Supplementary Information

Machine Learning-Driven Optimization of Output Force in Photo-Actuated Organic Crystals

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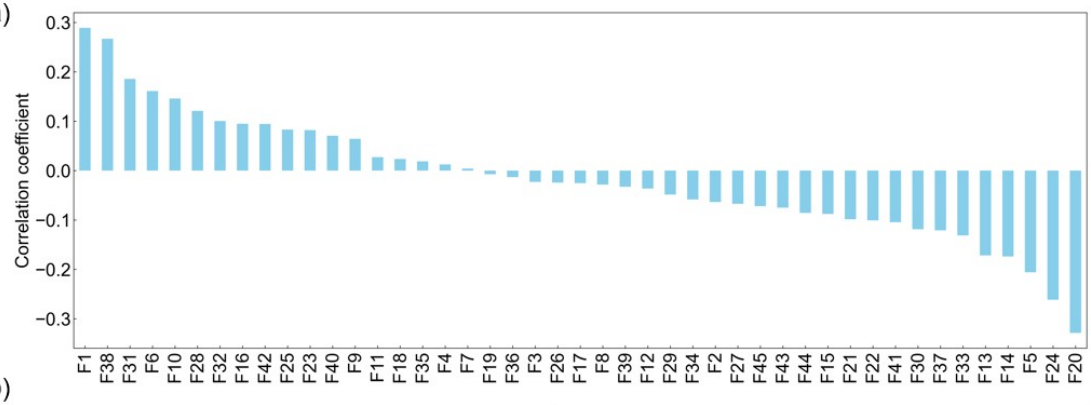
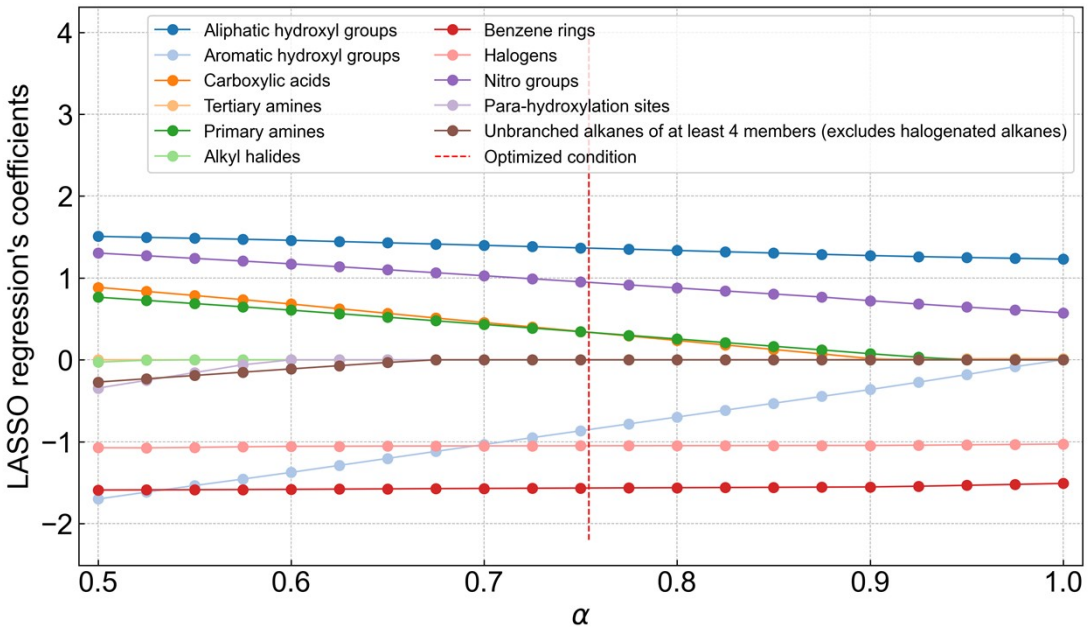
(a)



