Supporting information for:

Thermoelasticity in Organic Semiconductors Determined with Terahertz Spectroscopy and Quantum Quasi-Harmonic Simulations

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1 Rubrene

1.1 Structural Parameters

Table S1: Theoretical cell coordinates for rubrene corresponding to $0\,\mathrm{K}$ (no ZPE), calculated with the 6-31G(d,p)/PBE-D3 method. The crystal structure has symmetry $Cmca$, with corresponding cell parameters of: $a = 26.3024$ Å, $b = 7.1293$ Å, $c = 13.9250$ Å, and $\alpha = \beta = \gamma = 90°$

Atom	X/A	Y/B	Z/C
C	0.0280	0.0000	0.0000
C	0.0545	-0.1460	0.0495
C	0.0276	-0.3005	0.0882
C	0.0533	-0.4520	0.1361
C	0.0270	0.4051	0.1802
C	0.1097	-0.1335	0.0745
C	0.1460	-0.2553	0.0341
C	0.1962	-0.2562	0.0671
C	0.2115	-0.1322	0.1399
C	0.1759	-0.0089	0.1803
C	0.1254	-0.0112	0.1482
H	0.0948	-0.4510	0.1386
H	0.0474	0.2936	0.2180
H	0.1344	-0.3491	-0.0241
H	0.2238	-0.3538	0.0361
H	0.2511	-0.1327	0.1645
H	0.1870	0.0890	0.2373
H	0.0974	0.0833	0.1803

Table S2: Theoretical cell coordinates for rubrene corresponding to $0\,K$, calculated with the 6-31G(d,p)/PBE-D3 method. The crystal structure has symmetry *Cmca*, with corresponding cell parameters of: $a = 26.4074$ Å, $b = 7.1469$ Å, $c = 14.0632$ Å, and $\alpha = \beta = \gamma = 90°$

Atom	X/A	Y/B	Z/C
C	0.0279	0.0000	0.0000
C	0.0543	-0.1452	0.0494
C	0.0275	-0.2988	0.0883
C	0.0531	-0.4494	0.1362
C	0.0269	0.4085	0.1803
C	0.1093	-0.1324	0.0743
C	0.1456	-0.2533	0.0343
C	0.1956	-0.2539	0.0672
C	0.2106	-0.1307	0.1397
C	0.1749	-0.0084	0.1798
C	0.1247	-0.0109	0.1477
H	0.0944	-0.4484	0.1386
H	0.0472	0.2976	0.2179
H	0.1342	-0.3468	-0.0235
H	0.2232	-0.3506	0.0364
H	0.2499	-0.1310	0.1645
H	0.1858	0.0889	0.2365
H	0.0966	0.0826	0.1795

Table S3: Theoretical cell coordinates for rubrene corresponding to 100 K, calculated with the 6-31G(d,p)/PBE-D3 method. The crystal structure has symmetry *Cmca*, with corresponding cell parameters of: $a = 26.4315$ Å, $b = 7.1535$ Å, $c = 14.1023$ Å, and $\alpha = \beta = \gamma = 90°$

Atom	X/A	Y/B	Z/C
C	0.0279	0.0000	0.0000
C	0.0543	-0.1449	0.0494
C	0.0275	-0.2983	0.0883
C	0.0530	-0.4486	0.1363
C	0.0269	0.4096	0.1804
C	0.1092	-0.1320	0.0742
C	0.1455	-0.2528	0.0345
C	0.1955	-0.2530	0.0674
C	0.2104	-0.1297	0.1397
C	0.1747	-0.0076	0.1796
C	0.1245	-0.0103	0.1474
H	0.0943	-0.4476	0.1386
H	0.0472	0.2990	0.2180
H	0.1342	-0.3464	-0.0231
H	0.2231	-0.3495	0.0368
H	0.2497	-0.1297	0.1644
H	0.1855	0.0897	0.2361
H	0.0964	0.0830	0.1791

Table S4: Theoretical cell coordinates for rubrene corresponding to 200 K, calculated with the 6-31G(d,p)/PBE-D3 method. The crystal structure has symmetry *Cmca*, with corresponding cell parameters of: $a = 26.4788$ Å, $b = 7.1649$ Å, $c = 14.1800$ Å, and $\alpha = \beta = \gamma = 90°$

Atom	X/A	Y/B	Z/C
C	0.0279	0.0000	0.0000
C	0.0542	-0.1444	0.0494
C	0.0274	-0.2972	0.0884
C	0.0529	-0.4470	0.1363
C	0.0269	0.4117	0.1804
C	0.1090	-0.1313	0.0741
C	0.1454	-0.2517	0.0347
C	0.1952	-0.2516	0.0676
C	0.2100	-0.1286	0.1396
C	0.1742	-0.0069	0.1791
C	0.1241	-0.0098	0.1470
H	0.0942	-0.4460	0.1387
H	0.0471	0.3014	0.2180
H	0.1341	-0.3453	-0.0226
H	0.2229	-0.3478	0.0372
H	0.2491	-0.1284	0.1644
H	0.1850	0.0902	0.2354
H	0.0961	0.0831	0.1784

Table S5: Theoretical cell coordinates for rubrene corresponding to 300 K, calculated with the 6-31G(d,p)/PBE-D3 method. The crystal structure has symmetry *Cmca*, with corresponding cell parameters of: $a = 26.5250$ Å, $b = 7.1795$ Å, $c = 14.2851$ Å, and $\alpha = \beta = \gamma = 90°$

