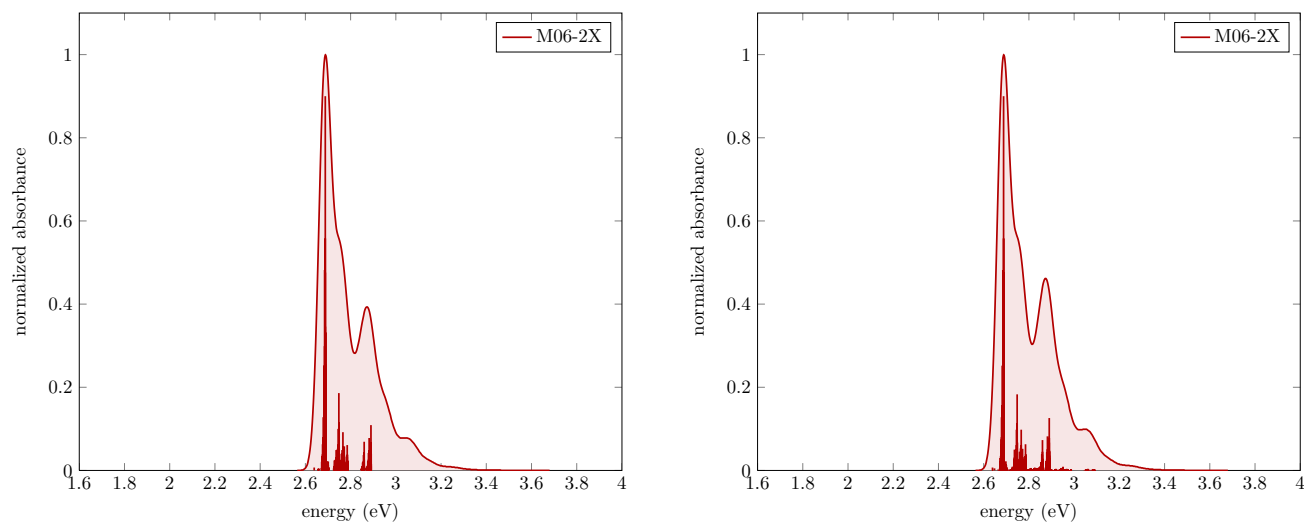
## 2.7 BMK



**Fig. S17** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at BMK/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

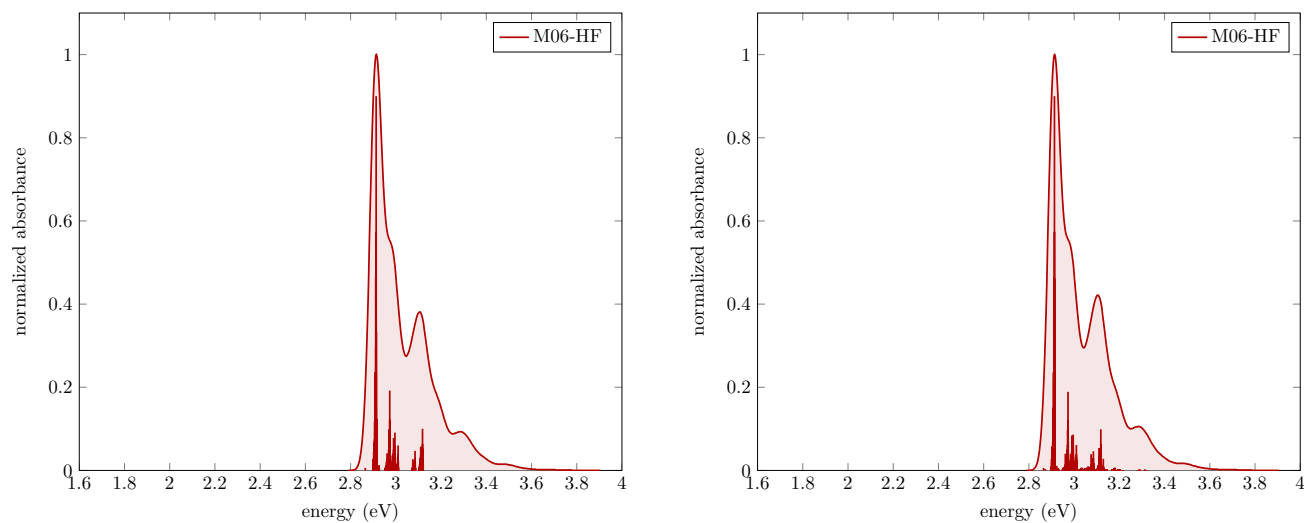


## 2.8 M06-2X



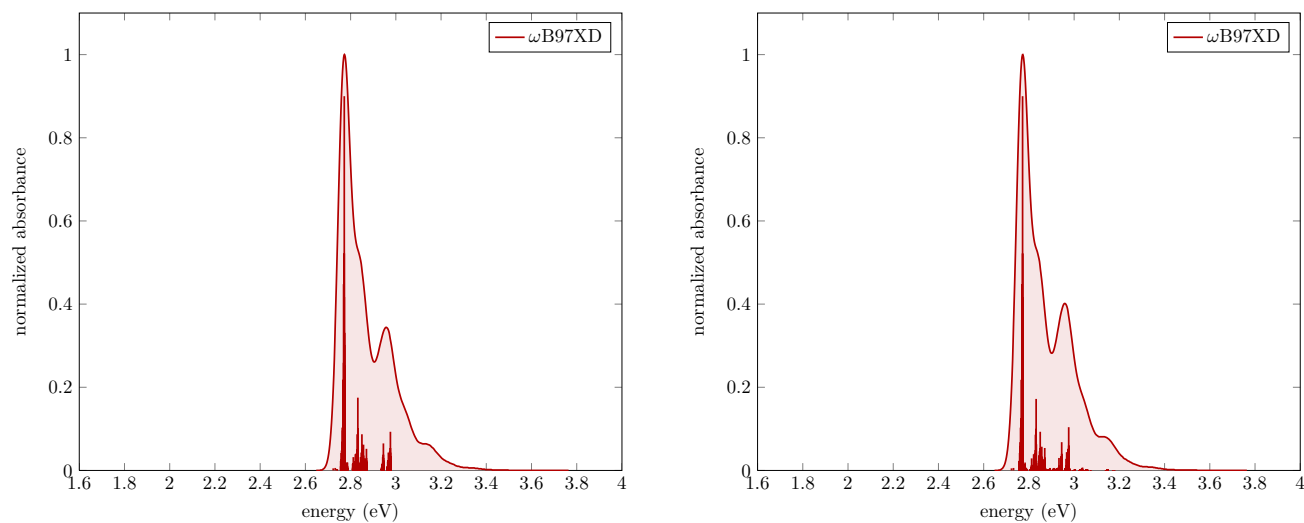
**Fig. S18** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06-2X/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 2.9 M06-HF



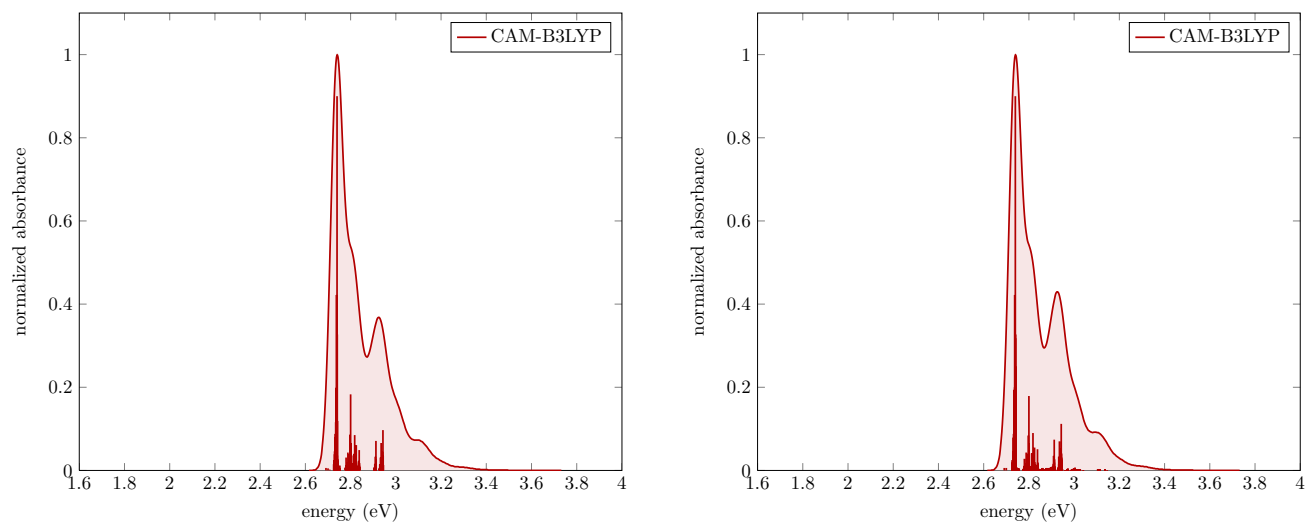
**Fig. S19** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06-HF/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 2.10 $\omega$ B97XD



**Fig. S20** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at  $\omega$ B97XD/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

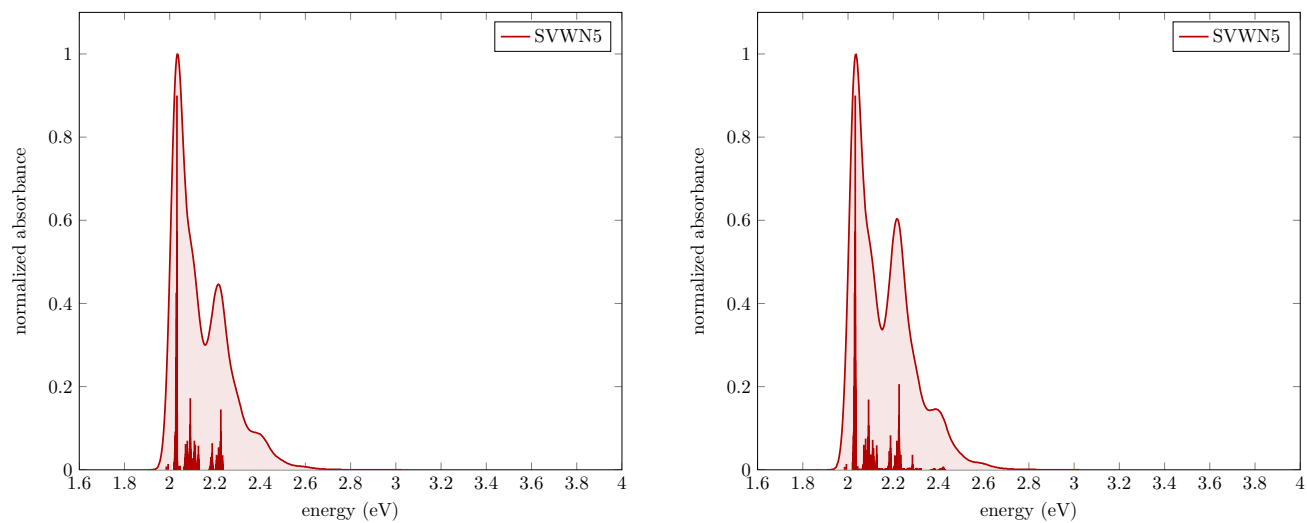
## 2.11 CAM-B3LYP



**Fig. S21** UV/vis absorption spectra of ADOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at CAM-B3LYP/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

