Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information for

The Influences of dynamic and static disorders on carrier mobility of BTBT-C12 derivatives: a multiscale computational study

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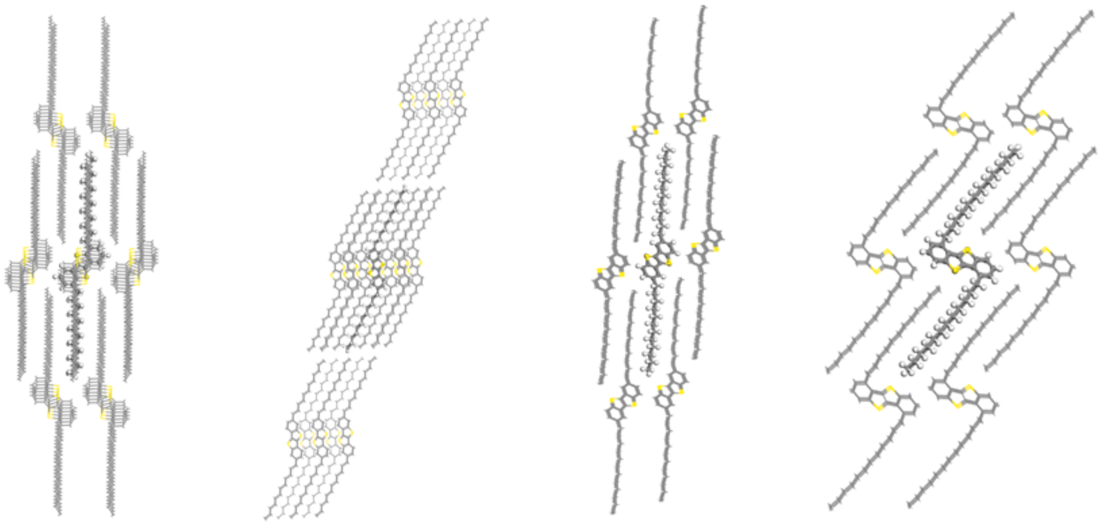


BTBT1 **BTBT2** **BTBT3** **BTBT4**

Figure S1. The ONIOM model for crystal structure calculations. The central molecule is represented by balls (QM part), and the surrounding molecules are represented by wireframes (MM part).