Atom	X/A	Y/B	Z/C
C	0.0278	0.0000	0.0000
C	0.0541	-0.1437	0.0494
C	0.0274	-0.2957	0.0886
C	0.0529	-0.4447	0.1366
C	0.0268	0.4147	0.1807
C	0.1088	-0.1303	0.0740
C	0.1452	-0.2505	0.0351
C	0.1949	-0.2503	0.0679
C	0.2095	-0.1275	0.1395
C	0.1738	-0.0061	0.1785
C	0.1238	-0.0090	0.1464
H	0.0940	-0.4437	0.1389
H	0.0471	0.3051	0.2183
H	0.1340	-0.3441	-0.0218
H	0.2226	-0.3463	0.0378
H	0.2486	-0.1273	0.1643
H	0.1844	0.0908	0.2345
H	0.0957	0.0837	0.1775

Table S6: Theoretical cell coordinates for rubrene corresponding to 300 K, calculated with the 6-31G(d,p)/PBE0-D3 method. The crystal structure has symmetry *Cmca*, with corresponding cell parameters of: $a = 26.5407$ Å, $b = 7.1277$ Å, $c = 14.1557$ Å, and $\alpha = \beta = \gamma = 90°$

Atom	X/A	Y/B	Z/C
C	0.0276	0.0000	0.0000
C	0.0536	-0.1443	0.0493
C	0.0271	-0.2962	0.0881
C	0.0525	-0.4465	0.1358
C	0.0267	0.4126	0.1793
C	0.1082	-0.1323	0.0737
C	0.1442	-0.2498	0.0325
C	0.1937	-0.2502	0.0647
C	0.2084	-0.1309	0.1377
C	0.1729	-0.0124	0.1791
C	0.1232	-0.0147	0.1476
H	0.0934	-0.4457	0.1380
H	0.0468	0.3021	0.2163
H	0.1330	-0.3412	-0.0252
H	0.2211	-0.3442	0.0329
H	0.2473	-0.1313	0.1621
H	0.1836	0.0817	0.2363
H	0.0954	0.0758	0.1805

1.2 Predicted Elastic Tensors

1.2.1 PBE

0 K (no ZPE)

$$c_{ij} = \begin{bmatrix} 28.530 & 8.480 & 9.390 & 0.000 & 0.000 & 0.000 \\ 8.480 & 19.74 & 11.690 & 0.000 & 0.000 & 0.000 \\ 9.390 & 11.690 & 13.650 & 0.000 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 8.470 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 3.500 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 0.000 & 3.710 \end{bmatrix} \text{GPa} \tag{1}$$

0 K

$$c_{ij} = \begin{bmatrix} 24.720 & 7.190 & 7.940 & 0.000 & 0.000 & 0.000 \\ 7.190 & 17.640 & 10.310 & 0.000 & 0.000 & 0.000 \\ 7.940 & 10.310 & 11.830 & 0.000 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 7.590 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 3.310 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 0.000 & 3.150 \end{bmatrix} \text{GPa} \tag{2}$$

100 K

$$c_{ij} = \begin{bmatrix} 23.693 & 6.768 & 7.584 & 0.000 & 0.000 & 0.000 \\ 6.768 & 17.033 & 9.961 & 0.000 & 0.000 & 0.000 \\ 7.584 & 9.961 & 11.367 & 0.000 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 7.322 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 3.255 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 0.000 & 3.003 \end{bmatrix} \text{GPa} \tag{3}$$

200 K

$$c_{ij} = \begin{bmatrix} 21.715 & 6.137 & 6.920 & 0.000 & 0.000 & 0.000 \\ 6.137 & 15.931 & 9.270 & 0.000 & 0.000 & 0.000 \\ 6.920 & 9.270 & 10.451 & 0.000 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 6.827 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 3.155 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 0.000 & 2.718 \end{bmatrix} \text{GPa} \tag{4}$$

300 K

$$c_{ij} = \begin{bmatrix} 19.500 & 5.530 & 6.240 & 0.000 & 0.000 & 0.000 \\ 5.530 & 14.470 & 8.210 & 0.000 & 0.000 & 0.000 \\ 6.240 & 8.210 & 9.040 & 0.000 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 6.170 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 2.990 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 0.000 & 2.390 \end{bmatrix} \text{GPa} \tag{5}$$