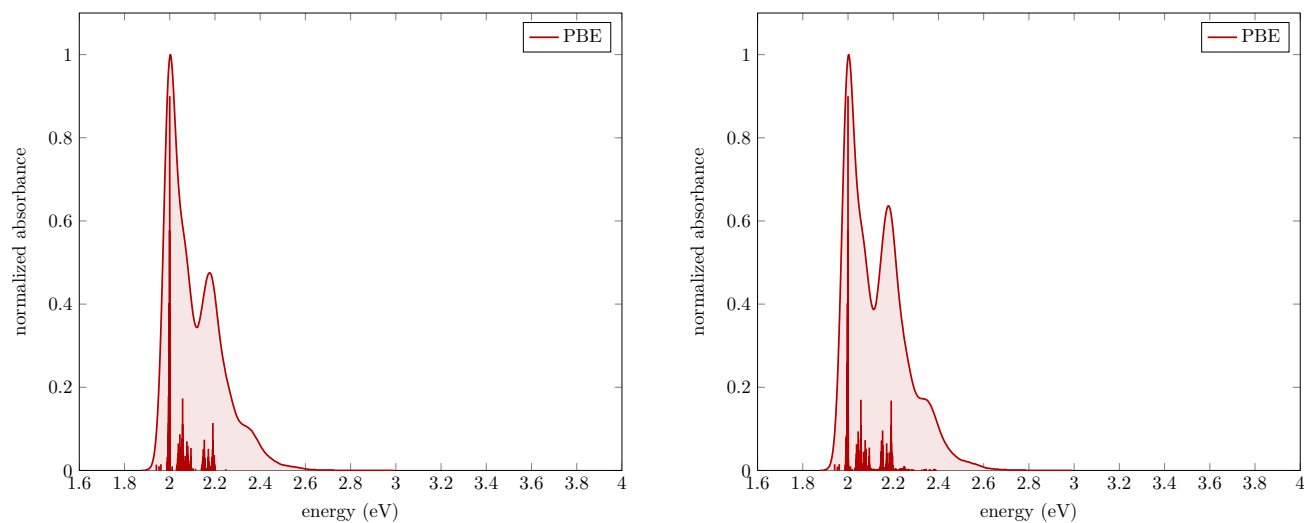
### 3 DAOTA

#### 3.1 SVWN5



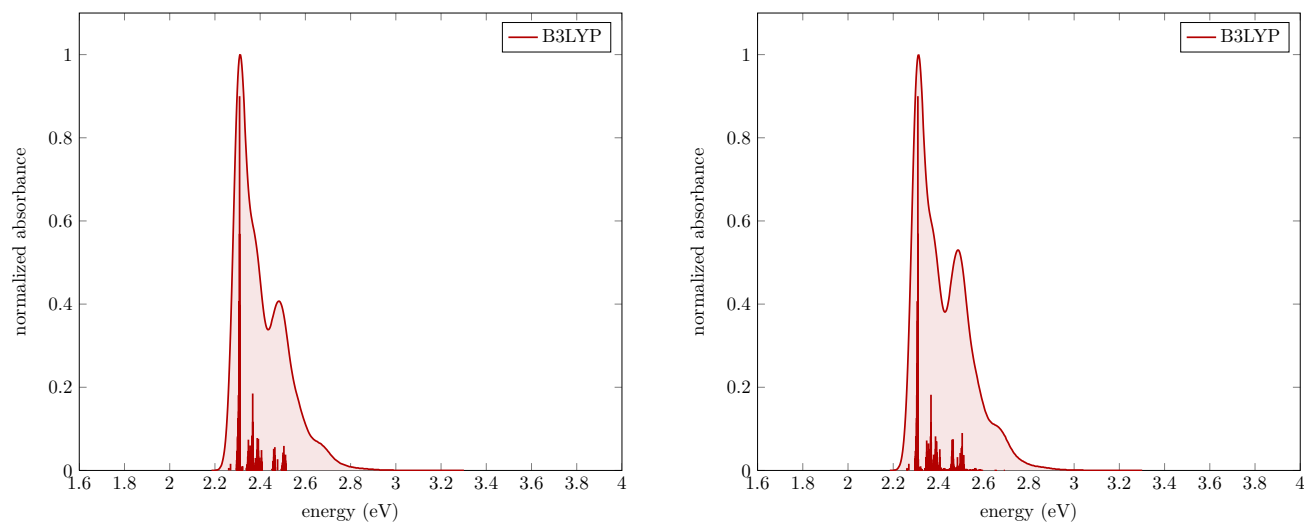
**Fig. S22** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at SVWN5/6-31+G<sup>\*</sup> level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 3.2 PBE



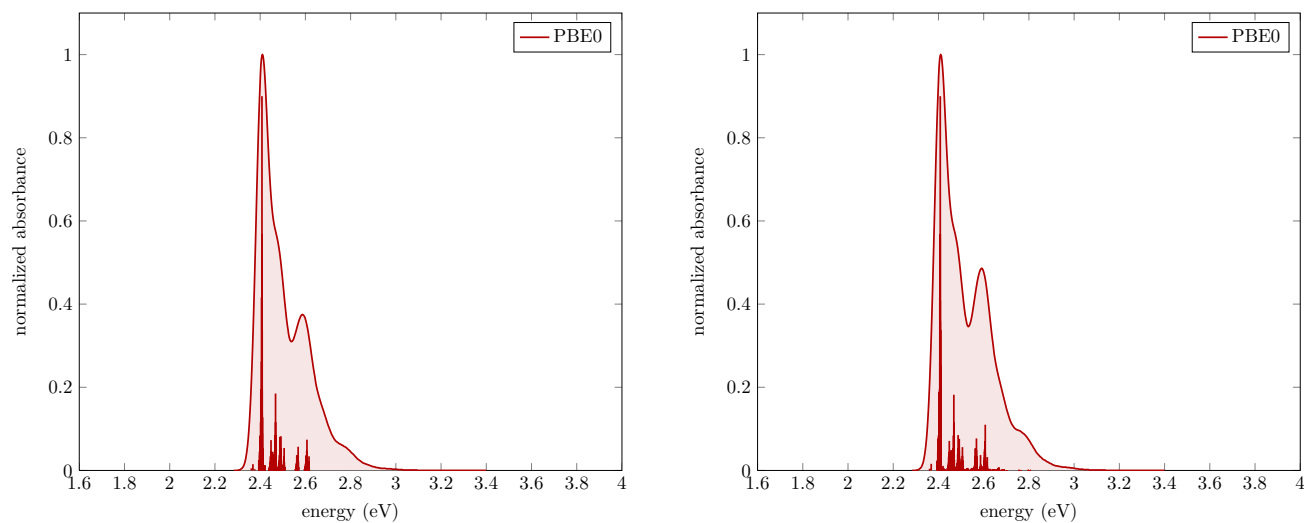
**Fig. S23** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

### 3.3 B3LYP



**Fig. S24** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at B3LYP/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