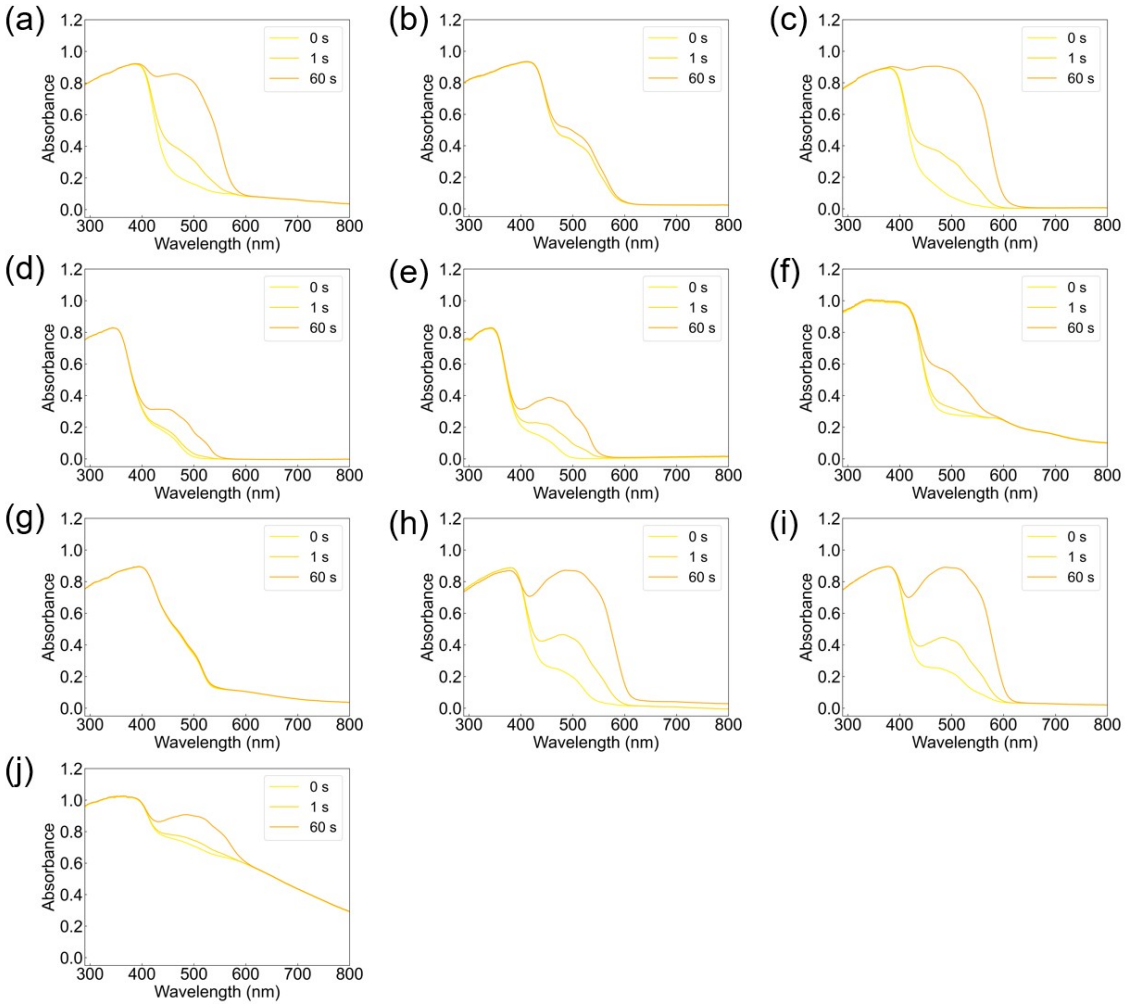
(b)