1.2.2   PBE0

300 K

$$c_{ij} = \begin{bmatrix} 18.645 & 5.713 & 6.148 & 0.000 & 0.000 & 0.000 \\ 5.713 & 15.541 & 8.544 & 0.000 & 0.000 & 0.000 \\ 6.148 & 8.544 & 10.121 & 0.000 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 6.842 & 0.000 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 3.355 & 0.000 \\ 0.000 & 0.000 & 0.000 & 0.000 & 0.000 & 2.450 \end{bmatrix} \text{GPa} \tag{6}$$
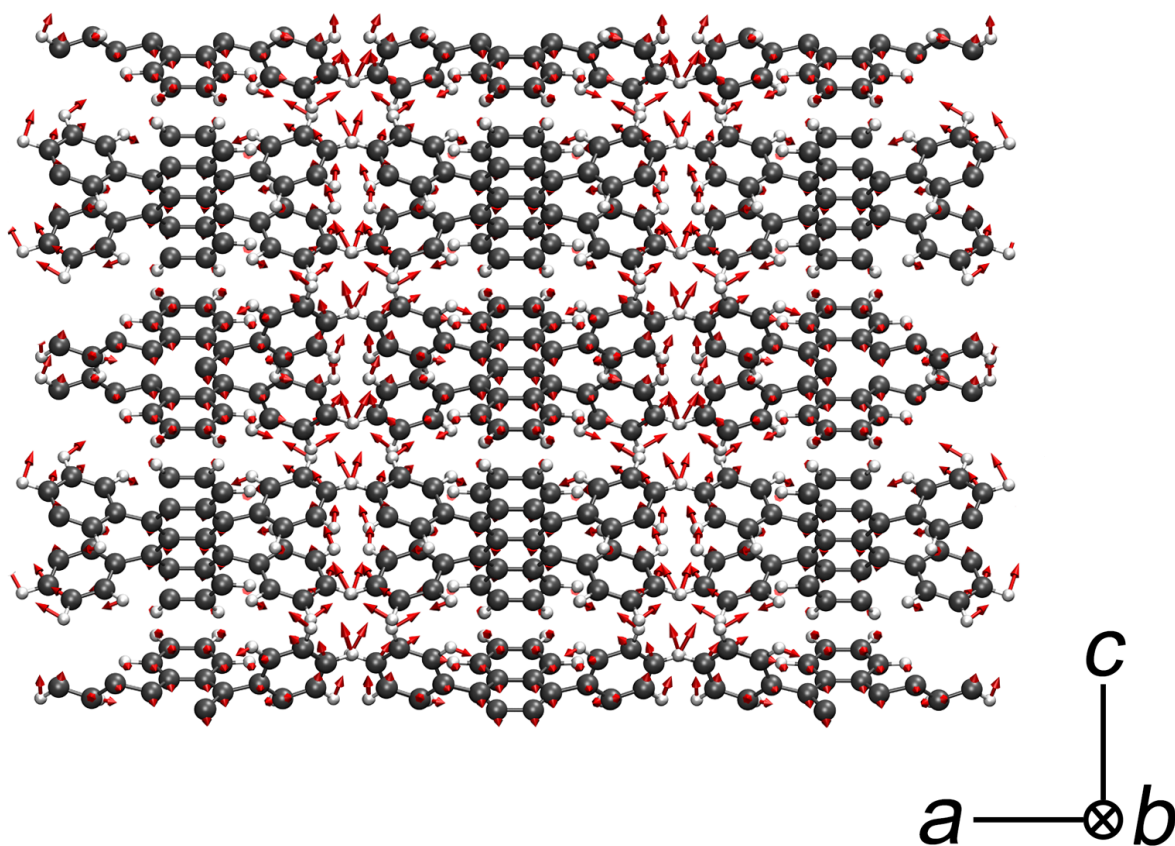
1.3   Vibrational Mode Types

Figure S1: Calculated molecular motions of rubrene low-frequency vibrational mode with vibrational transition frequency of $36.50\,\mathrm{cm}^{-1}$, with atomic displacements illustrated with red arrows.
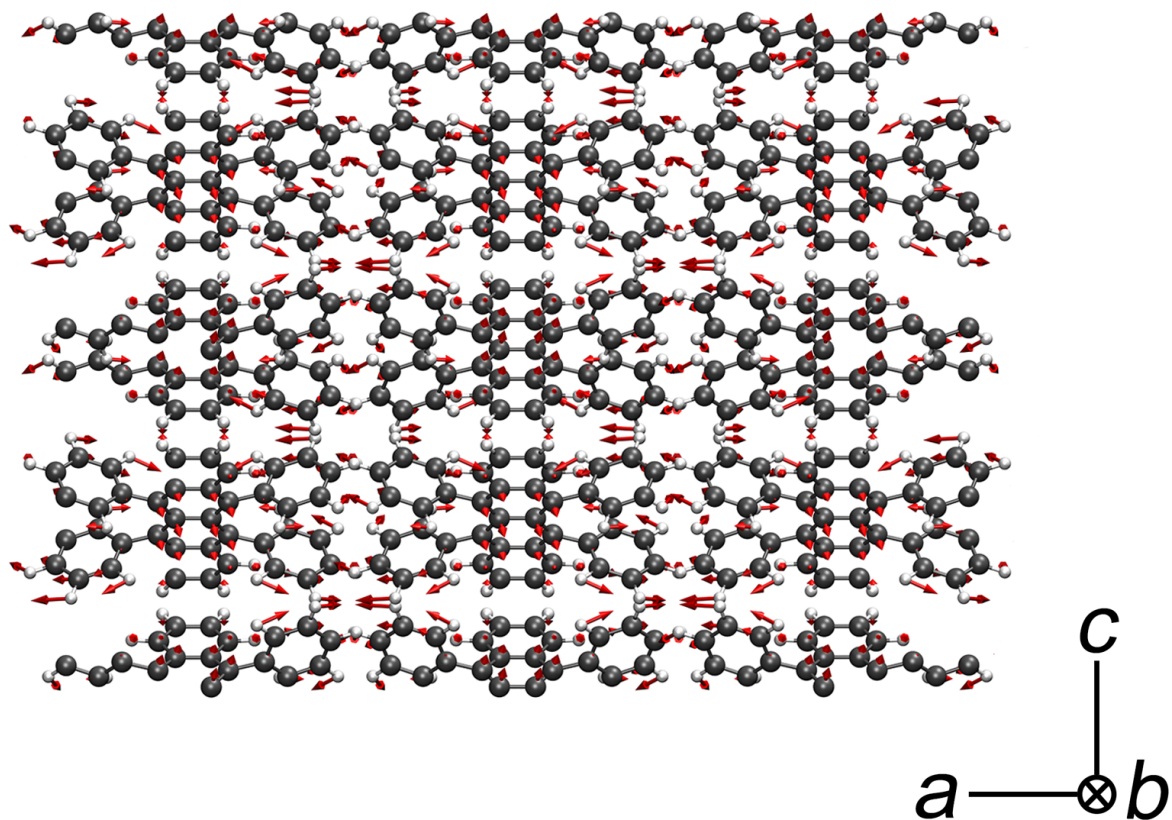
Figure S2: Calculated molecular motions of rubrene low-frequency vibrational mode with vibrational transition frequency of $49.67\,\mathrm{cm}^{-1}$, with atomic displacements illustrated with red arrows.
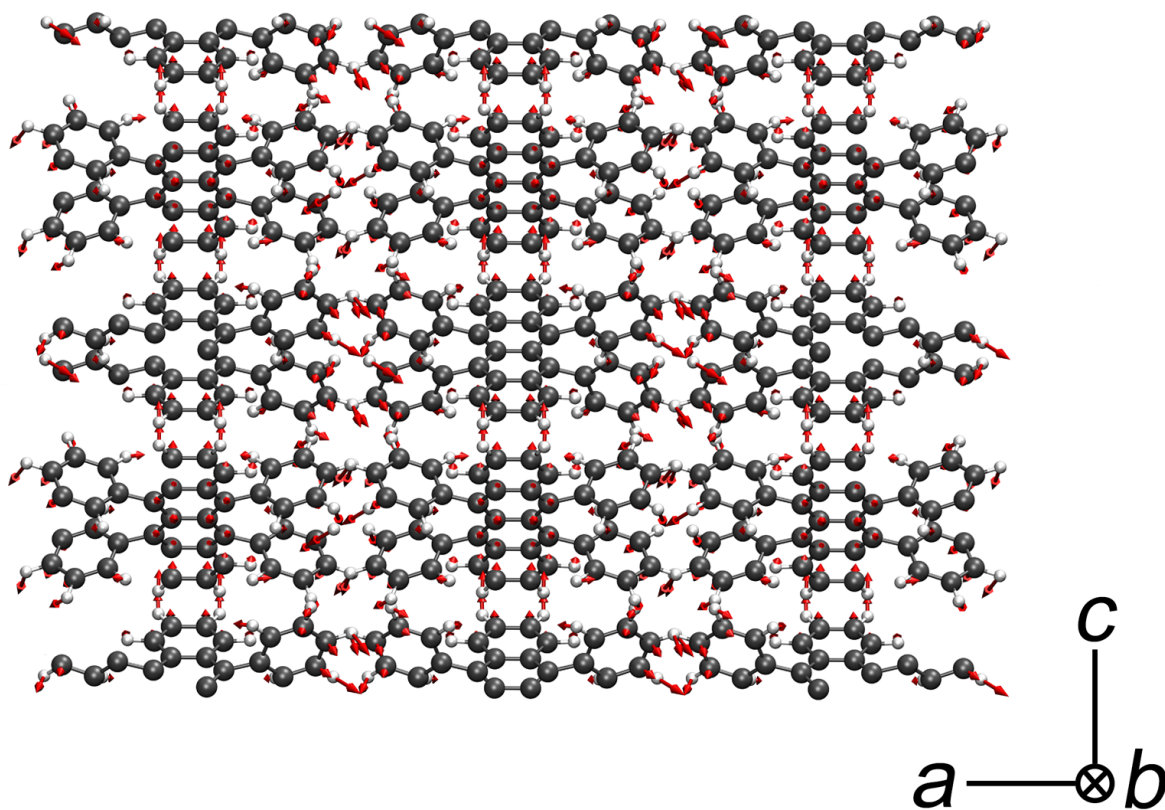
Figure S3: Calculated molecular motions of rubrene low-frequency vibrational mode with vibrational transition frequency of $74.62\,\mathrm{cm}^{-1}$, with atomic displacements illustrated with red arrows.