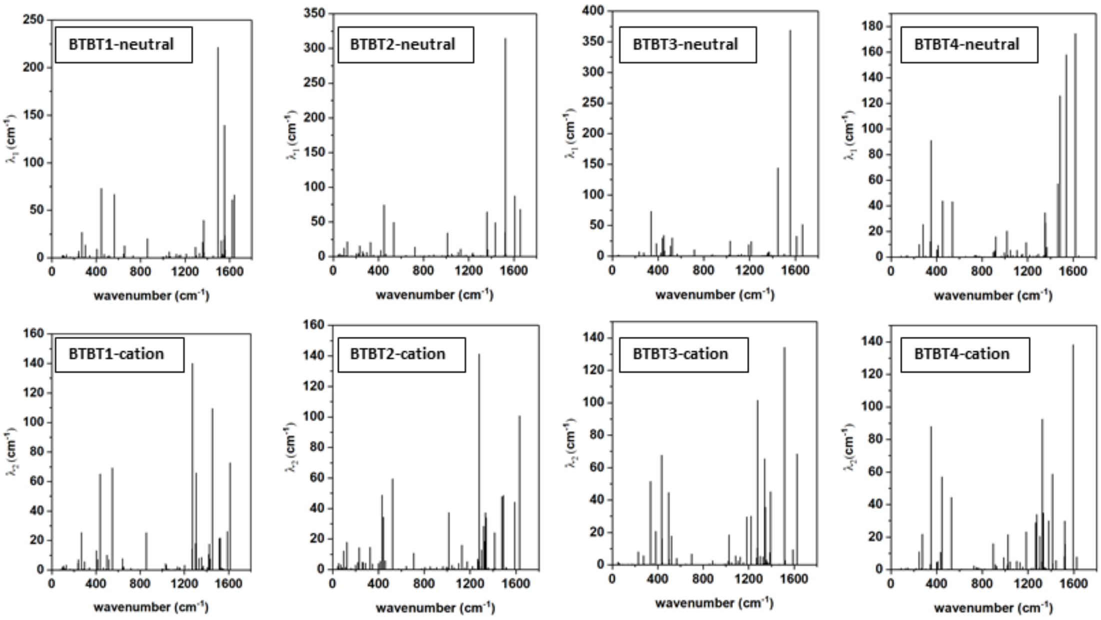


Figure S2. Contribution of vibrational modes to geometry relaxation for the neutral and charged states of four BTBT