Name	Functional Group	Name	Functional Group
F1	Aliphatic hydroxyl groups	F24	Halogens
F2	Aromatic carboxylic acids	F25	Imidazole rings
F3	Aromatic nitrogens	F26	Imide groups
F4	Aromatic amines	F27	Ketones
F5	Aromatic hydroxyl groups	F28	Cyclic esters (lactones)
F6	Carboxylic acids	F29	Methoxy groups -OCH3
F7	Carbonyl O	F30	Nitriles
F8	Imines	F31	Nitro groups
F9	Tertiary amines	F32	Nitro benzene ring substituents
F10	Primary amines	F33	Para-hydroxylation sites
F11	XCCNR groups	F34	Piperdine rings
F12	Aldehydes	F35	Piperzine rings
F13	Alkyl halides	F36	Primary amides
F14	Allylic oxidation sites excluding steroid dienone	F37	Pyridine rings
F15	Amides	F38	Quarternary nitrogens
F16	Amidine groups	F39	Thioether
F17	Anilines	F40	Sulfonamides
F18	Aryl methyl sites for hydroxylation	F41	Terminal acetylenes
F19	Azo groups	F42	Thiazole rings
F20	Benzene rings	F43	Thiophene rings
F21	Bicyclic	F44	Unbranched alkanes of at least 4 members (excludes halogenated alkanes)
F22	Esters	F45	Urea groups
F23	Guanidine groups		

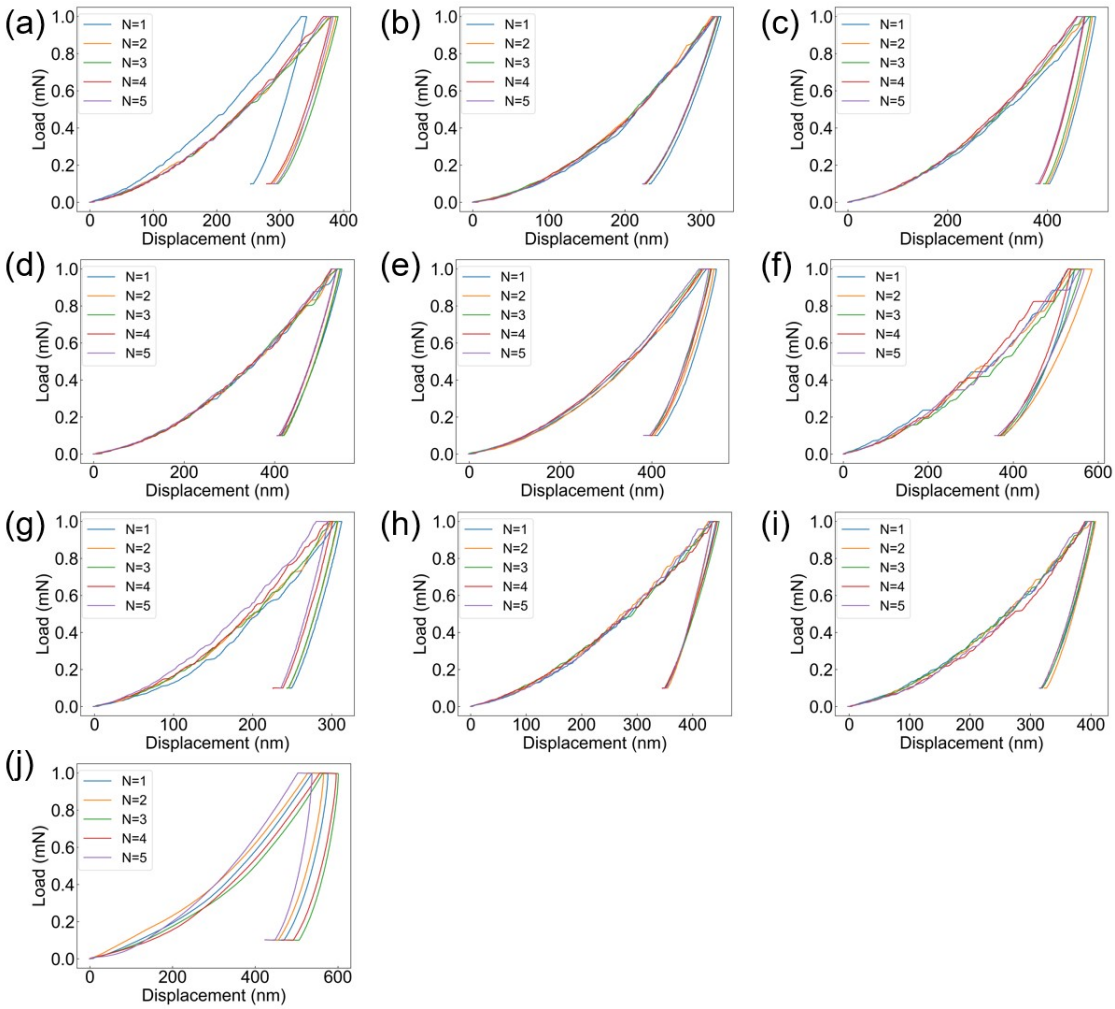
Supplementary Figure 1. The correlation coefficient of variables in the dataset used for LASSO regression. (a) Correlation coefficients between Young's modulus and explanatory variables. (b) List of explanatory variables of the functional groups.