2 BTBT

2.1 Structural Parameters

Table S7: Experimental cell coordinates for BTBT, collected at $100\,\mathrm{K}$. The crystal structure has symmetry $P2_1/c$, with corresponding cell parameters of: $a = 11.8134$ Å, $b = 5.8786$ Å, $c = 7.9700$ Å, $\alpha = \gamma = 90°$, and $\beta = 106.1180°$.

Atom	X/A	Y/B	Z/C
S	0.4204	0.2135	0.3590
C	0.6510	0.4119	0.5174
C	0.5248	0.4062	0.4724
C	0.6927	0.6143	0.6109
C	0.7319	0.2581	0.4813
C	0.8130	0.6627	0.6694
C	0.8915	0.5080	0.6321
C	0.8512	0.3076	0.5387
H	0.907	0.205	0.514
H	0.8388	0.799	0.734
H	0.7057	0.128	0.422
H	0.9724	0.542	0.673

Table S8: Experimental cell coordinates for BTBT, collected at $200\,\text{K}$. The crystal structure has symmetry $P2_1/c$, with corresponding cell parameters of: $a = 11.8458$ Å, $b = 5.8857$ Å, $c = 8.0368$ Å, $\alpha = \gamma = 90°$, and $\beta = 106.3150°$.

Atom	X/A	Y/B	Z/C
S	0.5796	0.7844	0.6418
C	0.5246	0.4070	0.4721
C	0.6506	0.4128	0.5171
C	0.6922	0.6137	0.6109
C	0.8123	0.6615	0.6696
C	0.7311	0.2600	0.4806
C	0.8500	0.3093	0.5381
C	0.8903	0.5083	0.6318
H	0.7042	0.1330	0.4190
H	0.8387	0.7980	0.7330
H	0.9712	0.5420	0.6720
H	0.9043	0.2090	0.5110

Table S9: Experimental cell coordinates for BTBT, collected at 300 K. The crystal structure has symmetry $P2_1/c$, with corresponding cell parameters of: $a = 11.8780$ Å, $b = 5.8820$ Å, $c = 8.0920$ Å, $\alpha = \gamma = 90.0000$ °, and $\beta = 106.4890°$.