isomers in crystal state, which is calculated at the B3LYP 6-31G*/UFF level.

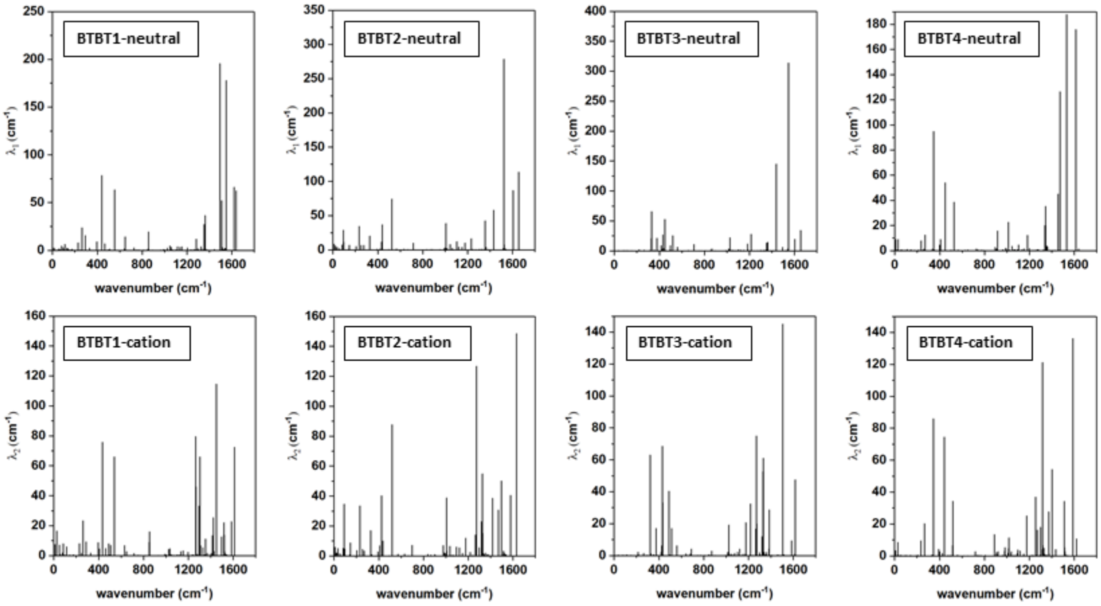
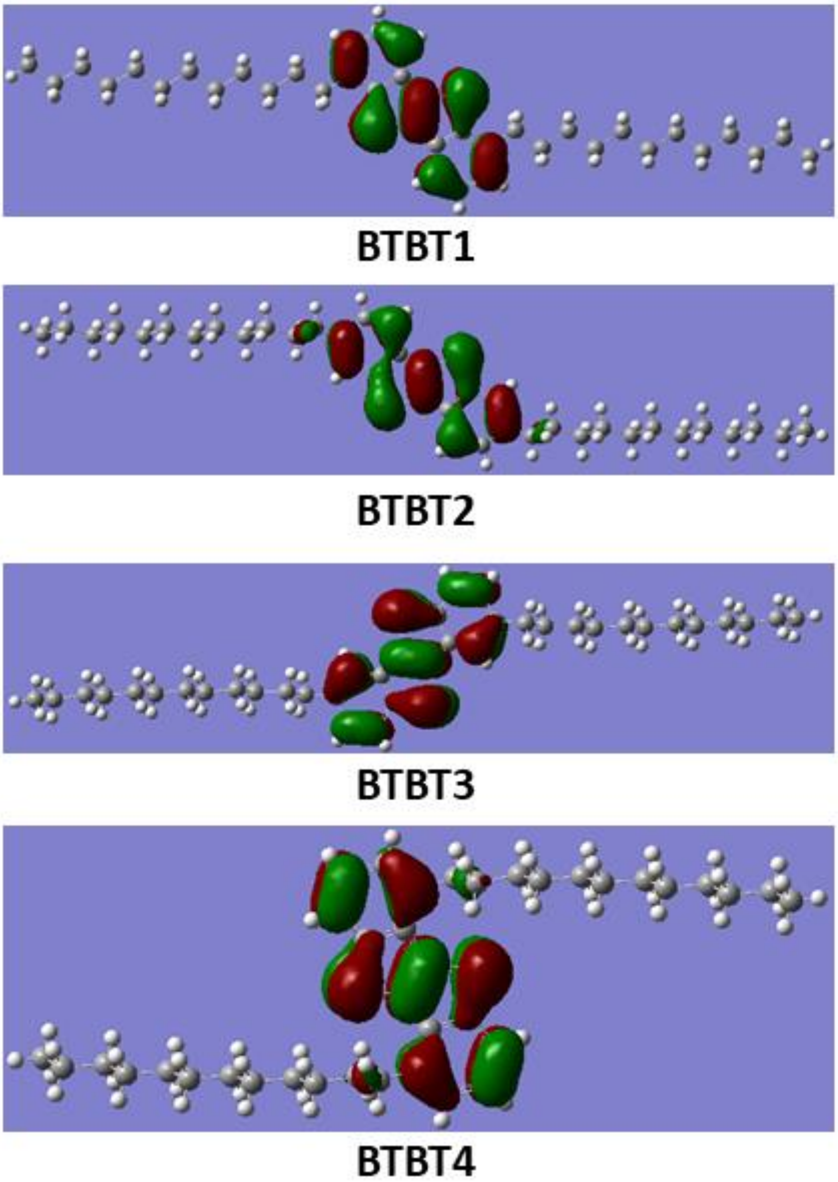