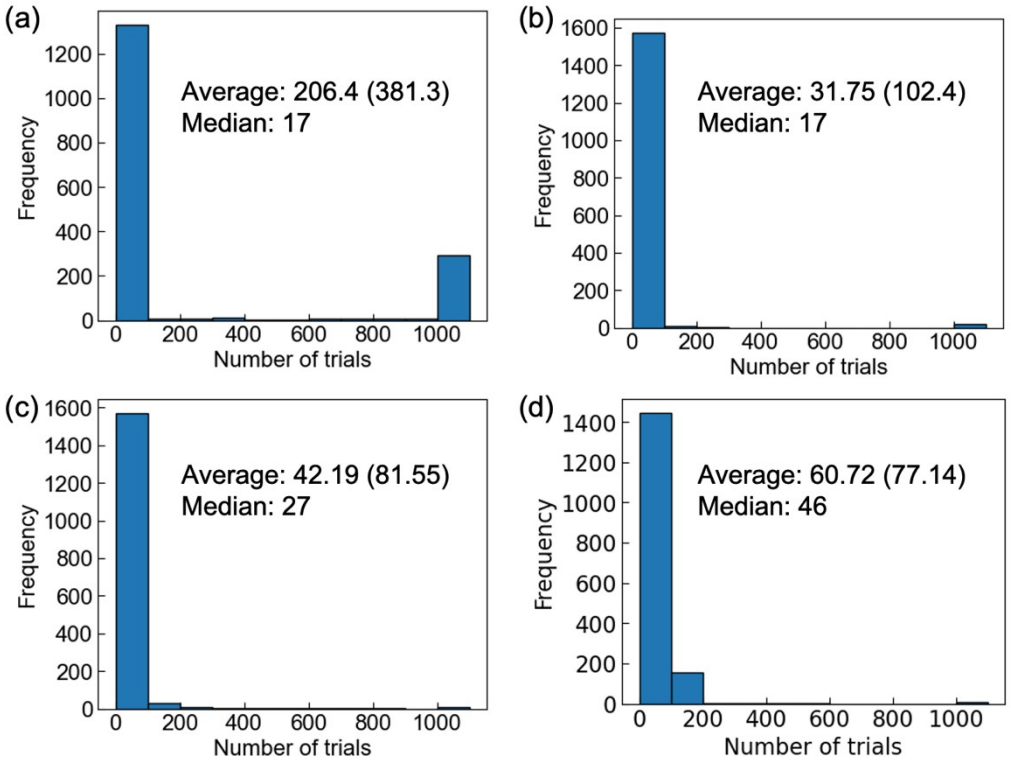
Supplementary Figure 2. The LASSO regression coefficients in different hyperparameters (α).



Suppelementary Figure 3. UV-Vis diffuse reflections spectra of salicylideneamine derivatives in the crystalline powder. (a-j) Results of compound **1**-**10** in the order.