Atom	X/A	Y/B	Z/C
S	0.5795	0.7826	0.6425
C	0.3083	0.3872	0.3883
C	0.3491	0.5871	0.4836
C	0.4750	0.5921	0.5276
C	0.2692	0.7387	0.5197
C	0.1510	0.6875	0.4620
C	0.1862	0.3432	0.3309
C	0.1100	0.4935	0.3680
H	0.2949	0.8715	0.5812
H	0.0973	0.7869	0.4873
H	0.1583	0.2122	0.2682
H	0.0297	0.4656	0.3299

Table S10: Theoretical cell coordinates for BTBT corresponding to 0 K, without inclusion of zero point energy. The crystal structure has symmetry $P2_1/c$, with corresponding cell parameters of: $a = 11.8080$ Å, $b = 5.8280$ Å, $c = 7.8291$ Å, $\alpha = \gamma = 90°$, and $\beta = 105.9956°$

Atom	X/A	Y/B	Z/C
S	0.4202	-0.2065	0.3590
C	0.3063	-0.3833	0.3881
C	0.1859	-0.3339	0.3300
H	0.1554	-0.1760	0.2574
C	0.1070	-0.4910	0.3676
H	0.1308	-0.4533	0.3250
C	0.1475	0.3052	0.4612
H	0.8460	0.1858	0.4912
C	0.2675	0.2544	-0.4825
H	0.2979	0.0965	-0.4103
C	0.3487	0.4099	0.4817
C	0.4746	0.4040	-0.4729

Table S11: Theoretical cell coordinates for BTBT corresponding to 0 K, with the inclusion of zero point energy. The crystal structure has symmetry $P2_1/c$, with corresponding cell parameters of: $a = 11.8578$ Å, $b = 5.8588$ Å, $c = 7.8904$ Å, $\alpha = \gamma = 90°$, and $\beta = 106.0777°$

Atom	X/A	Y/B	Z/C
C	0.4747	0.4048	-0.4728
C	0.3492	0.4108	0.4820
H	0.2988	0.0996	-0.4098
C	0.2681	0.2563	-0.4820
H	0.0862	0.1880	0.4915
C	0.1488	0.3066	0.4616
H	0.0148	-0.4538	0.3254
C	0.1085	-0.4910	0.3680
H	0.1565	-0.1783	0.2578
C	0.1870	-0.3351	0.3304
C	0.3069	-0.3840	0.3884
S	0.4204	-0.2086	0.3589

Table S12: Theoretical cell coordinates for BTBT corresponding to 100 K. The crystal structure has symmetry $P2_1/c$, with corresponding cell parameters of: $a = 11.8740$ Å, $b = 5.8703$ Å, $c = 7.9096$ Å, $\alpha = \gamma = 90°$, and $\beta = 106.0873°$

Atom	X/A	Y/B	Z/C
S	0.4206	-0.2093	0.3589
C	0.3072	-0.3842	0.3885
C	0.1874	-0.3354	0.3305
H	0.1569	-0.1791	0.2580
C	0.1089	-0.4909	0.3682
H	0.0154	-0.4538	0.3256
C	0.1493	0.3072	0.4618
H	0.0867	0.1889	0.4917
C	0.2684	0.2570	-0.4819
H	0.2990	0.1008	-0.4096
C	0.3494	0.4111	0.4822
C	0.4748	0.4051	-0.4727

Table S13: Theoretical cell coordinates for BTBT corresponding to 200 K. The crystal structure has symmetry $P2_1/c$, with corresponding cell parameters of: $a = 11.9092$ Å, $b = 5.8904$ Å, $c = 7.9548$ Å, $\alpha = \gamma = 90°$, and $\beta = 106.0937°$

Atom	X/A	Y/B	Z/C
S	0.4208	-0.2108	0.3588
C	0.3077	-0.3848	0.3886
C	0.1883	-0.3362	0.3308
H	0.1579	-0.1806	0.2583
C	0.1100	-0.4909	0.3686
H	0.0167	-0.4540	0.3260
C	0.1502	0.3083	0.4622
H	0.0878	0.1905	0.4921
C	0.2690	0.2584	-0.4815
H	0.2996	0.1030	-0.4091
C	0.3498	0.4117	0.4825
C	0.4748	0.4057	-0.4726

Table S14: Theoretical cell coordinates for BTBT corresponding to 300 K. The crystal structure has symmetry $P2_1/c$, with corresponding cell parameters of: $a = 11.9357$ Å, $b = 5.9084$ Å, $c = 8.0010$ Å, $\alpha = \gamma = 90°$, and $\beta = 105.8121°$

Atom	X/A	Y/B	Z/C
S	0.4217	-0.2123	0.3585
C	0.3086	-0.3851	0.3885
C	0.1896	-0.3367	0.3306
H	0.1595	-0.1820	0.2580
C	0.1112	-0.4906	0.3683
H	0.0183	-0.4540	0.3254
C	0.1509	0.3098	0.4619
H	0.0885	0.1927	0.4917
C	0.2692	0.2601	-0.4815
H	0.2994	0.1056	-0.4089
C	0.3501	0.4125	0.4825
C	0.4747	0.4064	-0.4725

2.2 Elastic Tensors

0 K (no ZPE)

$$
c_{ij} = \begin{bmatrix}
33.487 & 4.254 & 6.004 & 0.000 & 5.948 & 0.000 \\
4.254 & 15.515 & 10.892 & 0.000 & -1.442 & 0.000 \\
6.004 & 10.892 & 12.541 & 0.000 & -1.161 & 0.000 \\
0.000 & 0.000 & 0.000 & 7.520 & 0.000 & 0.205 \\
5.948 & -1.442 & -1.161 & 0.000 & 2.412 & 0.000 \\
0.000 & 0.000 & 0.000 & 0.205 & 0.000 & 6.491
\end{bmatrix} \text{GPa} \tag{7}
$$

0 K

$$
c_{ij} = \begin{bmatrix}
28.486 & 3.541 & 5.125 & 0.000 & 5.026 & 0.000 \\
3.541 & 13.585 & 9.589 & 0.000 & -1.346 & 0.000 \\
5.125 & 9.589 & 10.947 & 0.000 & -1.090 & 0.000 \\
0.000 & 0.000 & 0.000 & 6.825 & 0.000 & 0.044 \\
5.026 & -1.346 & -1.090 & 0.000 & 2.005 & 0.000 \\
0.000 & 0.000 & 0.000 & 0.044 & 0.000 & 5.539
\end{bmatrix} \text{GPa} \tag{8}
$$