Figure S3. Contribution of vibrational modes to geometry relaxation for the neutral and charged states of four BTBT isomers in gas state, which is calculated at the B3LYP 6-31G* level.



BTBT1

BTBT2

BTBT3

BTBT4

Figure S4. HOMO orbitals for the four BTBT isomers calculated at the PW91PW91/6-31G* level.

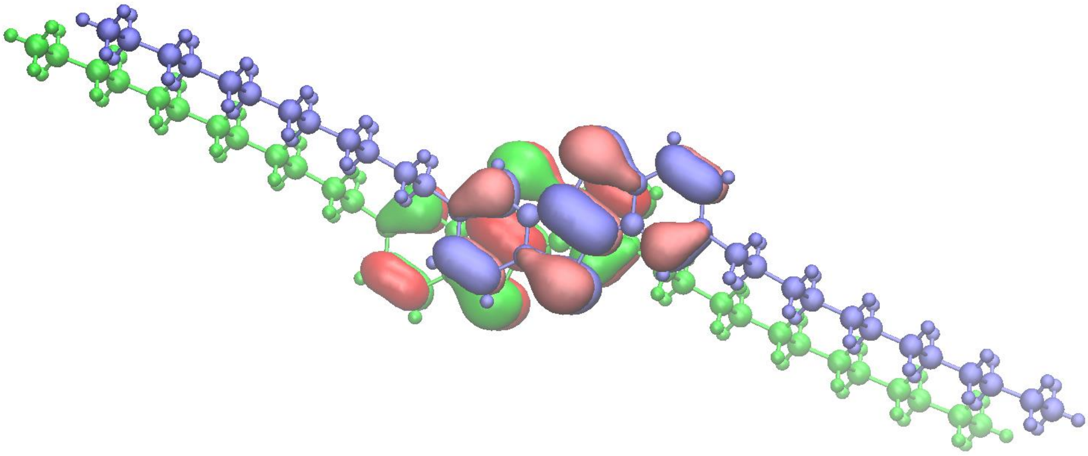


Figure S5. Overlap of HOMO orbitals for the two D1 molecules of BTBT3.

Table S1. The positional parameters for the nearest in-plane dimers (only one dimer is listed for the dimers with equivalent related positions), including distance of center of mass (COM) of benzothiophene ring (d_{COM}), slip distance of benzothiophene along the long axis (d_L) and the short axis (d_S), and angles between benzothiophene rings (angle). The detailed parameter definition is shown in Figure S4.

		D1	D2	D3
BTBT1	d_{COM} (**Å**)	4.08	9.01	9.89
	d_L (**Å**)	1.7	5.79	7.5
	d_S (**Å**)	0.71	6.76	6.05
	angle (°)	0.0	0.0	0.0
	Transfer integral (meV)	30.88	0.44	5.38
BTBT2	d_{COM} (**Å**)	4.86	5.86	
	d_L (**Å**)	0.06	0.16	
	d_S (**Å**)	4.34	5.26	
	angle (°)	52.6	0.0	
	Transfer integral (meV)	36.90	53.48	
BTBT3	d_{COM} (**Å**)	4.72	8.48	
	d_L (**Å**)	3.17	3.36	
	d_S (**Å**)	0.28	5.17	
	angle (°)	0.0	84.6	
	Transfer integral (meV)	5.70	21.33	
BTBT4	d_{COM} (**Å**)	4.65	10.39	
	d_L (**Å**)	1.61	10.01	
	d_S (**Å**)	2.62	1.24	
	angle (°)	0.0	83.0	
	Transfer integral (meV)	150.34	5.22	