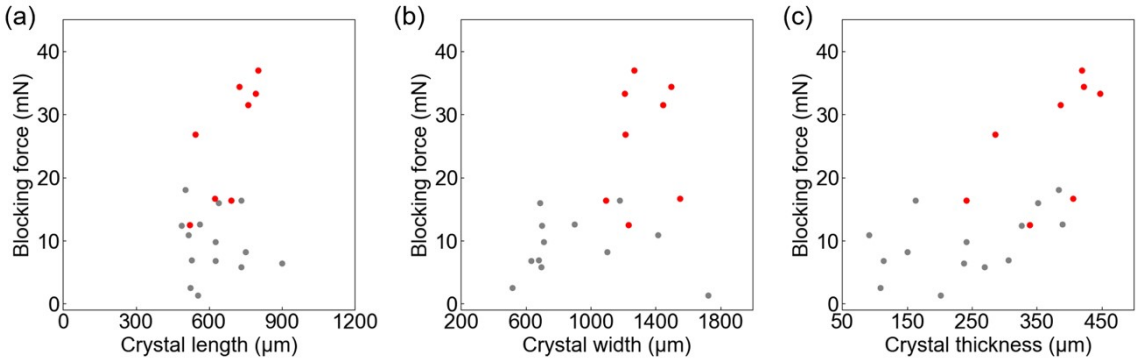
Suppelementary Figure 4. Load-displacement curves measured by nanoindentation test. (a-j) Results of compound **1**-**10** in the order.



Suppelementary Figure 5. Simulation results for optimizing the hyperparameter κ of the acquisition function for Bayesian optimization. Histograms represent the number of trials required to find the optimal conditions at (a) κ = 1, (b) κ = 5, (c) κ = 10, and (d) κ = 20.

[Description of the simulation]

The true function was assumed to have a peak in each dimension, and $f = 15exp\sum\left(-\left(x_i - \bar{x}_i\right)^2/2{\sigma_i}^2\right)$ was used as the assumed true function, where subscript i indicates a parameter (E_{bending}, L, W, T, I). The deviation σ_i was fixed to be a specific value. We simulated $2^5 = 32$ patterns, consisting of two possibilities for each axis peak position $\bar{x}_i$: either within the search range or outside it. For each pattern, we performed 50 runs, resulting in a total of 1,600 runs of Bayesian optimization with a specific κ. Five random data was used as initial data for each run, and the number of trials until reaching the terminate condition was counted. Each run was terminated when the suggested conditions produced values exceeding 90% of the global maximum or the number of trials reached at 1000. The figure above shows the histogram of the number of trials required to reach the terminate condition using Bayesian optimization. We determined that κ = 5 provided the optimal balance between exploration and exploitation in the parameter space.



Supplementary Figure 6. The relationship between crystal size and maximum blocking force. (a) Crystal length, (b) width, and (c) thickness with blocking force at higher intensity ragion (I > 1900 mW/cm^2). Compound **3** was shown in red, and other compounds were shown in gray.

Supplementary Table 1. Lattice parameters of prepared crystals.

	1	**2**	**3**	(*S*)-**4**	(*S*)-**5**
Crystal system	Monoclinic	Monoclinic	Triclinic	Triclinic	Orthorhombic
Space group	$P2_1/c$	$P2_1/n$	*P*-1	*P*1	$P2_12_12_1$
a (Å)	17.612(2)	6.2038(12)	6.048(2)	6.2330(12)	6.375(2)
b (Å)	6.4089(8)	19.580(4)	10.323(4)	9.9318(17)	9.847(4)
c (Å)	18.883(2)	16.221(4)	16.808(6)	19.711(4)	39.354(12)
α (°)	90	90	101.144(8)	85.466(6)	90
β (°)	107.368(8)	91.568(6)	92.208(8)	86.216(6)	90
γ (°)	90	90	101.564(8)	88.759(6)	90
V (Å^3)	2034.2(4)	1969.7(7)	1005.4(6)	1213.6(4)	2470.4(14)
Z	4	4	2	2	4
d (g/cm^3)	1.154	1.192	1.171	1.061	1.042
$R_1[I > 2\sigma(I)]$	0.0741	0.0615	0.0554	0.593	0.0607
wR_2	0.2017	0.1598	0.1470	0.1719	0.1724
GOF	1.085	1.017	1.030	1.127	0.929
CCDC	2391778	2391779	2391780	1879559	2391781