100 K

$$
c_{ij} = \begin{bmatrix}
26.764 & 3.308 & 4.757 & 0.000 & 4.801 & 0.000 \\
3.308 & 12.940 & 9.186 & 0.000 & -1.270 & 0.000 \\
4.757 & 9.186 & 10.442 & 0.000 & -1.063 & 0.000 \\
0.000 & 0.000 & 0.000 & 6.606 & 0.000 & 0.013 \\
4.801 & -1.270 & -1.063 & 0.000 & 1.914 & 0.000 \\
0.000 & 0.000 & 0.000 & 0.013 & 0.000 & 5.215
\end{bmatrix} \text{GPa} \tag{9}
$$

200 K

$$
c_{ij} =
\begin{bmatrix}
23.947 & 2.923 & 4.185 & 0.000 & 4.275 & 0.000 \\
2.923 & 11.733 & 8.390 & 0.000 & -1.273 & 0.000 \\
4.815 & 8.390 & 9.573 & 0.000 & -1.002 & 0.000 \\
0.000 & 0.000 & 0.000 & 6.223 & 0.000 & -0.086 \\
4.275 & -1.273 & -1.002 & 0.000 & 1.665 & 0.000 \\
0.000 & 0.000 & 0.000 & -0.086 & 0.000 & 4.616
\end{bmatrix}
\text{GPa}
\tag{10}
$$

300 K

$$
c_{ij} =
\begin{bmatrix}
20.241 & 2.470 & 3.615 & 0.000 & 3.588 & 0.000 \\
2.470 & 10.492 & 7.613 & 0.000 & -1.239 & 0.000 \\
3.615 & 7.613 & 8.644 & 0.000 & -0.945 & 0.000 \\
0.000 & 0.000 & 0.000 & 5.762 & 0.000 & -0.197 \\
3.588 & -1.239 & -0.945 & 0.000 & 1.457 & 0.000 \\
0.000 & 0.000 & 0.000 & -0.197 & 0.000 & 4.010
\end{bmatrix}
\text{GPa}
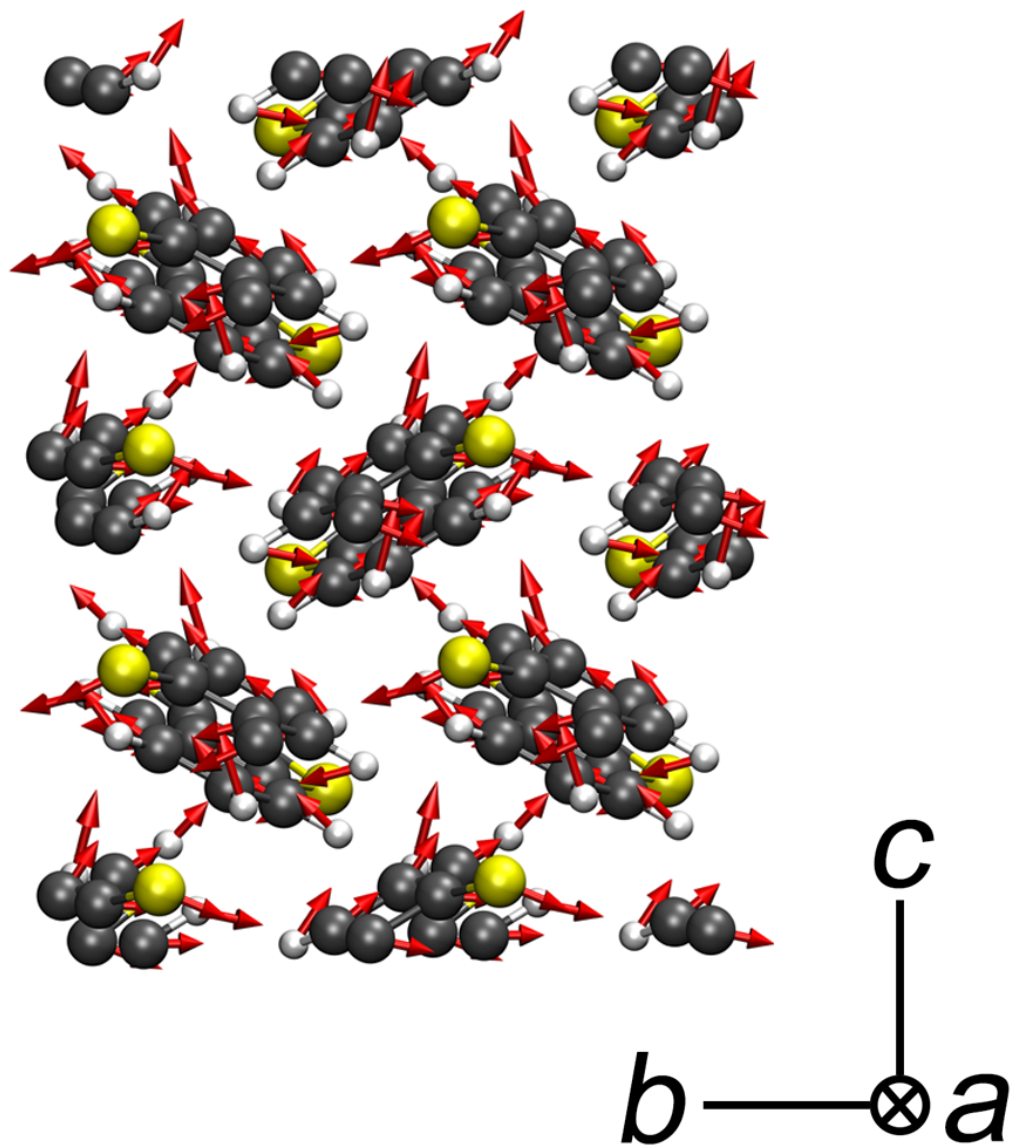\tag{11}
$$

