

Magnetic properties of CrX_3 ($\text{X}=\text{Cl},\text{Br},\text{I}$) monolayers in excited states

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Electronic Supplementary Information:

Table S1: Particle-hole state, excitation energy (E_{ex} , magnetic anisotropy energy (MAE), Dipole moment, Transition dipole moment (TDM), Average spin-charge on Cr sites for CrI_3 for singly excited states spin conserving particle-hole state.

Transition	E (eV)	MAE (K)	Dipole (D)	TDM	Spin charge (e)		
					Integrated	Mulliken	Lowdin
H \rightarrow L	2.24	60.60	11.64	0.1532	2.9809	3.5103	3.1495
H \rightarrow L+1	1.34	63.63	0.91	0.2795	2.9790	3.5093	3.1482
H \rightarrow L+2	1.50	72.81	3.37	0.0385	2.9791	3.5098	3.1491
H \rightarrow L+3	1.45	66.34	2.13	0.2517	2.9790	3.5098	3.1497
H \rightarrow L+4	1.50	64.41	2.81	0.2432	2.9789	3.5091	3.1499
H \rightarrow L+5	1.61	82.87	4.63	0.1699	2.9763	3.5058	3.1469
H \rightarrow L+6	1.46	61.50	3.24	0.1171	2.9755	3.5055	3.1459
H \rightarrow L+7	2.24	139.76	4.36	0.7114	2.9848	3.5169	3.1564
H \rightarrow L+8	1.63	83.53	3.23	0.1015	2.9747	3.5053	3.1459
H \rightarrow L+9	2.65	36.71	13.59	0.0100	2.9721	3.5021	3.1435
H \rightarrow L+10	2.36	78.04	11.57	0.3713	2.9726	3.5028	3.1444
H \rightarrow L+11	2.26	62.50	9.80	0.3562	2.9715	3.5016	3.1437
H-1 \rightarrow L	1.67	73.41	7.79	0.0941	2.9822	3.5105	3.1509
H-1 \rightarrow L+1	1.66	69.96	2.76	0.2357	2.9842	3.5152	3.1541
H-1 \rightarrow L+2	1.71	80.73	4.88	0.0186	2.9856	3.5154	3.1559
H-1 \rightarrow L+3	1.43	64.31	1.80	0.1080	2.9790	3.5082	3.149
H-1 \rightarrow L+4	1.46	90.37	1.86	0.0719	2.9792	3.5079	3.1498
H-1 \rightarrow L+5	1.81	75.29	8.06	0.2542	2.9812	3.5104	3.1523
H-1 \rightarrow L+6	1.71	117.04	3.57	0.1412	2.9875	3.5181	3.1592
H-1 \rightarrow L+7	1.71	85.55	5.36	0.3215	2.9778	3.5086	3.1493
H-1 \rightarrow L+8	1.92	67.66	2.28	0.1300	2.9774	3.5078	3.149
H-1 \rightarrow L+9	1.65	74.60	3.81	0.0590	2.9789	3.5098	3.152

H-1 \rightarrow L+10	3.97	76.16	16.77	0.1886	2.9771	3.5068	3.1487
H-1 \rightarrow L+11	2.33	81.91	8.72	1.1130	2.9749	3.5034	3.1468
H-2 \rightarrow L	2.63	123.60	16.44	0.0384	3.0043	3.5333	3.1753
H-2 \rightarrow L+1	4.64	97.26	11.74	0.1138	2.9883	3.5105	3.1542
H-2 \rightarrow L+2	1.69	116.31	6.33	0.0781	2.9940	3.5266	3.1678
H-2 \rightarrow L+3	1.51	131.10	2.28	0.2068	3.0007	3.5328	3.1754
H-2 \rightarrow L+4	1.60	111.10	3.45	0.1951	2.9995	3.5312	3.1746
H-2 \rightarrow L+5	2.30	121.63	14.47	0.0606	2.9962	3.5278	3.1706
H-2 \rightarrow L+6	1.74	155.39	9.18	0.1372	2.9895	3.5252	3.1661
H-2 \rightarrow L+7	3.05	102.10	18.49	0.0170	2.9981	3.5291	3.1706
H-2 \rightarrow L+8	1.97	115.36	3.73	0.2426	2.9965	3.5283	3.1708
H-2 \rightarrow L+9	3.07	111.72	18.81	0.0818	2.9974	3.5289	3.1717
H-2 \rightarrow L+10	4.18	158.20	22.08	0.1326	2.9988	3.5298	3.1727
H-2 \rightarrow L+11	1.78	81.97	3.16	0.0692	2.9897	3.5285	3.1686
H-3 \rightarrow L	2.61	118.66	15.85	0.0995	3.0047	3.5342	3.1762
H-3 \rightarrow L+1	1.62	121.03	0.92	0.1116	3.0043	3.5367	3.1774
H-3 \rightarrow L+2	2.87	117.88	14.79	0.1175	3.0016	3.5318	3.1734
H-3 \rightarrow L+3	1.57	121.27	2.84	0.0768	2.9990	3.5323	3.1748
H-3 \rightarrow L+4	1.68	101.88	7.47	0.0245	2.9950	3.5296	3.1727
H-3 \rightarrow L+5	1.86	105.73	7.76	0.0959	2.9959	3.5269	3.1697
H-3 \rightarrow L+6	2.90	86.30	11.39	0.0304	2.9989	3.5232	3.1663
H-3 \rightarrow L+7	2.87	119.49	15.69	0.0435	2.9971	3.5286	3.1701
H-3 \rightarrow L+8	1.75	92.16	1.45	0.0831	2.9952	3.5288	3.1711
H-3 \rightarrow L+9	2.60	82.29	12.92	0.0877	3.0002	3.5293	3.1733
H-3 \rightarrow L+10	3.60	97.63	16.44	0.1745	3.0013	3.5322	3.1755
H-3 \rightarrow L+11	2.35	134.92	7.56	0.0183	3.0004	3.5284	3.1735