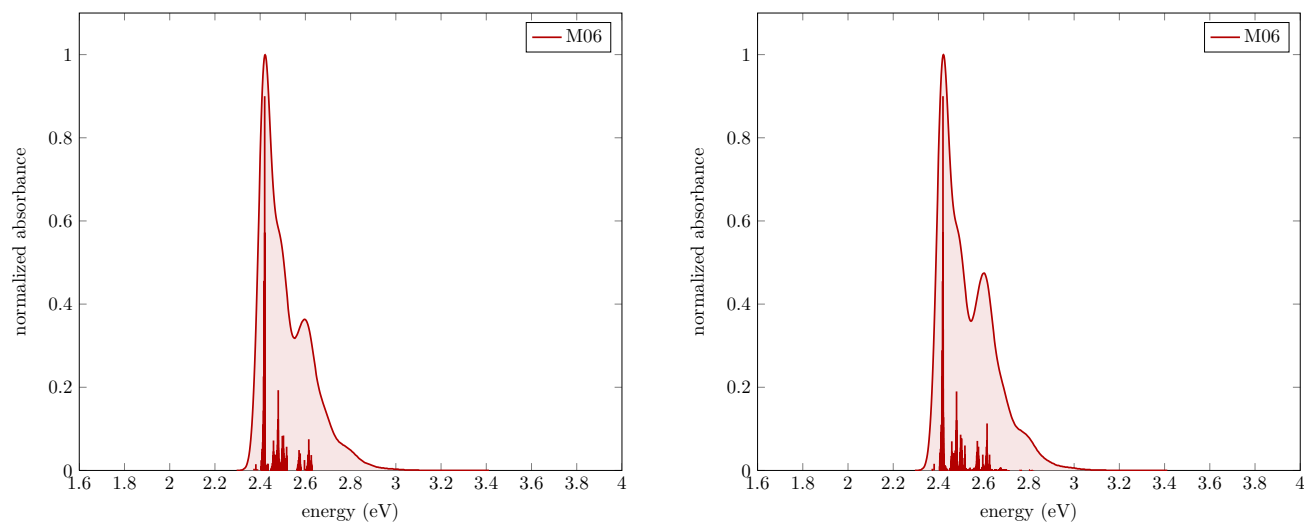
### 3.4 PBE0



**Fig. S25** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

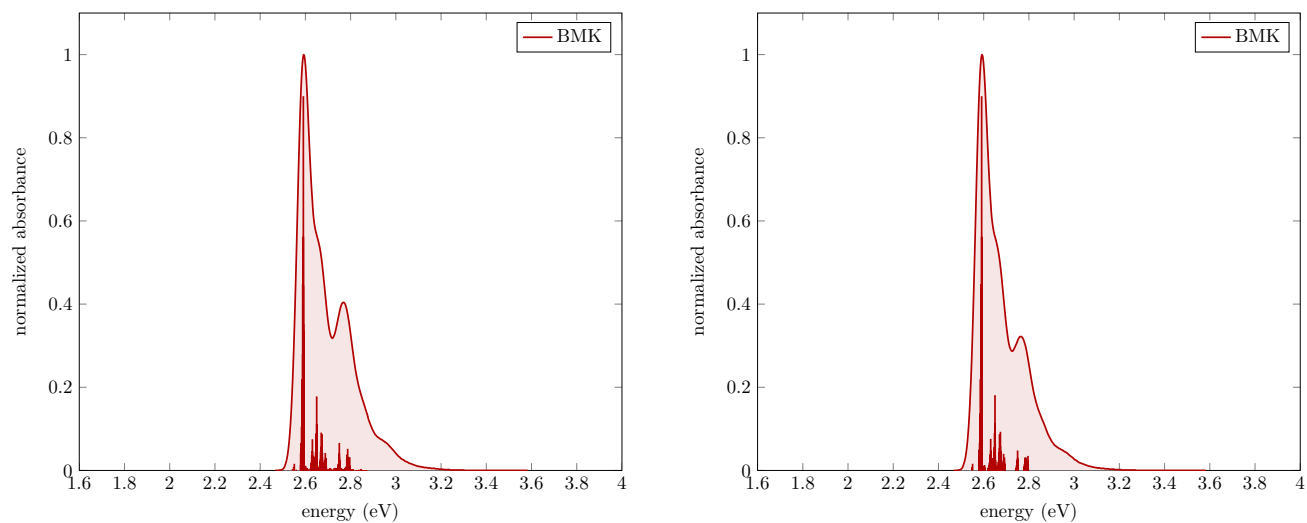


### 3.5 M06



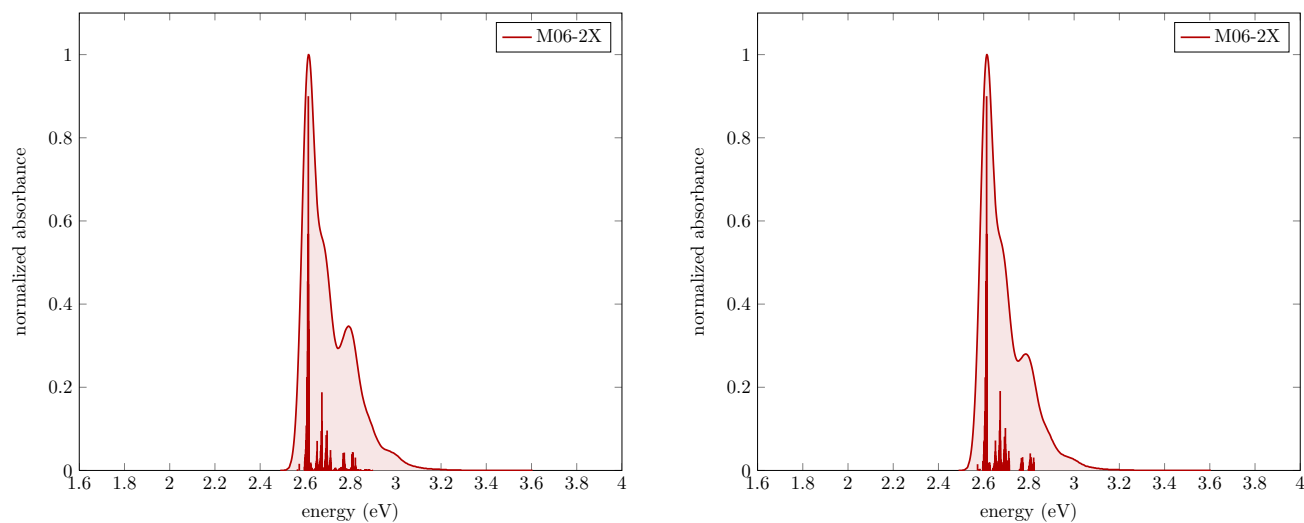
**Fig. S26** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

### 3.6 BMK



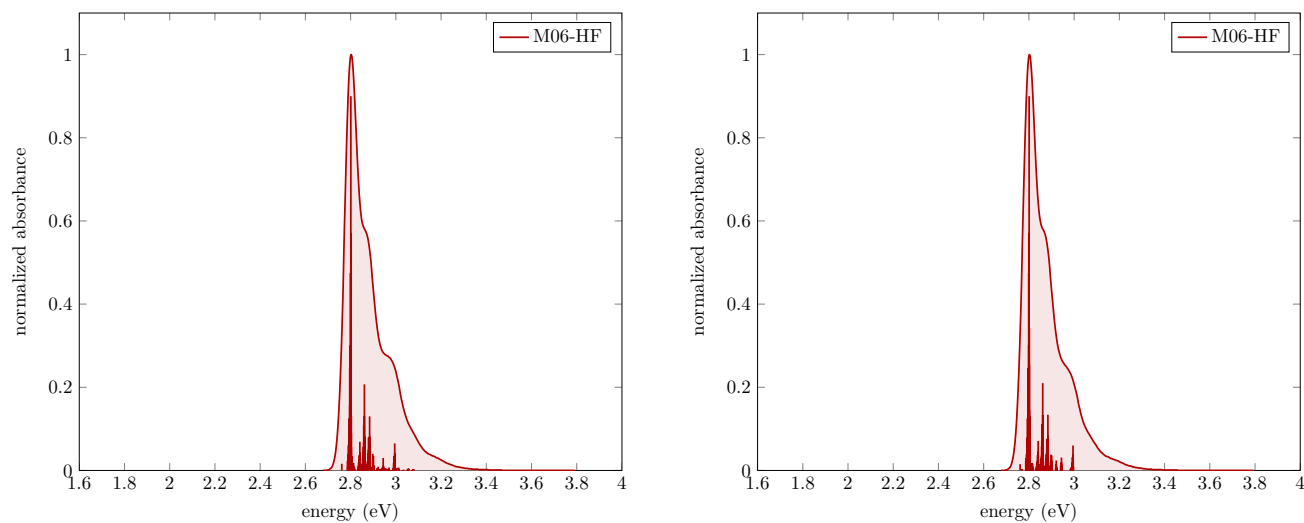
**Fig. S27** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at BMK/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

### 3.7 M06-2X



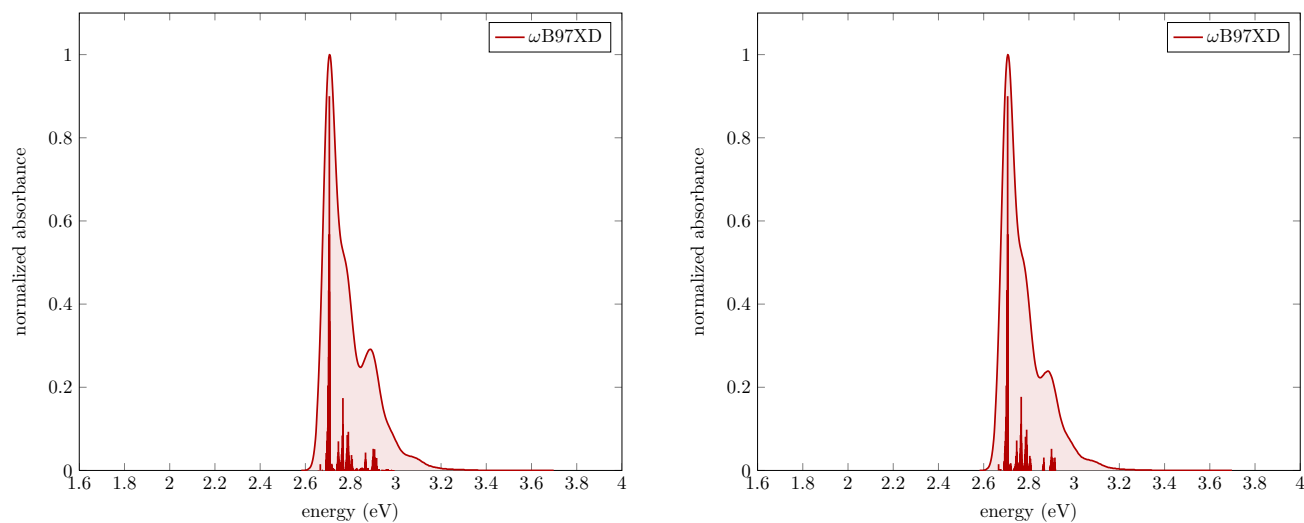
**Fig. S28** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06-2X/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

### 3.8 M06-HF



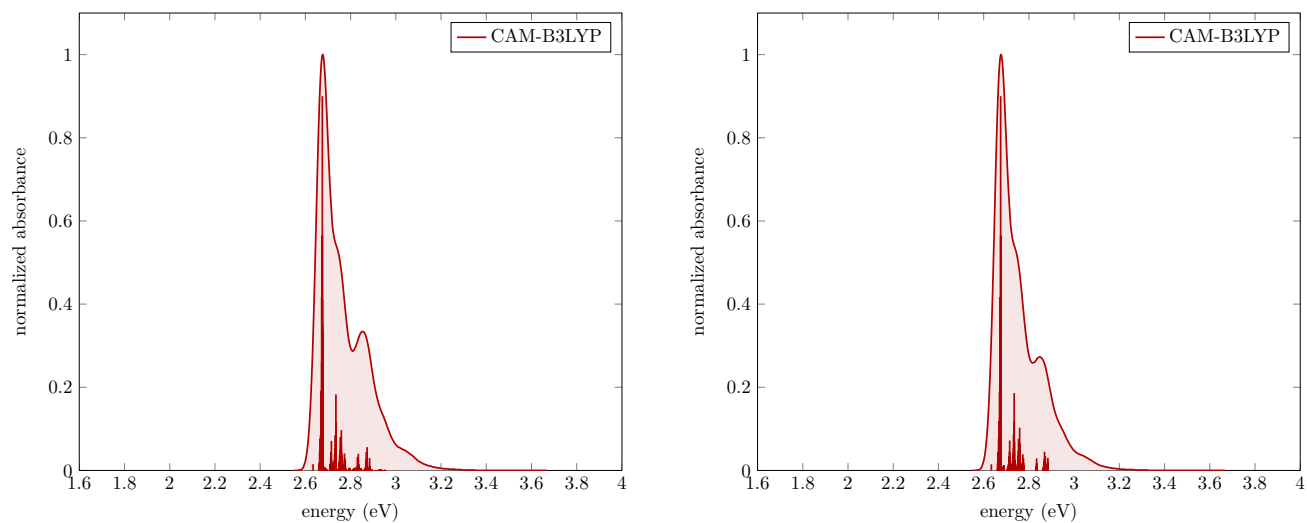
**Fig. S29** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06-HF/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

### 3.9 $\omega$ B97XD



**Fig. S30** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at  $\omega$ B97XD/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