2.3   Vibrational Mode Types

Figure S4: Calculated molecular motions of BTBT low-frequency vibrational mode with vibrational transition frequency of $58.98\,\mathrm{cm}^{-1}$, with atomic displacements illustrated with red arrows.
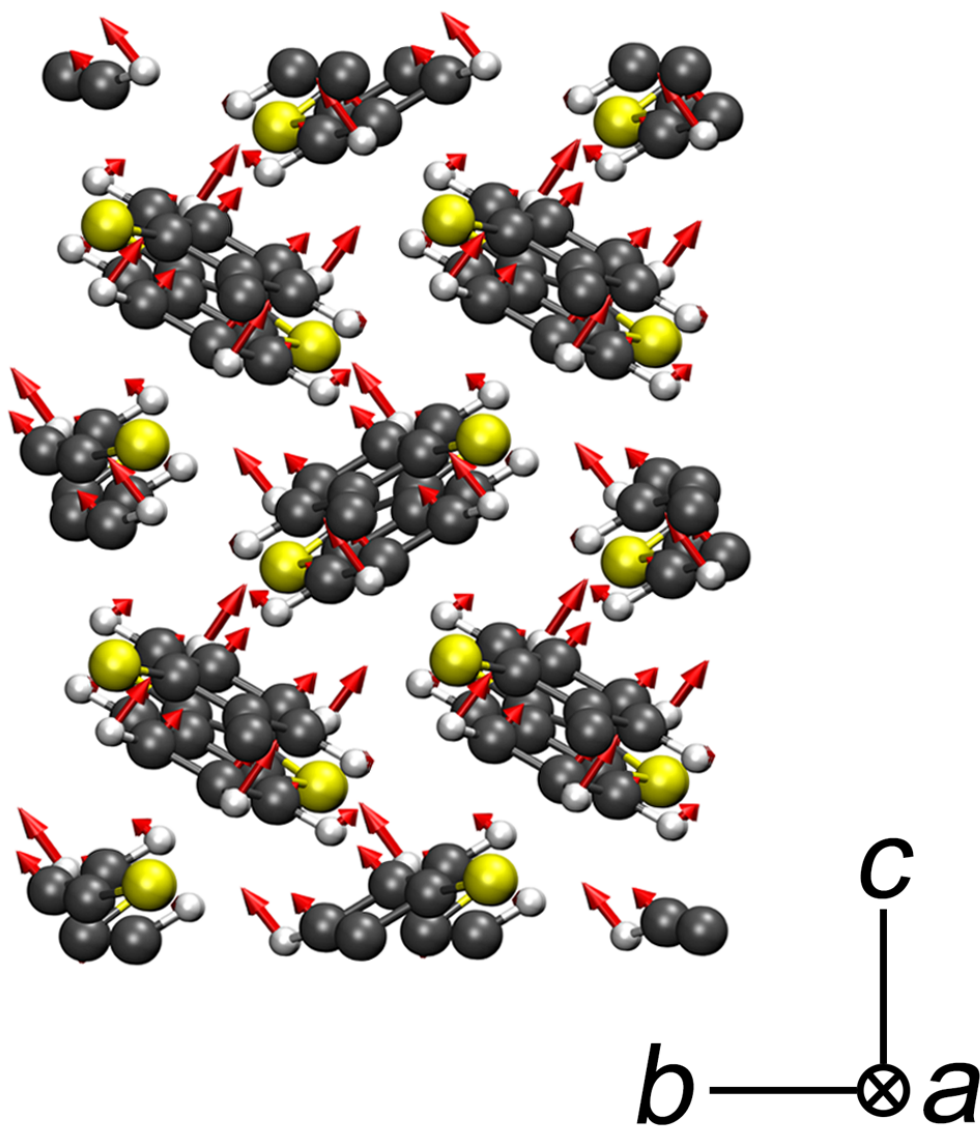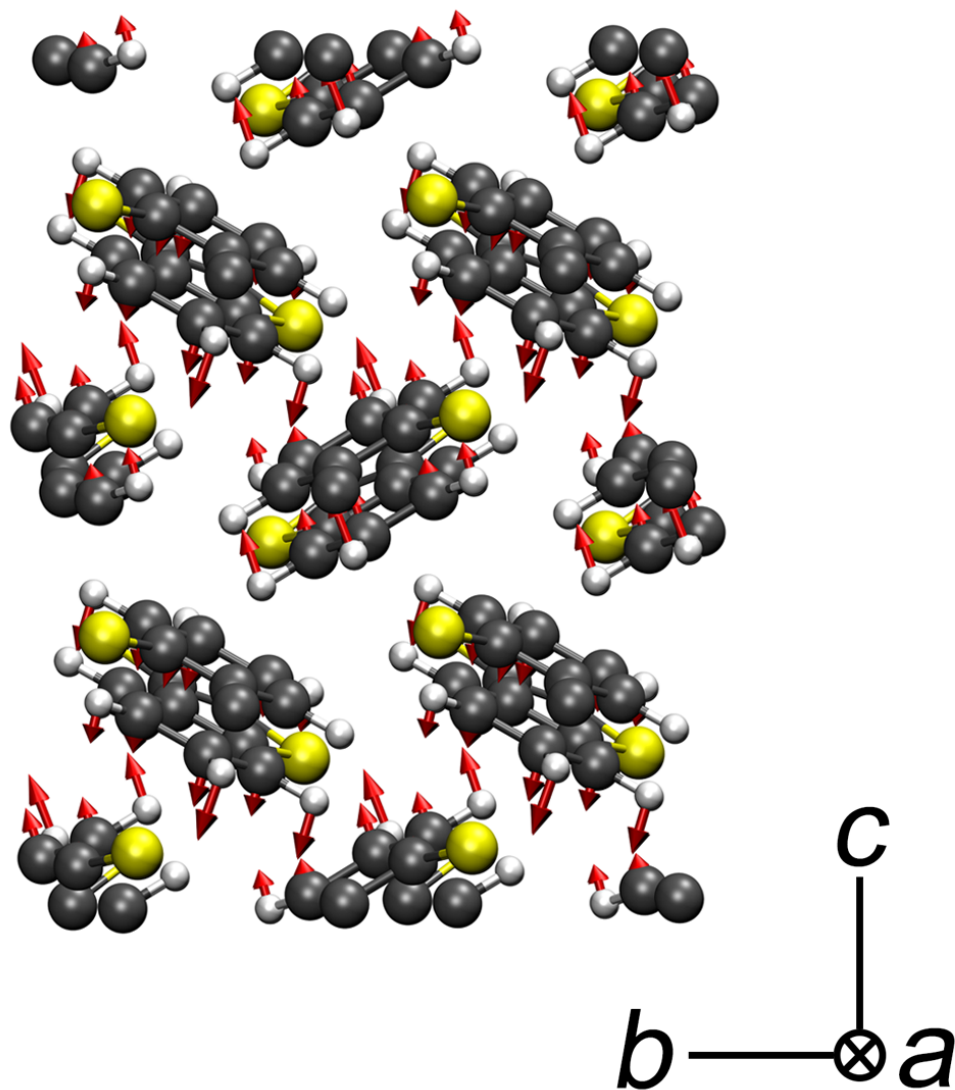
Figure S5: Calculated molecular motions of BTBT low-frequency vibrational mode with vibrational transition frequency of $65.85\,\mathrm{cm}^{-1}$, with atomic displacements illustrated with red arrows.