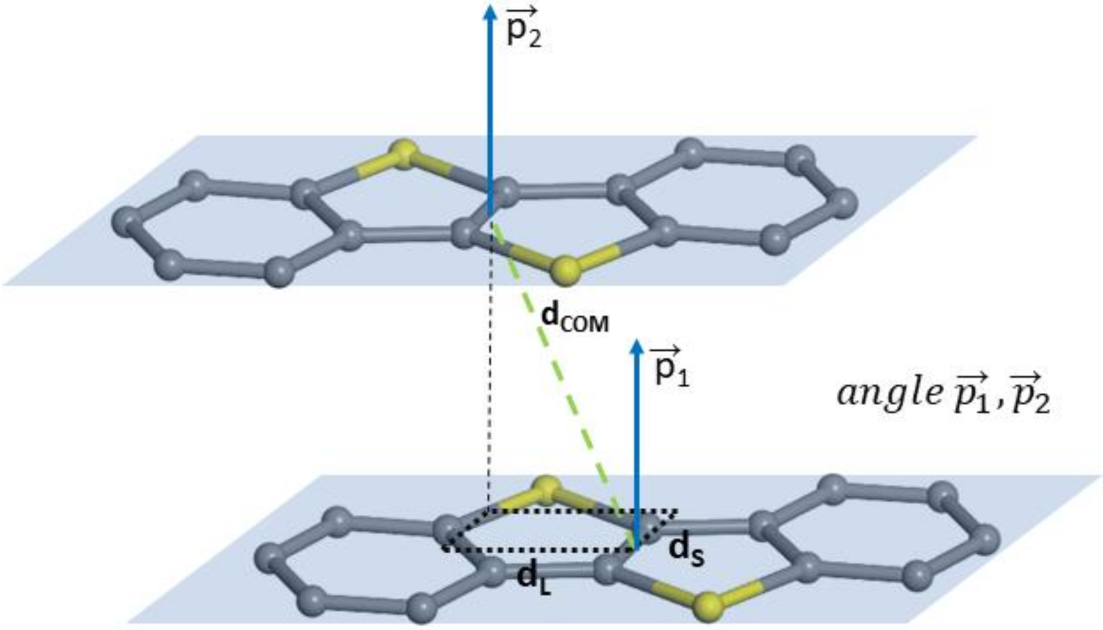


Figure S6. Detailed parameter definition for d_{COM}, d_L, d_S, and angle.

Table S2. Transfer integrals (at the PW91PW91/6-31G* level) of the nearest in-plane hopping pathways (Figure 2) for the four BTBT isomer crystals, and transfer rates calculated by Marcus theory and quantum nuclear tunneling method. The unit is in meV and s^{-1}.

		D1	D2	D3
Transfer integral (meV)	BTBT1	30.88	0.44	5.38
	BTBT2	36.90	53.48	
	BTBT3	5.70	21.33	
	BTBT4	150.34	5.22	
Marcus (1/s)	BTBT1	3.85E+12	7.92E+08	1.17E+11
	BTBT2	4.83E+12	1.01E+13	
	BTBT3	1.26E+11	1.76E+12	
	BTBT4	8.63E+13	1.04E+11	
Quantum Nuclear Tunneling (1/s)	BTBT1	1.86E+13	3.84E+09	5.65E+11
	BTBT2	1.52E+13	3.19E+13	
	BTBT3	3.74E+11	5.23E+12	
	BTBT4	3.95E+14	4.76E+11	