H-4 \rightarrow L	2.21	83.11	2.68	0.0786	3.0041	3.5338	3.1751
H-4 \rightarrow L+1	1.71	78.09	1.63	0.0455	2.9996	3.5317	3.1718
H-4 \rightarrow L+2	1.65	44.30	4.97	0.8167	2.9799	3.5132	3.1545
H-4 \rightarrow L+3	1.74	87.18	2.49	0.1803	3.0015	3.5341	3.1761
H-4 \rightarrow L+4	1.81	83.68	1.36	0.1493	3.0028	3.5343	3.1773
H-4 \rightarrow L+5	2.03	73.04	7.43	0.2264	2.9965	3.5287	3.1709
H-4 \rightarrow L+6	2.09	65.68	9.47	0.2015	2.9940	3.5246	3.1668
H-4 \rightarrow L+7	3.44	68.06	12.77	0.1115	2.9966	3.52	3.1637
H-4 \rightarrow L+8	1.93	72.20	1.81	0.1103	2.9959	3.5276	3.1697
H-4 \rightarrow L+9	2.11	67.54	7.36	0.1534	3.0021	3.5385	3.179
H-4 \rightarrow L+10	3.94	62.17	17.29	0.3176	2.9975	3.5282	3.1709
H-4 \rightarrow L+11	1.95	74.03	7.12	0.0847	2.9845	3.5175	3.1603
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H-5 \rightarrow L	2.25	114.96	13.66	0.1520	2.9839	3.513	3.1533
H-5 \rightarrow L+1	1.62	97.97	4.02	0.0703	2.9769	3.5073	3.1468
H-5 \rightarrow L+2	1.82	97.75	5.99	0.2253	2.9905	3.5215	3.1618
H-5 \rightarrow L+3	1.60	111.66	2.63	0.0951	2.9779	3.5086	3.1494
H-5 \rightarrow L+4	1.60	87.30	2.47	0.0718	2.9739	3.5042	3.1457
H-5 \rightarrow L+5	1.92	116.40	8.02	0.2118	2.9808	3.5102	3.1521
H-5 \rightarrow L+6	1.87	134.43	5.75	0.0230	2.9861	3.5166	3.1579
H-5 \rightarrow L+7	2.85	103.40	14.18	0.0830	2.9750	3.5056	3.146
H-5 \rightarrow L+8	1.75	86.03	2.29	0.0979	2.9714	3.5019	3.1433
H-5 \rightarrow L+9	2.06	76.32	7.24	0.0322	2.9826	3.5168	3.157
H-5 \rightarrow L+10	3.42	93.40	15.17	0.1257	2.9743	3.5042	3.1464
H-5 \rightarrow L+11	2.87	114.17	15.62	0.0015	2.9778	3.5082	3.151
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H-6 \rightarrow L	3.00	95.31	21.71	0.1357	3.0040	3.5344	3.1756
H-6 \rightarrow L+1	1.83	102.44	9.43	0.2920	2.9924	3.5242	3.1644