Figure S6: Calculated molecular motions of BTBT low-frequency vibrational mode with vibrational transition frequency of $93.64\,\mathrm{cm}^{-1}$, with atomic displacements illustrated with red arrows.
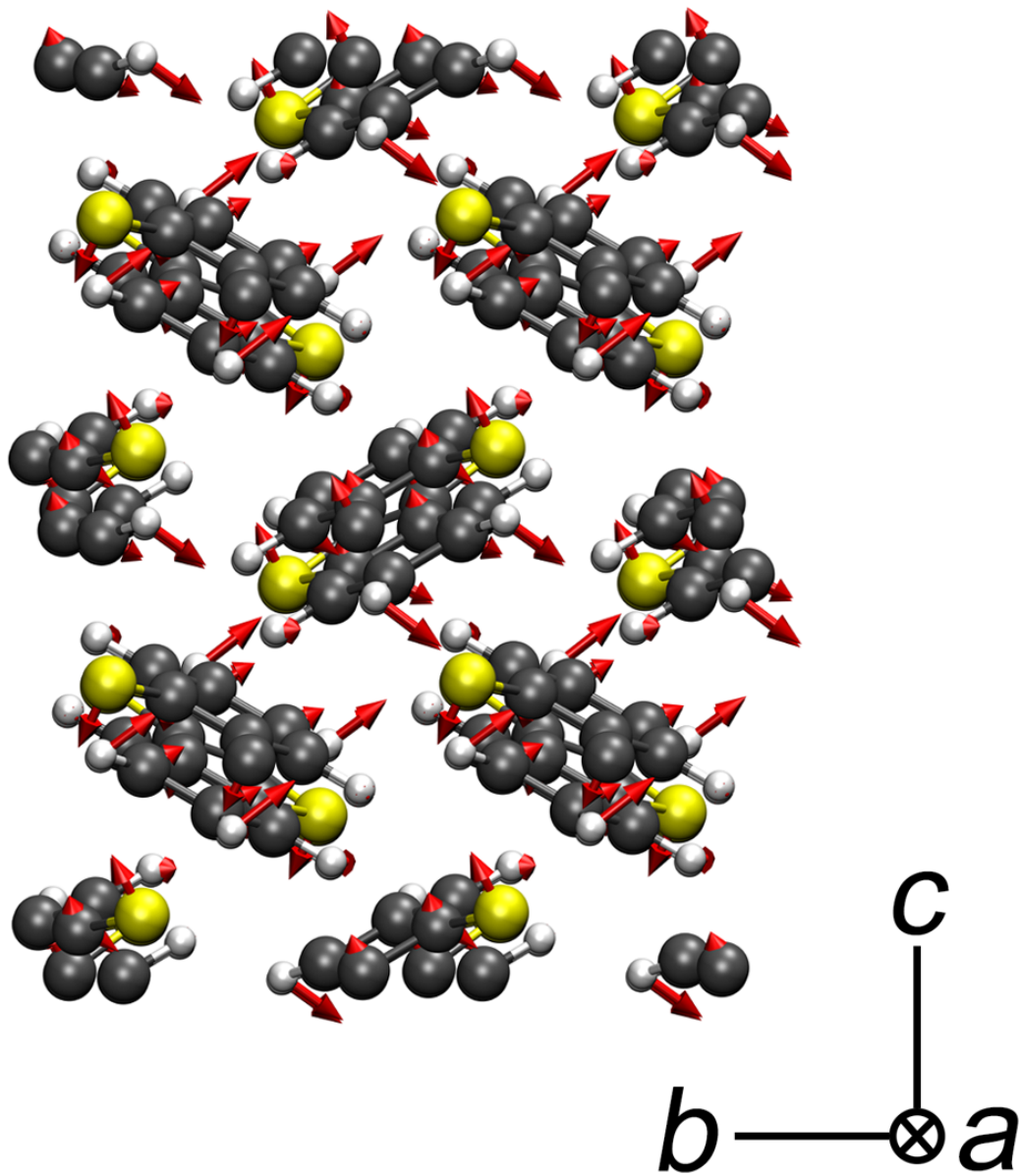
Figure S7: Calculated molecular motions of BTBT low-frequency vibrational mode with vibrational transition frequency of $109.91\,\mathrm{cm}^{-1}$, with atomic displacements illustrated with red arrows.