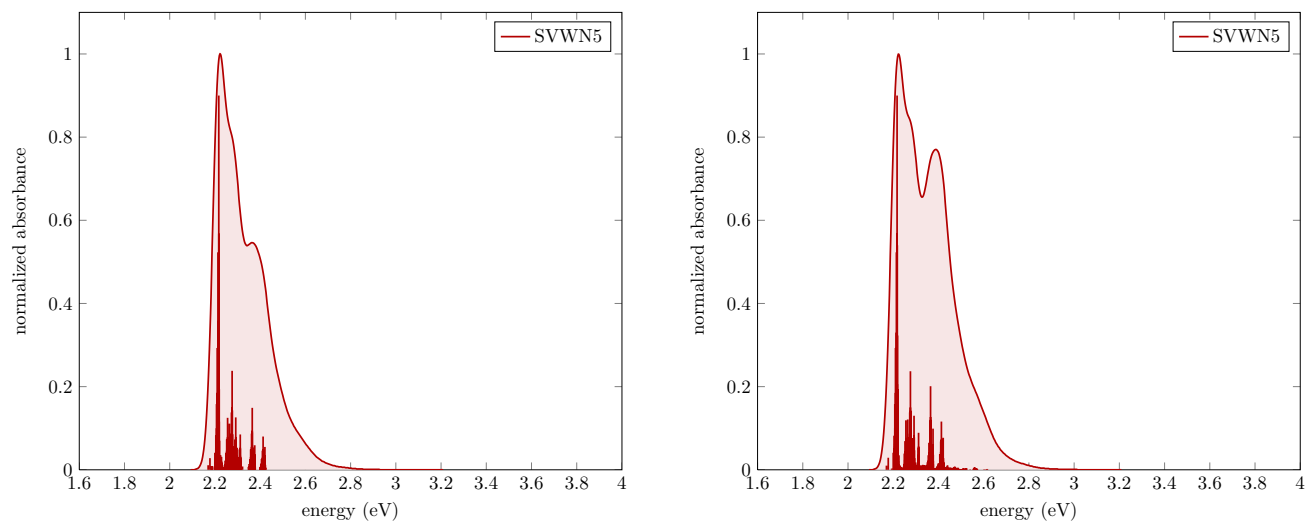
### 3.10 CAM-B3LYP



**Fig. S31** UV/vis absorption spectra of DAOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at CAM-B3LYP/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

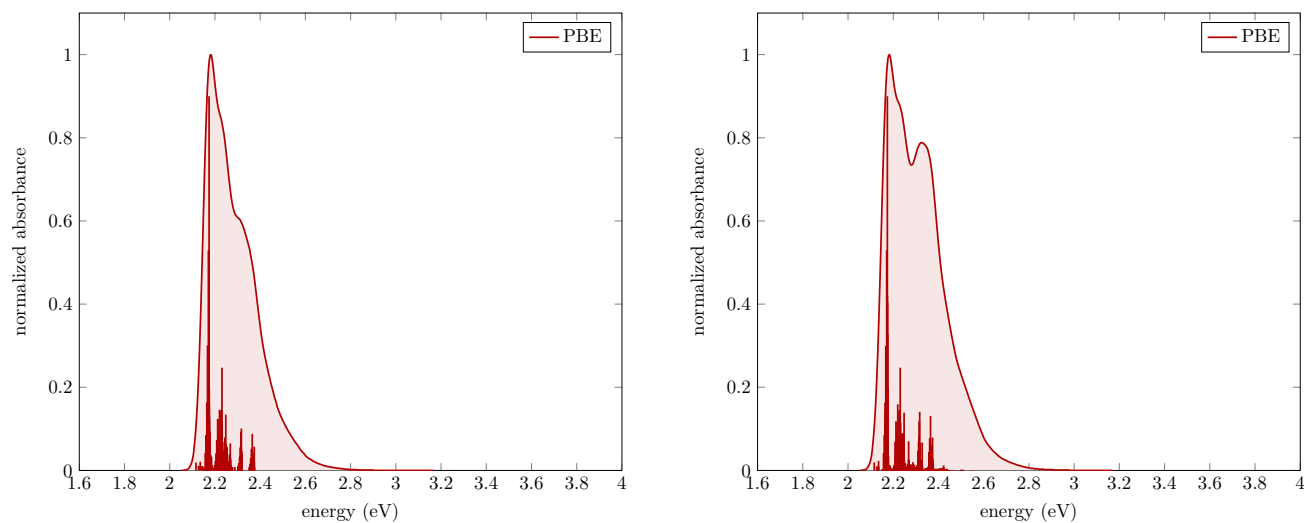
## 4 TOTA

### 4.1 SVWN5



**Fig. S32** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at SVWN5/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

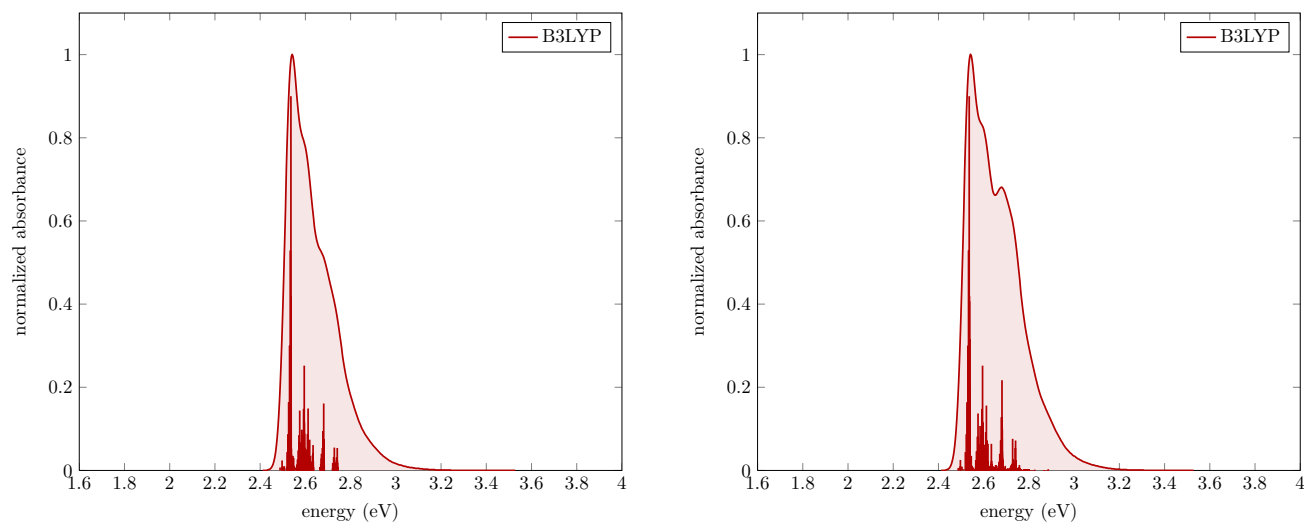
## 4.2 PBE



**Fig. S33** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

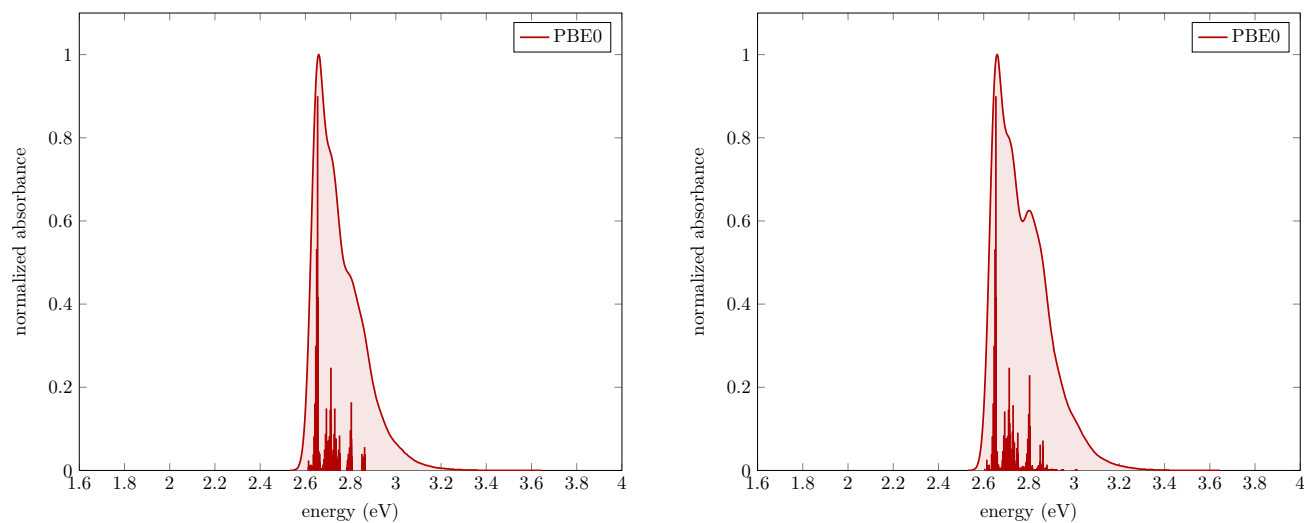


### 4.3 B3LYP



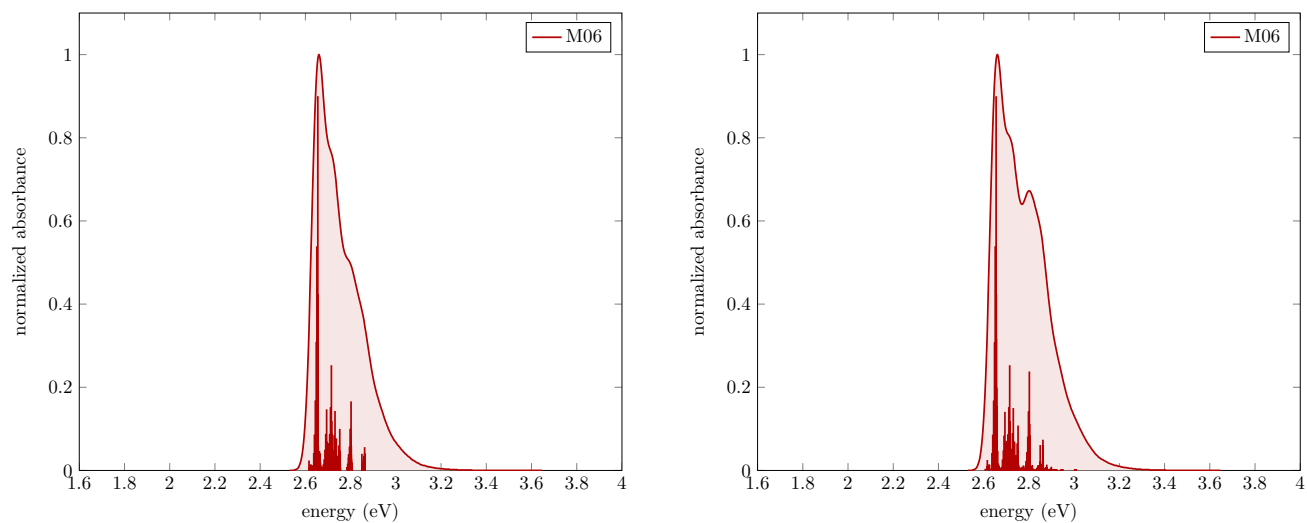
**Fig. S34** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at B3LYP/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 4.4 PBE0