	6	**7**	**8**	**9**	**10**
Crystal system	Triclinic	Orthorhombic	Monoclinic	Monoclinic	Monoclinic
Space group	*P*-1	$Pna2_1$	$P2_1/c$	$P2_1/c$	$P2_1/c$
a (Å)	7.6777(5)	12.399(3)	17.8843(15)	17.953(8)	18.2167(9)
b (Å)	8.9801(5)	9.031(3)	10.4746(9)	10.621(5)	10.7674(6)
c (Å)	16.5297(10)	16.721(4)	10.3528(9)	10.457(4)	10.3547(6)
α (°)	93.611(7)	90	90	90	90
β (°)	91.357(6)	90	92.529(6)	93.358(11)	93.088(7)
γ (°)	98.030(7)	90	90	90	90
V (Å^3)	1125.64(12)	1872.4(9)	1937.5(3)	1990.5(15)	2028.09(19)
Z	2	4	4	4	4
d (g/cm^3)	1.293	1.161	1.179	1.296	1.426
$R_1[I > 2\sigma(I)]$	0.0653	0.0378	0.0758	0.0548	0.0752
wR_2	0.1364	0.0991	0.1643	0.1285	0.1789
GOF	1.186	1.047	1.194	1.013	1.148
CCDC	2391782	2391783	2391784	2391785	2391786

Supplemtanry Table 2. Suggested and experimental conditions, and measured blocking force in the process of Bayesian optimization.

ID	$E_{bending}$ (GPa)	*Compound*	L (μm)	W (μm)	T (μm)	I (mW/cm^2)	F_{above} (mN)	F_{below} (mN)
1	1.25	(*S*)-**5**	626	708	241	208	3.42	0.09
2	1.25	(*S*)-**5**	626	708	241	520	5.85	0.03
3	1.25	(*S*)-**5**	626	708	241	1040	8.34	0.02
4	1.25	(*S*)-**5**	626	708	241	1560	9.22	0.10
5	1.25	(*S*)-**5**	626	708	241	2080	9.81	0.19
6	0.664	**8**	625	631	113	208	0.73	0.19
7	0.664	**8**	625	631	113	520	1.89	0.33
8	0.664	**8**	625	631	113	1040	3.67	0.52
9	0.664	**8**	625	631	113	1560	5.07	1.41
10	0.664	**8**	625	631	113	2080	6.82	1.74
Suggest 1	2.29	**9**	626	694	250	2080	6.37	
11	2.29	**9**	731	693	269	2080	5.86	2.38
12	2.29	**9**	731	693	269	1560	5.90	1.61
13	2.29	**9**	731	693	269	1040	3.99	0.81
14	2.29	**9**	731	693	269	520	1.20	Broken
15	2.29	**9**	731	693	269	208	0.59	Broken
Suggest 2	0.602	**1**	509	741	237	2080	6.70	
16	0.602	**1**	523	516	109	2080	2.55	1.83
17	0.602	**1**	523	516	109	1560	2.34	1.28
18	0.602	**1**	523	516	109	1040	1.72	1.18
19	0.602	**1**	523	516	109	520	0.63	0.92
20	0.602	**1**	523	516	109	208	0.19	0.72
Suggest 3	0.602	**1**	604	887	211	2080	6.70	
21	0.602	**1**	639	687	352	2080	15.95	3.05
22	0.602	**1**	639	687	352	1560	11.35	1.47
23	0.602	**1**	639	687	352	1040	6.85	0.92
24	0.602	**1**	639	687	352	520	3.34	0.84
25	0.602	**1**	639	687	352	208	1.95	0.55
Suggest 4	0.602	**1**	583	795	496	2080	10.50	
26	0.602	**1**	486	697	326	2080	12.42	0.83