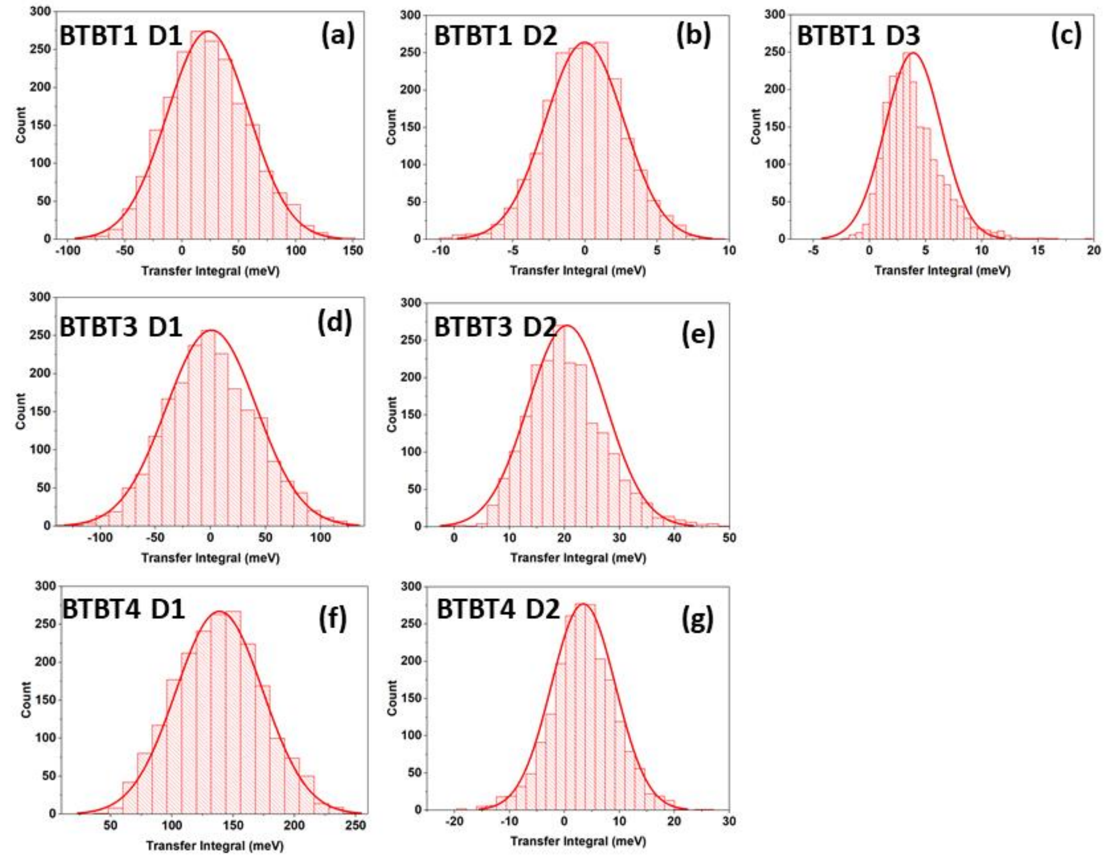
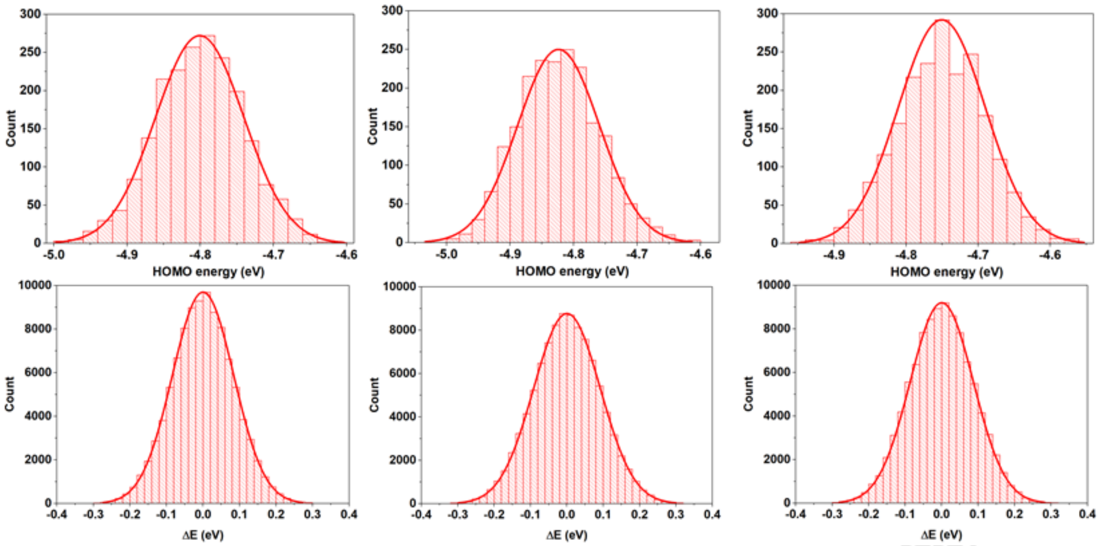


Figure S7. Transfer integral distributions for selected dimers of BTBT1, BTBT3 and BTBT4.

Table S3. Motilities for BTBT isomer crystals with dynamic transfer integral disorder by using Marcus theory, and ratio of mobility with disorder to mobility without disorder (unit in $cm^2\ V^{-1}\ s^{-1}$).

	μ(V-disorder)	μ(V-disorder)/ μ(disorder-free)
BTBT1	0.04	0.71
BTBT2	0.59	1.06
BTBT3	0.37	1.21
BTBT4	2.04	0.97



BTBT1 **BTBT3** **BTBT4**

Figure S8. Distributions of HOMO energies and HOMO energy differences for BTBT1, BTBT3, and BTBT4.