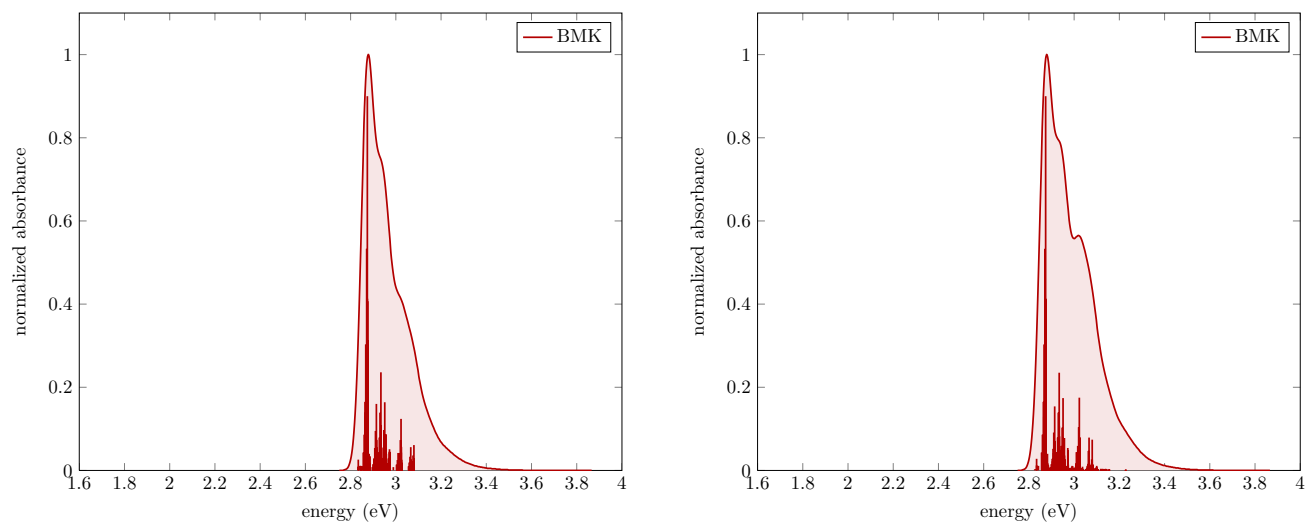
**Fig. S35** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 4.5 M06



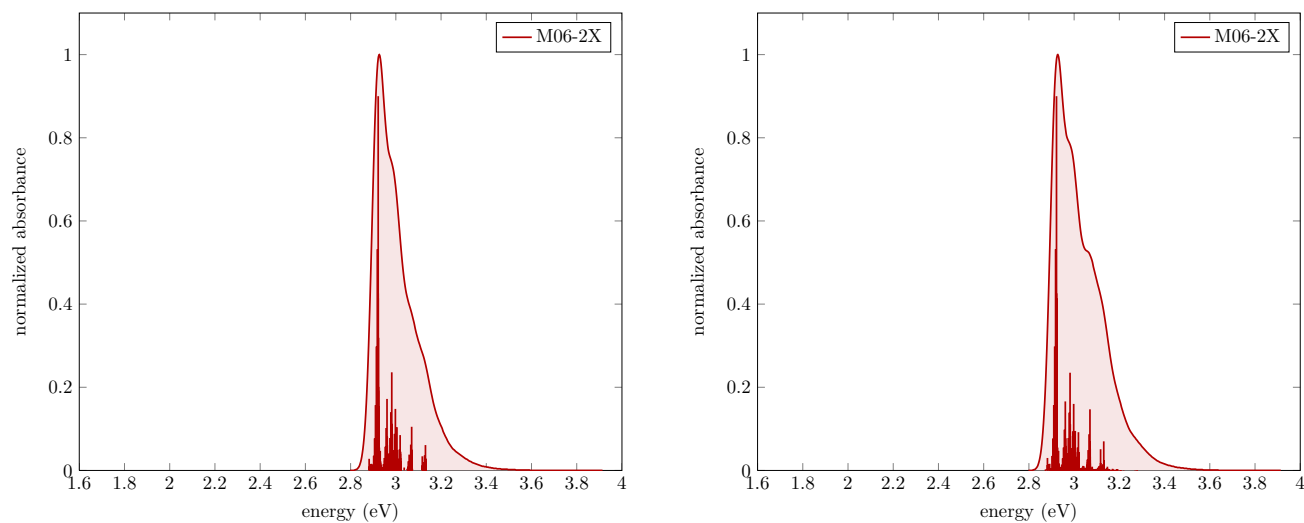
**Fig. S36** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 4.6 BMK



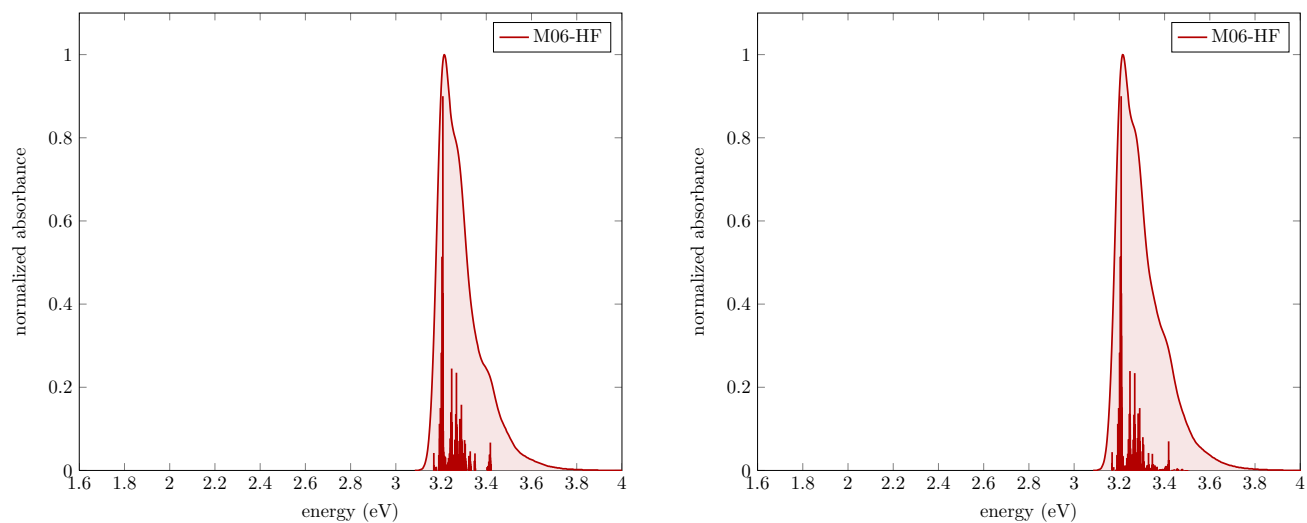
**Fig. S37** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at BMK/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 4.7 M06-2X



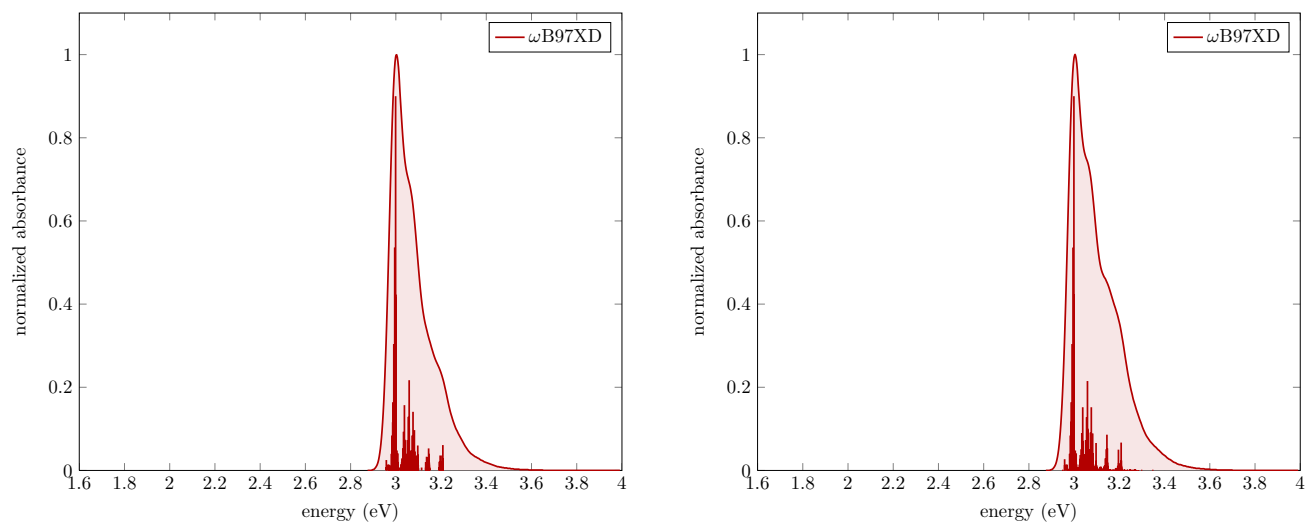
**Fig. S38** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06-2X/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 4.8 M06-HF



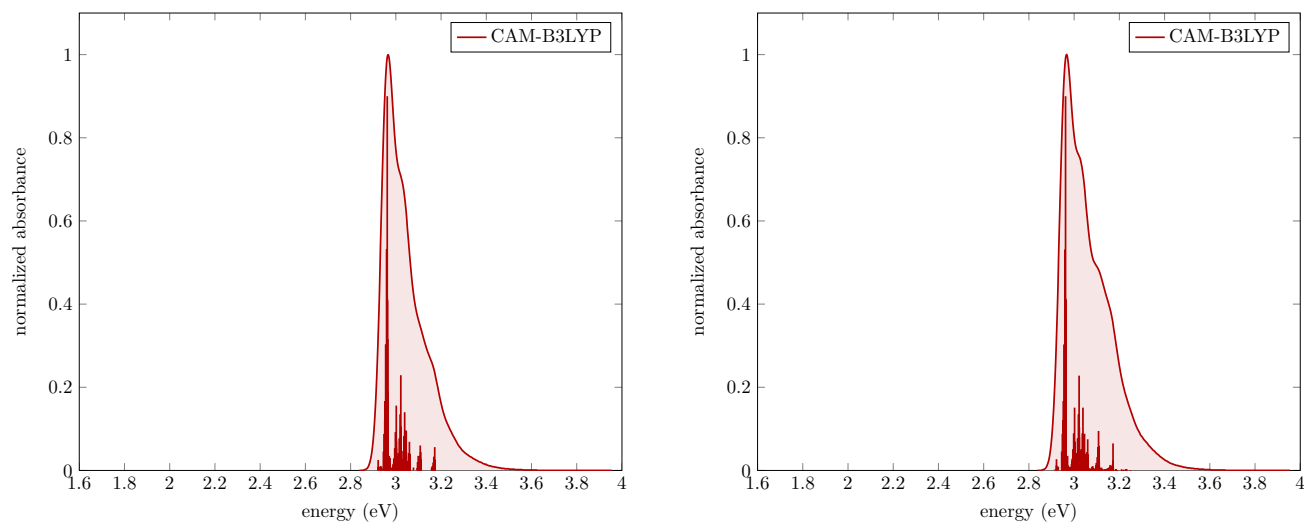
**Fig. S39** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at M06-HF/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 4.9 $\omega$ B97XD