27	0.602	**1**	486	697	326	1560	11.01	0.22
28	0.602	**1**	486	697	326	1040	10.67	0.12
29	0.602	**1**	486	697	326	520	2.43	0.32
30	0.602	**1**	486	697	326	208	0.95	0.00
Suggest 5	0.602	**1**	590	780	402	2080	10.81	
31	1.25	(*S*)-**5**	562	900	389	2080	12.57	2.58
32	1.25	(*S*)-**5**	562	900	389	1560	11.06	1.66
33	1.25	(*S*)-**5**	562	900	389	1040	8.77	1.42
34	1.25	(*S*)-**5**	562	900	389	520	4.85	0.82
35	1.25	(*S*)-**5**	562	900	389	208	2.96	0.51
Suggest 6	0.677	**3**	571	713	500	2080	11.77	
36	0.677	**3**	543	1211	286	2080	26.84	7.38
37	0.677	**3**	543	1211	286	1560	20.55	9.01
38	0.677	**3**	543	1211	286	1040	13.39	4.14
39	0.677	**3**	543	1211	286	520	5.96	3.81
40	0.677	**3**	543	1211	286	208	2.80	1.97
Suggest 7	0.602	**1**	654	1310	296	2080	18.56	
41	1.83	(*S*)-**4**	555	1722	202	2080	1.36	0.01
42	1.83	(*S*)-**4**	555	1722	202	1560	1.10	0.06
43	1.83	(*S*)-**4**	555	1722	202	1040	1.36	0.10
44	1.83	(*S*)-**4**	555	1722	202	520	1.89	0.20
45	1.83	(*S*)-**4**	555	1722	202	208	1.71	0.02
Suggest 8	0.602	**1**	579	1536	372	2080	19.77	
46	1.25	(*S*)-**5**	502	2393	383	2080	18.05	2.27
47	1.25	(*S*)-**5**	502	2393	383	1560	15.72	2.09
48	1.25	(*S*)-**5**	502	2393	383	1040	12.59	0.67
49	1.25	(*S*)-**5**	502	2393	383	520	7.18	0.66
50	1.25	(*S*)-**5**	502	2393	383	208	3.16	1.35
Suggest 9	0.602	**1**	584	1917	340	2080	23.45	
51	1.83	(*S*)-**4**	900	2672	237	2080	6.43	0.05
52	1.83	(*S*)-**4**	900	2672	237	1560	5.16	1.30
53	1.83	(*S*)-**4**	900	2672	237	1040	7.16	0.76
54	1.83	(*S*)-**4**	900	2672	237	520	1.15	0.11
55	1.83	(*S*)-**4**	900	2672	237	208	0.10	0.00
Suggest 10	0.602	**1**	601	2000	308	2080	24.25	