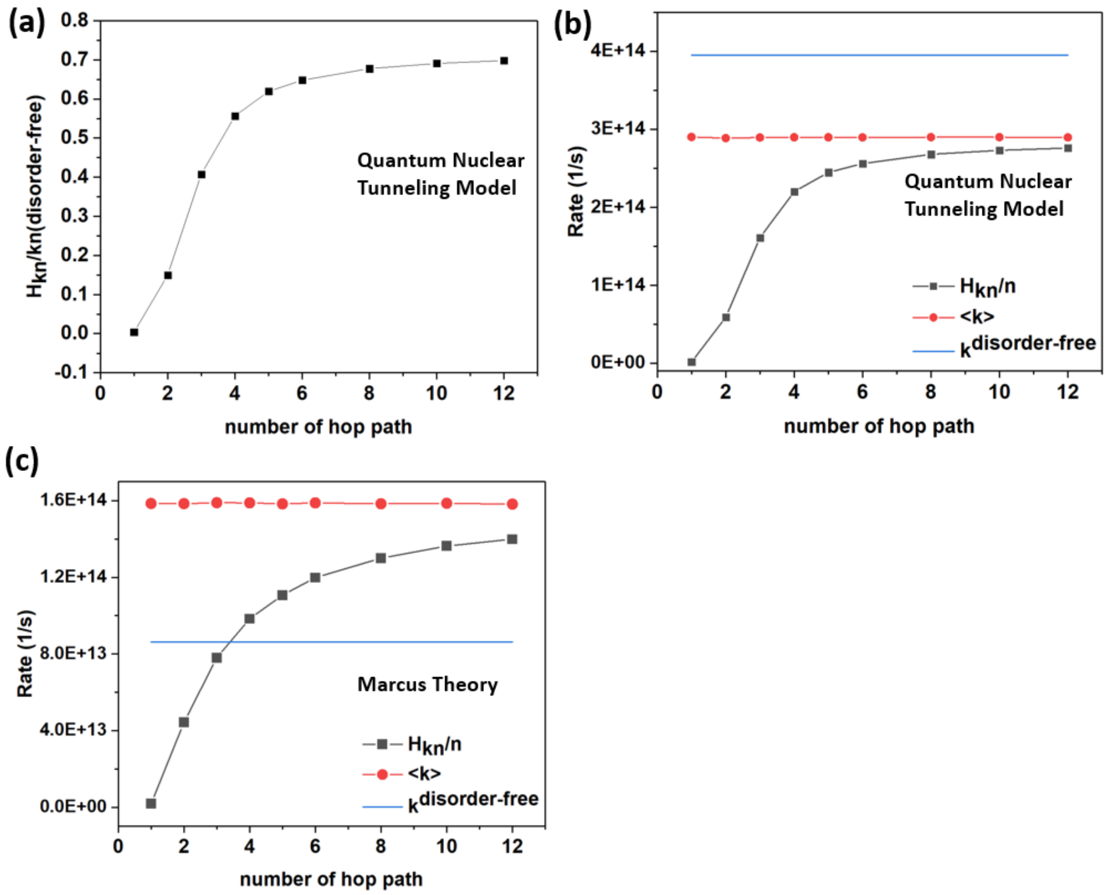


Figure S9. By using hop rates of BTBT4 D1 and after running 100,000 hopping steps, and then calculate the various parameters. (a)/(b): based on nuclear tunneling model, ratio of H_{kn} (harmonic mean of kn) to kn$^{disorder\text{-}free}$, as well as H_{kn}/n and <k> (mean of k) as a function of the number of hop paths *n*; (c): based on Marcus theory, H_{kn}/n and <k> as a function of the number of hop paths *n*.