**Fig. S40** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at  $\omega$ B97XD/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 4.10 CAM-B3LYP



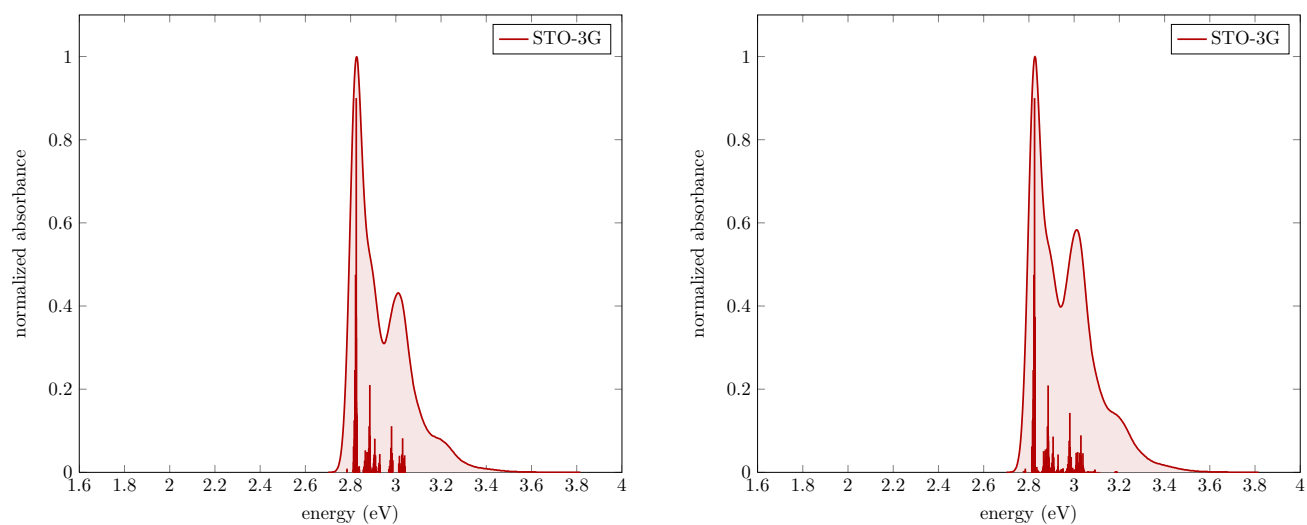
**Fig. S41** UV/vis absorption spectra of TOTA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at CAM-B3LYP/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.



# Basis-Set Benchmark

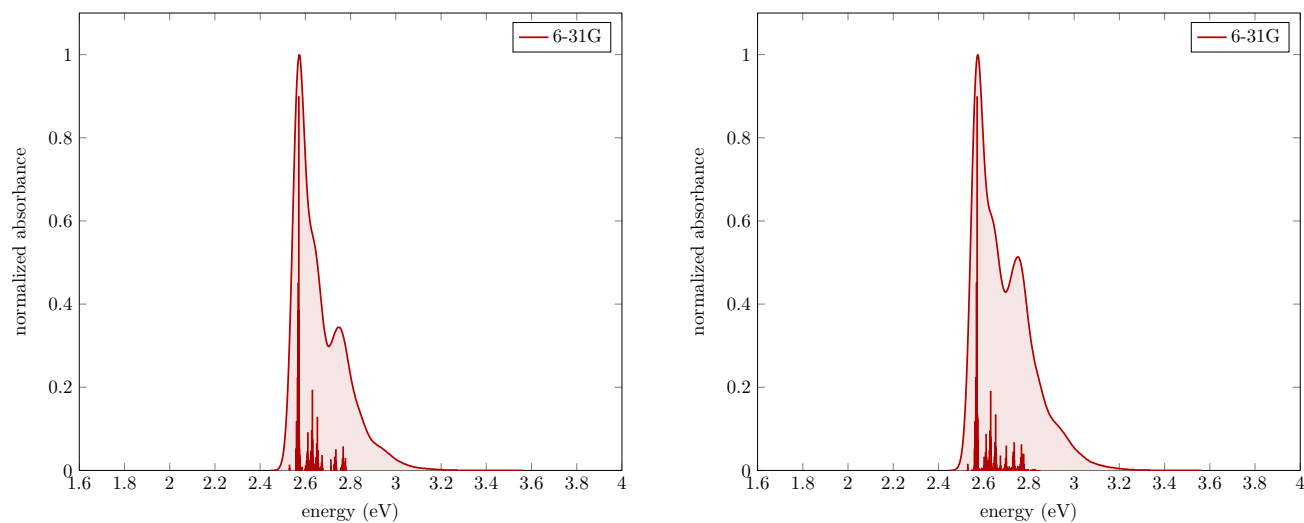
## 5 TATA

### 5.1 STO-3G



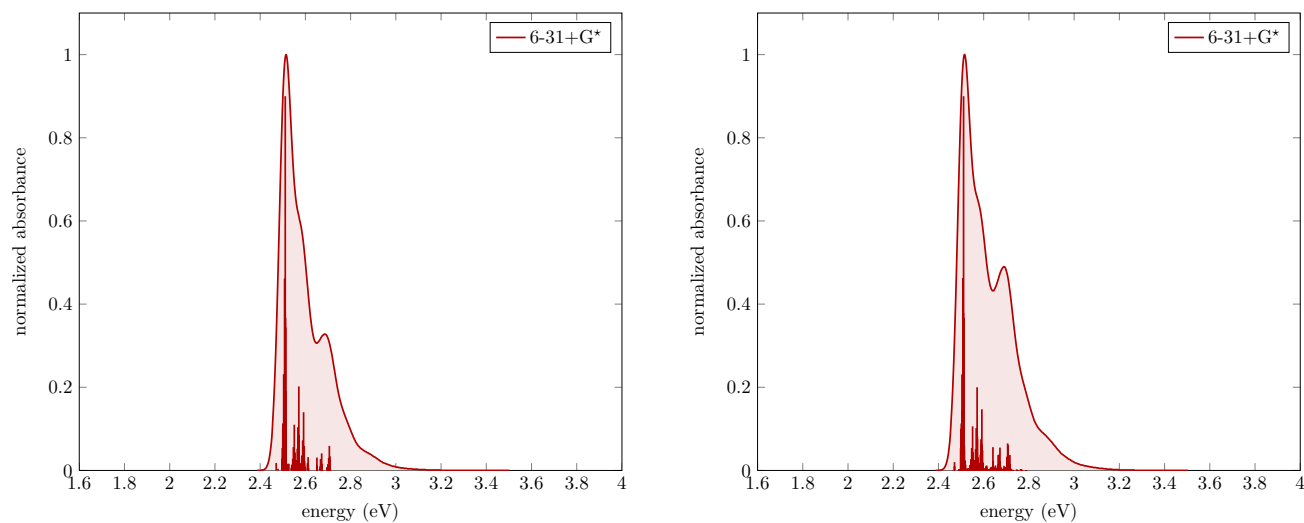
**Fig. S42** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/STO-3G level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution ( $\text{FWHM} = 250 \text{ cm}^{-1}$ ) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 5.2 6-31G



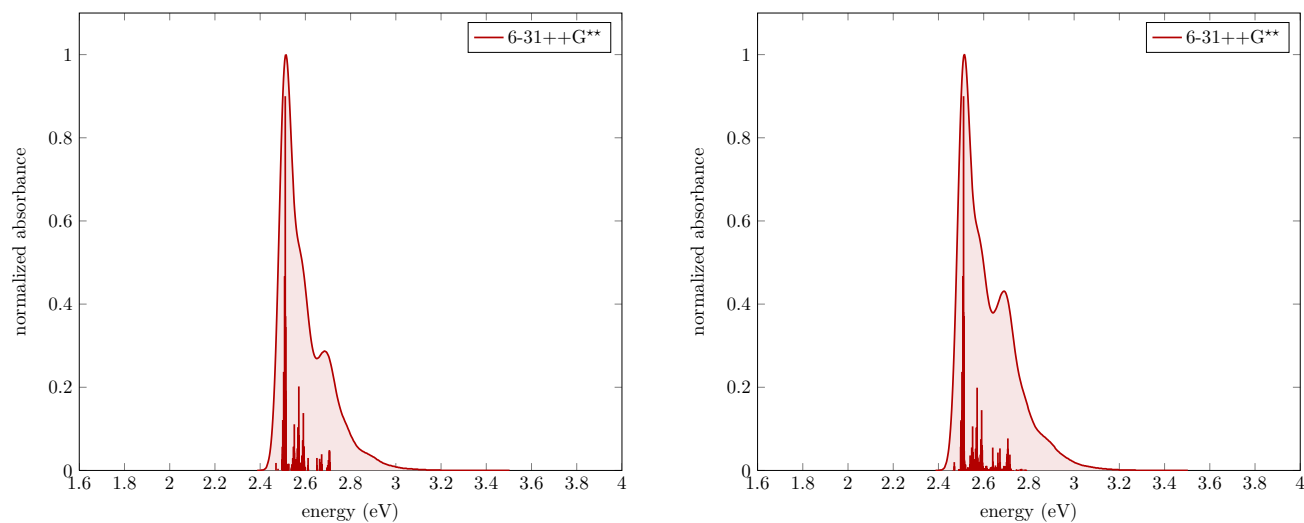
**Fig. S43** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31G level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

### 5.3 6-31+G\*



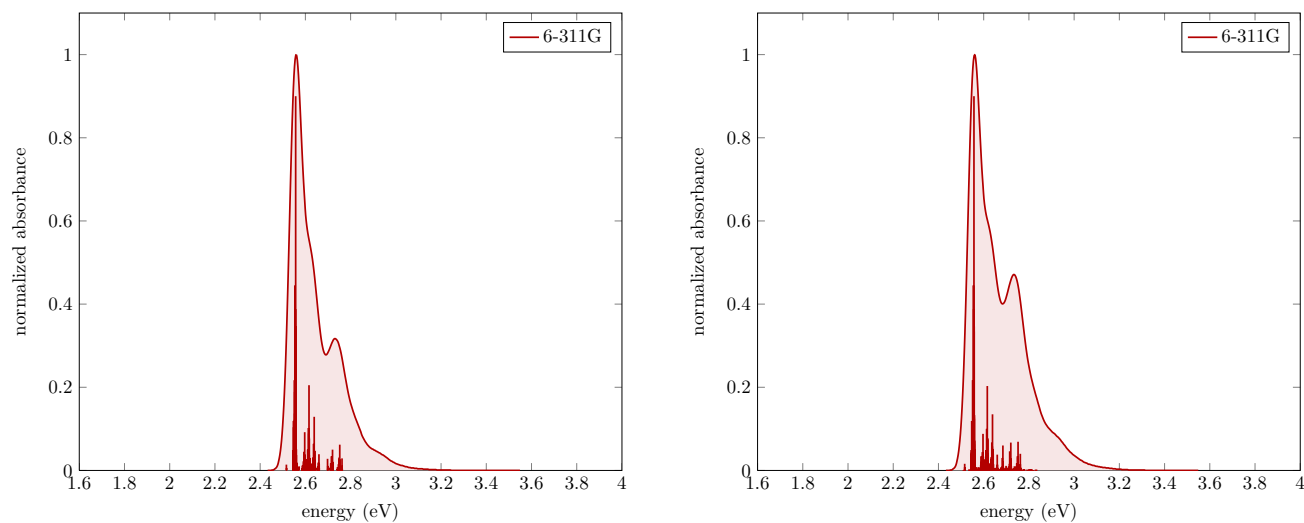
**Fig. S44** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 5.4 6-31++G\*\*