56	0.602	**1**	527	680	306	2080	6.94	2.12
57	0.602	**1**	527	680	306	1560	11.60	2.11
58	0.602	**1**	527	680	306	1040	2.44	0.34
59	0.602	**1**	527	680	306	520	0.67	0.24
60	0.602	**1**	527	680	306	208	0.19	0.23
Suggest 11	0.677	**3**	538	1700	329	2080	23.03	
61	0.677	**3**	521	1233	339	2080	12.45	6.12
62	0.677	**3**	521	1233	339	1560	15.00	3.76
63	0.677	**3**	521	1233	339	1040	8.68	1.79
64	0.677	**3**	521	1233	339	520	3.78	0.54
65	0.677	**3**	521	1233	339	208	0.98	0.54
Suggest 12	0.677	**3**	584	1567	257	2072	22.11	
66	0.677	**3**	725	1495	422	2072	34.41	9.33
67	0.677	**3**	725	1495	422	1560	32.74	9.50
68	0.677	**3**	725	1495	422	1040	22.74	5.83
69	0.677	**3**	725	1495	422	520	7.55	2.12
70	0.677	**3**	725	1495	422	208	2.20	0.63
Suggest 13	0.677	**3**	693	1686	329	1957	26.04	
71	0.677	**3**	624	1551	406	1957	16.66	8.49
72	0.677	**3**	624	1551	406	1560	17.21	7.82
73	0.677	**3**	624	1551	406	1040	11.20	5.19
74	0.677	**3**	624	1551	406	520	5.06	2.68
75	0.677	**3**	624	1551	406	208	1.16	0.87
Suggest 14	0.677	**3**	766	1513	340	1912	26.41	
76	0.677	**3**	761	1444	386	1912	31.52	8.25
77	0.677	**3**	761	1444	386	1560	22.73	8.88
78	0.677	**3**	761	1444	386	1040	17.08	6.71
79	0.677	**3**	761	1444	386	520	8.75	4.70
80	0.677	**3**	761	1444	386	208	3.16	2.21
Suggest 15	0.677	**3**	775	1320	500	1800	26.24	
81	0.677	**3**	801	1266	419	1800	14.88	8.63
82	0.677	**3**	801	1266	419	2080	36.97	19.45
83	0.677	**3**	801	1266	419	1040	12.58	5.99
84	0.677	**3**	801	1266	419	520	6.47	4.02
85	0.677	**3**	801	1266	419	208	1.42	2.06

Suggest 16	1.25	(*S*)-**5**	777	1565	419	2080	20.39	
86	1.25	(*S*)-**5**	750	1101	150	2080	8.23	0.04
87	1.25	(*S*)-**5**	750	1101	150	1560	10.72	0.11
88	1.25	(*S*)-**5**	750	1101	150	1040	9.64	0.03
89	1.25	(*S*)-**5**	750	1101	150	520	8.71	0.07
90	1.25	(*S*)-**5**	750	1101	150	208	6.24	0.09
Suggest 17	1.25	(*S*)-**5**	774	1514	418	2080	20.19	
91	1.25	(*S*)-**5**	514	1413	91	2080	10.93	1.18
92	1.25	(*S*)-**5**	514	1413	91	1560	9.84	0.02
93	1.25	(*S*)-**5**	514	1413	91	1040	10.94	0.00
94	1.25	(*S*)-**5**	514	1413	91	520	8.93	0.03
95	1.25	(*S*)-**5**	514	1413	91	208	4.92	0.00
Suggest 18	1.25	(*S*)-**5**	768	1452	418	2080	19.90	
96	1.25	(*S*)-**5**	733	1179	163	2080	16.37	0.00
97	1.25	(*S*)-**5**	733	1179	163	1560	6.94	0.00
98	1.25	(*S*)-**5**	733	1179	163	1040	8.32	0.01
99	1.25	(*S*)-**5**	733	1179	163	520	7.80	0.00
100	1.25	(*S*)-**5**	733	1179	163	208	5.62	0.00
Suggest 19	0.677	**3**	745	1237	404	2080	35.61	
101	0.677	**3**	791	1208	447	2080	33.32	9.42
102	0.677	**3**	791	1208	447	1560	23.87	6.26
103	0.677	**3**	791	1208	447	1040	15.59	3.32
104	0.677	**3**	791	1208	447	520	7.78	1.36
105	0.677	**3**	791	1208	447	208	2.35	0.35
Suggest 20	0.677	**3**	739	1211	394	2080	34.21	
106	0.677	**3**	690	1091	241	2080	16.33	4.79
107	0.677	**3**	690	1091	241	1560	8.29	4.95
108	0.677	**3**	690	1091	241	1040	5.48	3.38
109	0.677	**3**	690	1091	241	520	2.91	3.04
110	0.677	**3**	690	1091	241	208	0.69	1.22

($E_{bending}$: Young's modulus measured by bending test, L: crystal length, W: crystal width, T: crystal thickness, I: light intensity, F_{above}: maximum blocking force upon light from upper the crystal, F_{below}: maximum blocking force upon light irradiation from under the crystal.)