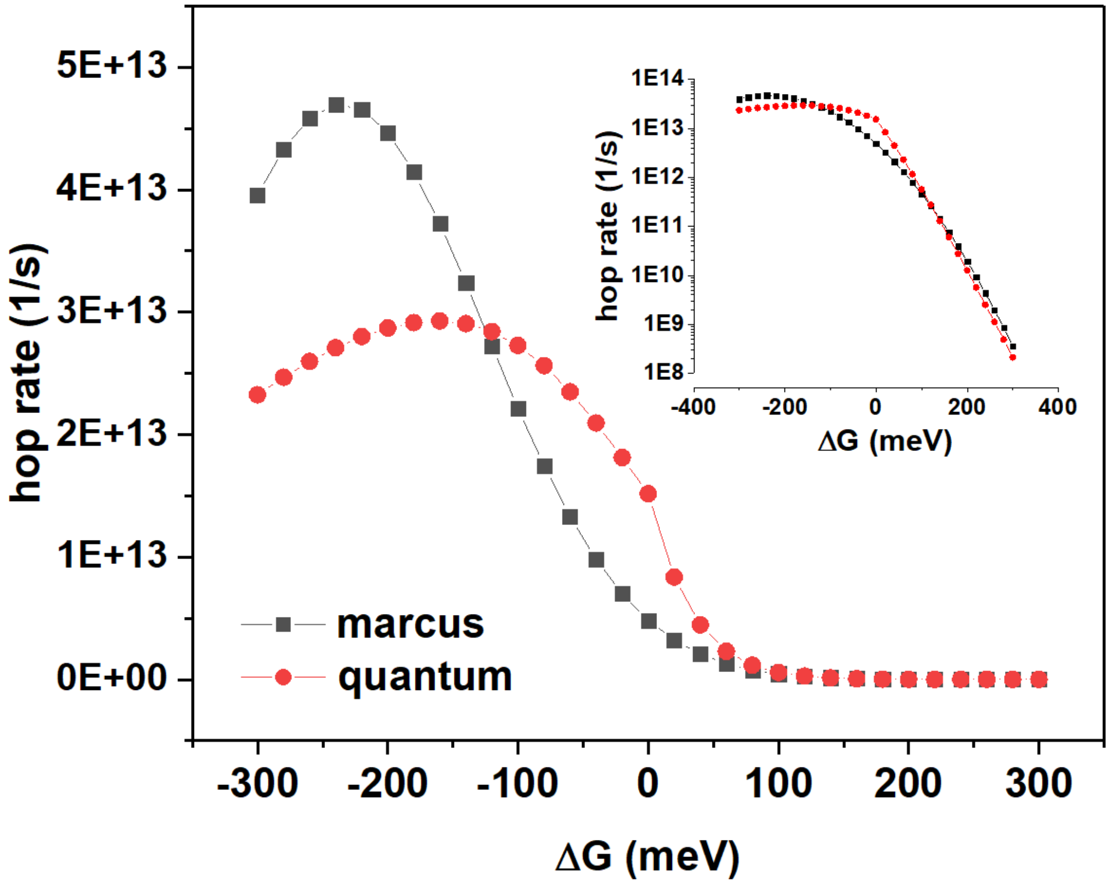


Figure S10. Hop rates of BTBT2 D2 calculated by using Marcus theory and quantum nuclear tunneling method as a function of site energy difference (ΔG). The insert is shown in logarithm of y axis.

Table S4. Motilities calculated by using Marcus theory for the four BTBT isomer crystals with dynamic site energy disorder, and ratio of mobility with disorder to mobility without disorder.

	μ(SE-disorder) (cm^2/Vs)	μ(SE-disorder)/ μ(disorder-free)
BTBT1	0.08	0.87
BTBT2	1.04	1.43
BTBT3	0.04	1.16
BTBT4	1.40	0.53

Table S5. The ∆G values with defect to be of the BTBT-oxide or BTBT-dioxide, which is calculated as follows: for example, for the BTBT-oxides, the energy of cation BTBT plus the energy of neutral BTBT-oxides, then minus the energy of neutral BTBT and the energy of cation BTBT-oxides.

	∆G (meV)	
	BTBT-oxides	BTBT-dioxides
BTBT1	-604	-1003
BTBT2	-552	-895
BTBT3	-574	-940
BTBT4	-681	-1035