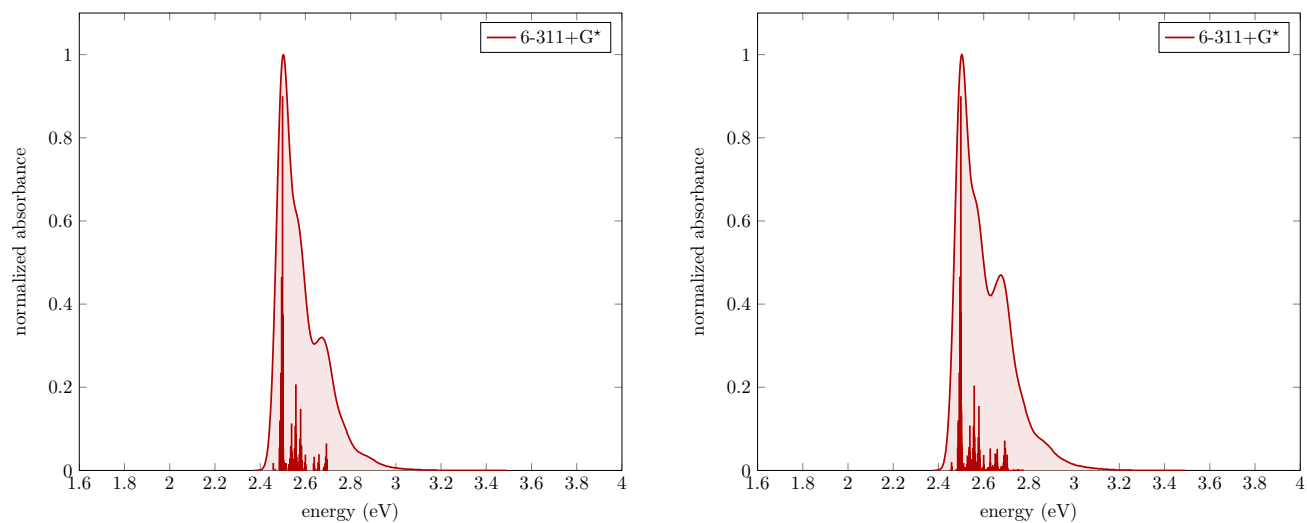
**Fig. S45** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31++G\*\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 5.5 6-311G



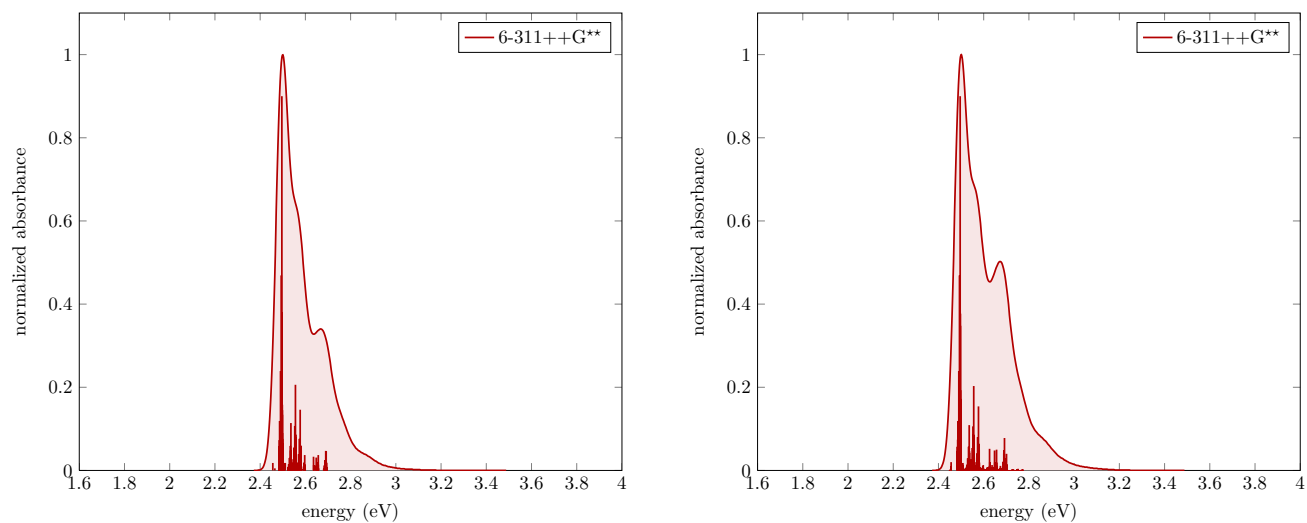
**Fig. S46** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-311G level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM =  $250\text{ cm}^{-1}$ ) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 5.6 6-311+G\*



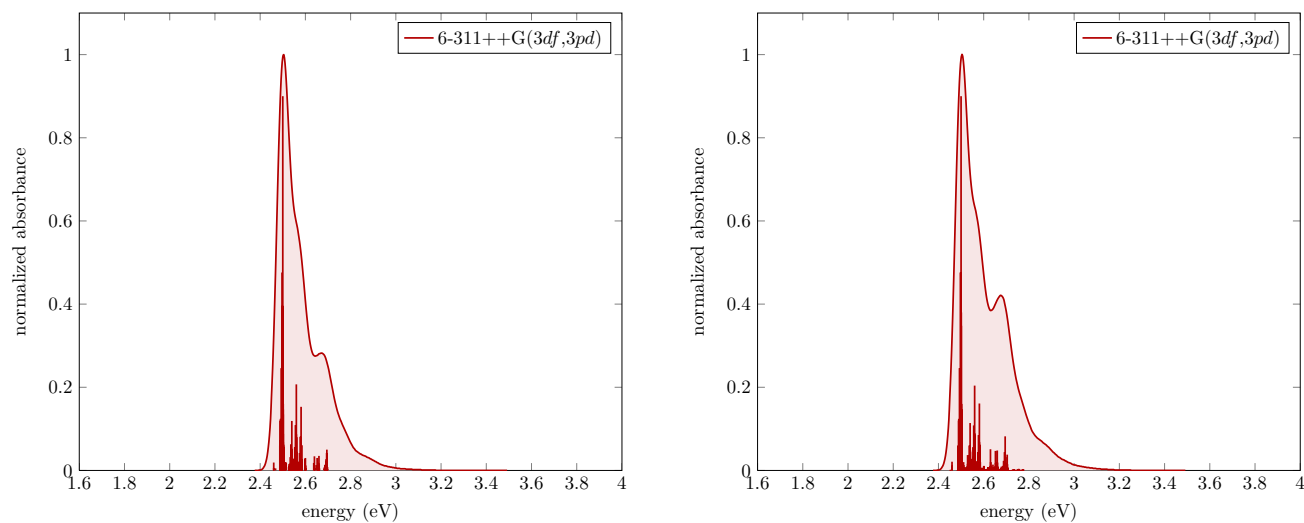
**Fig. S47** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-311+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 5.7 6-311++G\*\*



**Fig. S48** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-311++G\*\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 5.8 6-311++G(3df,3pd)



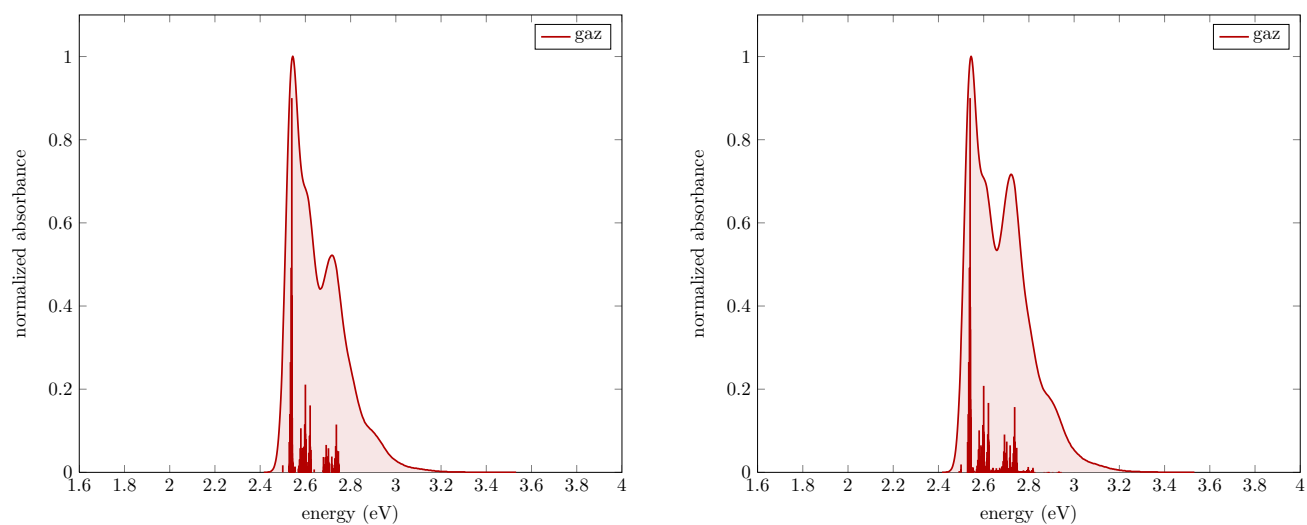
**Fig. S49** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-311++G(3df,3pd) level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.



# Solvent Benchmark

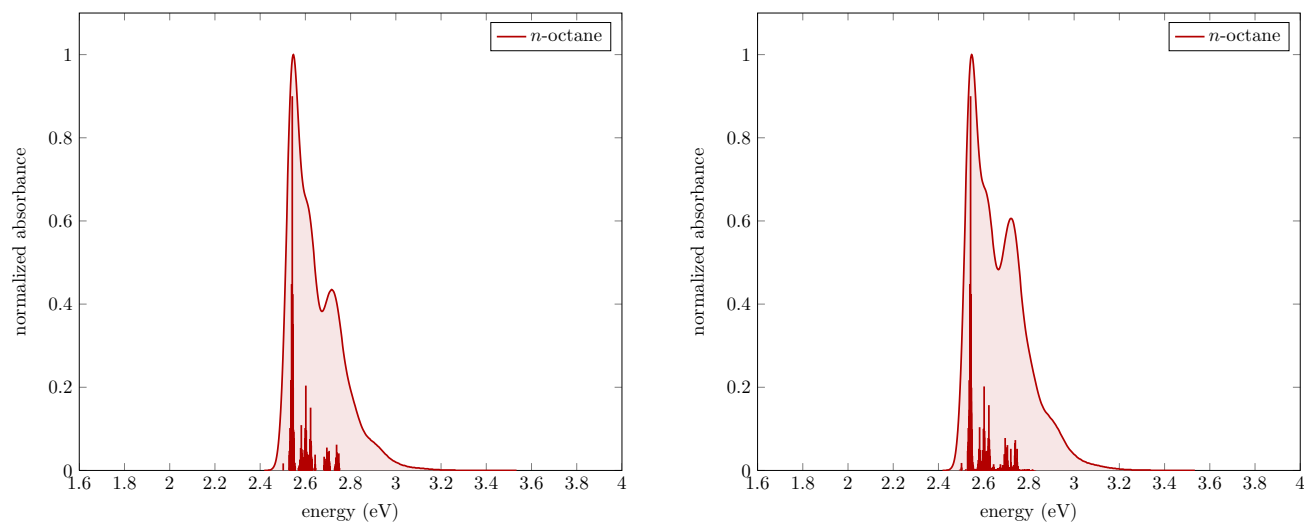
## 6 TATA

### 6.1 gaz



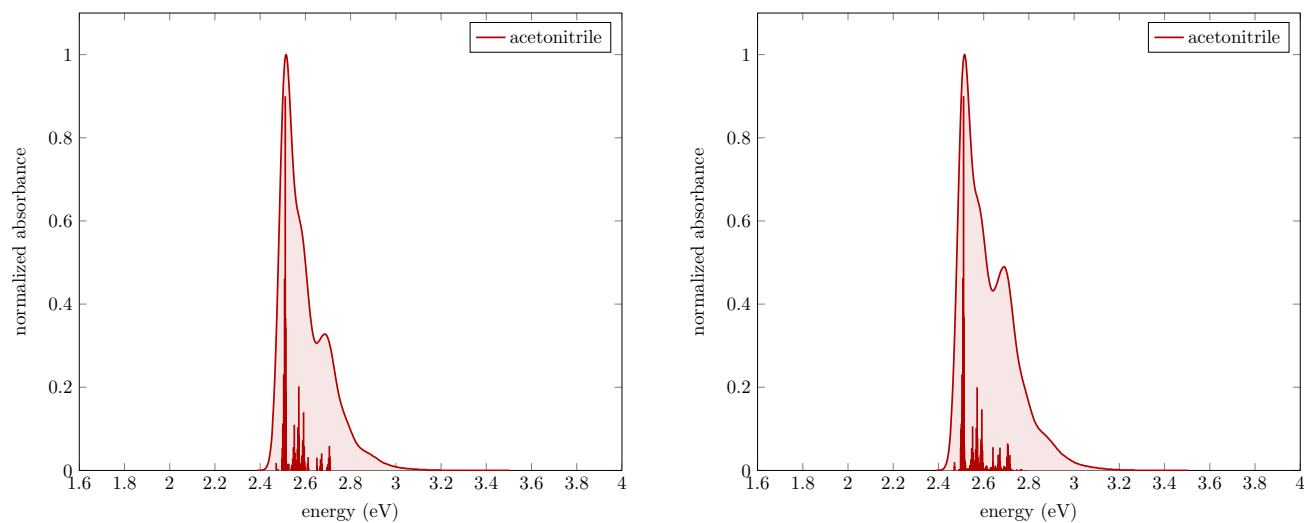
**Fig. S50** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in gaz solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 6.2 *n*-octane



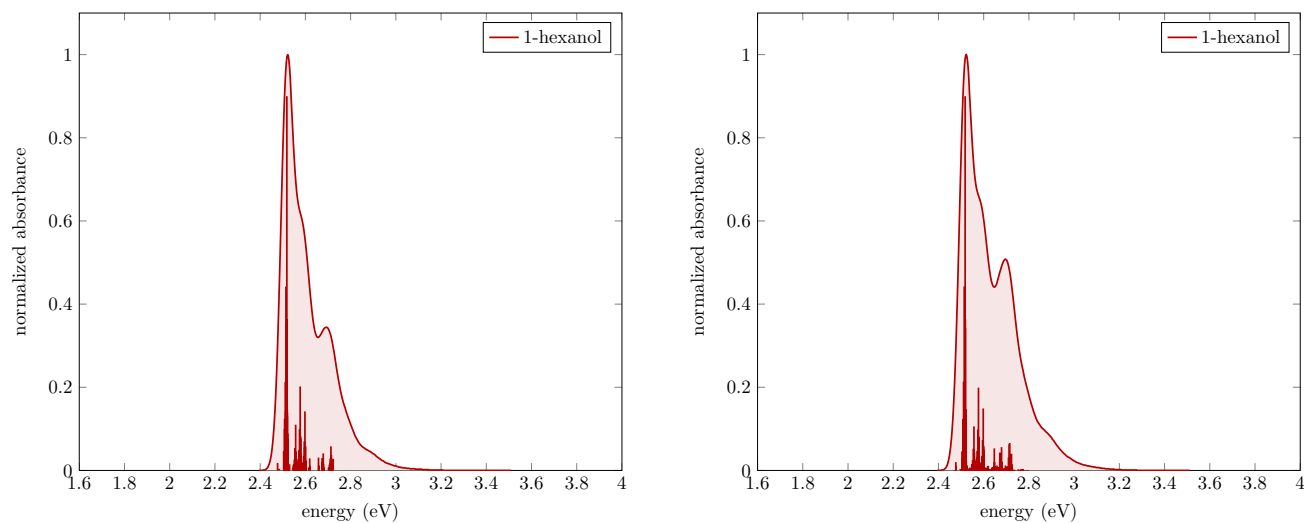
**Fig. S51** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in *n*-octane solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

### 6.3 acetonitrile



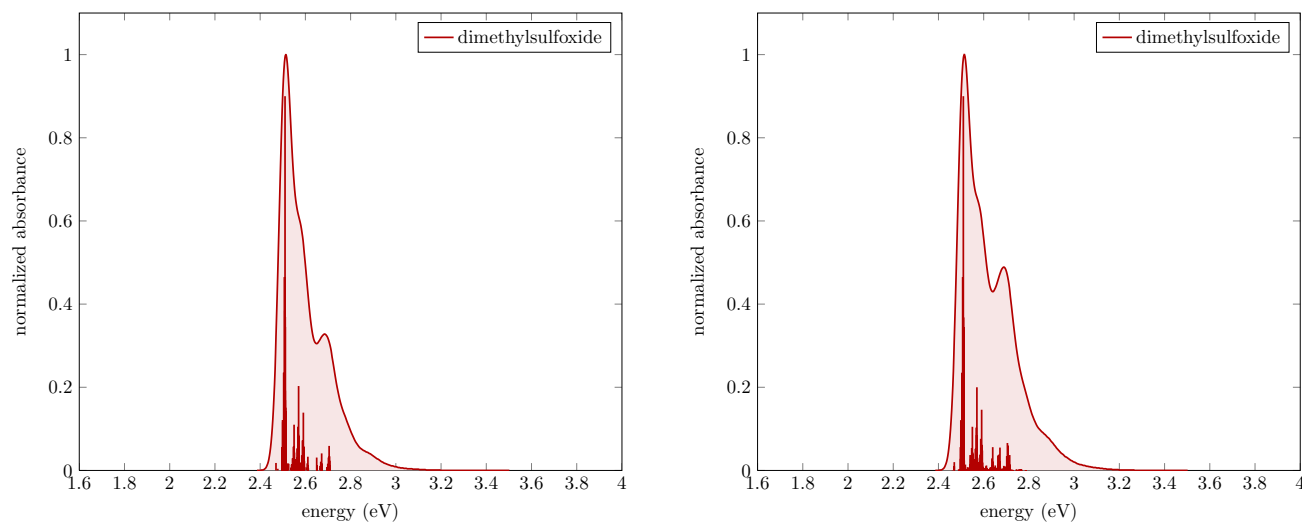
**Fig. S52** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in acetonitrile solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 6.4 1-hexanol



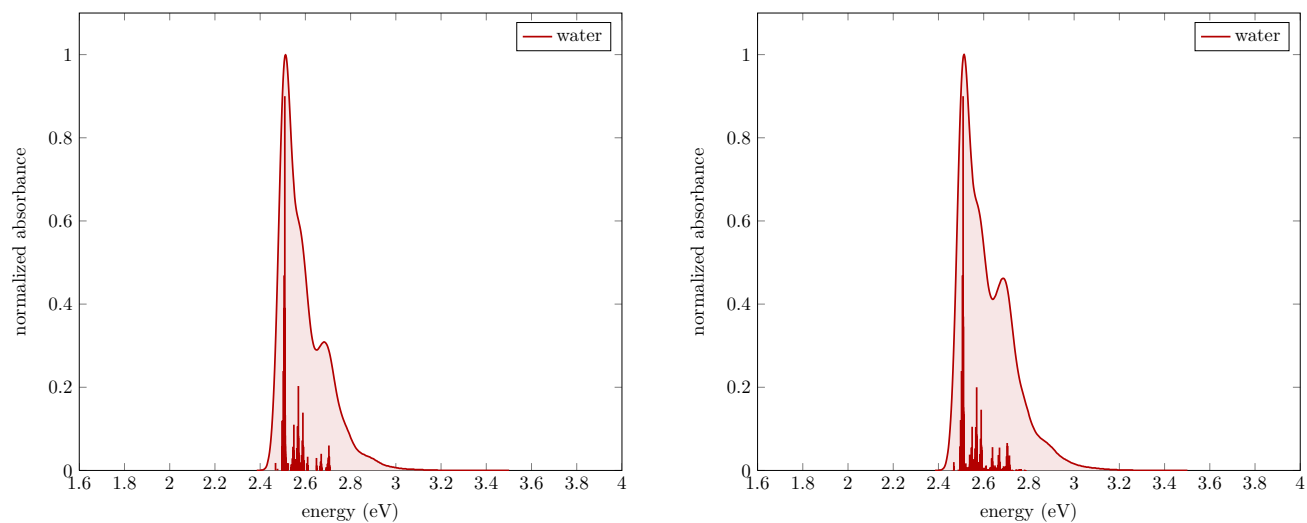
**Fig. S53** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in 1-hexanol solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM =  $250\text{ cm}^{-1}$ ) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 6.5 dimethylsulfoxide



**Fig. S54** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in dimethylsulfoxide solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM = 250 cm<sup>-1</sup>) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.

## 6.6 water



**Fig. S55** UV/vis absorption spectra of TATA computed within the (left) Franck-Condon and (right) Franck-Condon-Herzberg-Teller approximations at PBE0/6-31+G\* level of theory in water solvent. The envelope of each spectrum is obtained by a Gaussian convolution (FWHM =  $250\text{ cm}^{-1}$ ) of the vibronic transitions (sticks) between the  $S_0$  and  $S_1$  electronic states.