H-6 \rightarrow L+2	4.10	57.78	27.17	0.0385	2.9928	3.5218	3.1634
H-6 \rightarrow L+3	1.97	93.83	7.34	0.0421	2.9949	3.5268	3.1684
H-6 \rightarrow L+4	1.89	88.08	7.24	0.0508	2.9924	3.5243	3.1668
H-6 \rightarrow L+5	2.11	89.16	6.92	0.3598	2.9903	3.5201	3.1628
H-6 \rightarrow L+6	3.81	67.98	23.06	0.0627	2.9965	3.5236	3.1667
H-6 \rightarrow L+7	2.08	81.81	4.62	0.2785	2.9727	3.5088	3.1493
H-6 \rightarrow L+8	2.10	71.54	8.50	0.2354	2.9862	3.5184	3.1602
H-6 \rightarrow L+9	1.86	55.44	2.28	0.0762	2.9784	3.513	3.1547
H-6 \rightarrow L+10	4.28	104.22	22.57	0.0405	2.9905	3.5205	3.1637
H-6 \rightarrow L+11	3.44	82.59	24.39	0.1680	2.9901	3.5203	3.1641
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H-7 \rightarrow L	2.14	44.58	9.71	0.0595	2.9665	3.4936	3.1344
H-7 \rightarrow L+1	1.70	51.35	2.23	0.0655	2.9732	3.5021	3.1424
H-7 \rightarrow L+2	3.30	29.85	15.66	0.1686	2.9657	3.494	3.1348
H-7 \rightarrow L+3	1.63	43.42	2.85	0.0354	2.9654	3.4935	3.135
H-7 \rightarrow L+4	1.68	42.79	3.03	0.0286	2.9660	3.4931	3.1359
H-7 \rightarrow L+5	2.27	60.15	13.48	0.0087	2.9696	3.4974	3.1397
H-7 \rightarrow L+6	3.57	39.48	20.18	0.0677	2.9657	3.4921	3.1345
H-7 \rightarrow L+7	3.02	49.02	17.78	0.3264	2.9640	3.4928	3.1337
H-7 \rightarrow L+8	1.96	61.52	4.32	0.0711	2.9666	3.4951	3.1375
H-7 \rightarrow L+9	2.86	45.06	17.23	0.0732	2.9633	3.4917	3.1346
H-7 \rightarrow L+10	2.00	56.23	3.04	0.1686	2.9620	3.4896	3.1331
H-7 \rightarrow L+11	3.06	47.14	14.93	0.0354	2.9641	3.4924	3.136
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H-8 \rightarrow L	2.56	85.22	13.79	0.0286	2.9950	3.5238	3.1654
H-8 \rightarrow L+1	2.00	81.76	7.18	0.0087	2.9925	3.5225	3.1633
H-8 \rightarrow L+2	1.79	87.26	3.34	0.3709	2.9968	3.5275	3.1688
H-8 \rightarrow L+3	1.78	85.64	4.09	0.2463	2.9825	3.5124	3.1546

H-8 \rightarrow L+4	1.79	83.62	4.32	0.2320	2.9823	3.5119	3.1549
H-8 \rightarrow L+5	1.87	95.21	5.84	0.4239	2.9806	3.51	3.1528
H-8 \rightarrow L+6	2.03	133.11	3.67	0.2212	2.9885	3.5197	3.1616
H-8 \rightarrow L+7	2.00	79.14	5.39	0.0056	2.9662	3.4973	3.1382
H-8 \rightarrow L+8	2.06	75.86	4.38	0.1942	2.9861	3.5166	3.1592
H-8 \rightarrow L+9	1.91	59.75	5.83	0.0362	2.9702	3.5005	3.1434
H-8 \rightarrow L+10	3.14	93.98	8.05	0.1631	2.9863	3.5154	3.1591
H-8 \rightarrow L+11	3.12	78.98	18.58	0.0412	2.9818	3.5106	3.1551
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H-9 \rightarrow L	2.25	46.36	8.80	0.1170	2.9858	3.514	3.1556
H-9 \rightarrow L+1	1.84	79.11	4.84	0.2136	2.9909	3.5216	3.162
H-9 \rightarrow L+2	2.28	57.41	8.73	0.1702	2.9895	3.5197	3.1611
H-9 \rightarrow L+3	1.72	55.29	3.44	0.1223	2.9850	3.5154	3.1573
H-9 \rightarrow L+4	1.75	53.11	5.51	0.0883	2.9849	3.5154	3.1581
H-9 \rightarrow L+5	1.82	76.32	0.65	0.0648	2.9813	3.5123	3.1549
H-9 \rightarrow L+6	2.04	86.04	6.49	0.0131	2.9906	3.5225	3.1642
H-9 \rightarrow L+7	3.05	35.26	13.24	0.1123	2.9844	3.5146	3.1558
H-9 \rightarrow L+8	2.09	51.16	3.57	0.0915	2.9862	3.5171	3.1594
H-9 \rightarrow L+9	1.93	117.74	4.83	0.0833	2.9950	3.5315	3.1712
H-9 \rightarrow L+10	3.87	60.24	20.25	0.1027	2.9846	3.5143	3.1574
H-9 \rightarrow L+11	2.22	69.86	1.52	0.0973	2.9839	3.5112	3.1559
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H-10 \rightarrow L	2.99	69.47	17.98	0.3568	2.9799	3.5073	3.1487
H-10 \rightarrow L+1	2.02	50.60	6.74	0.0849	2.9839	3.5135	3.1539
H-10 \rightarrow L+2	2.04	79.85	7.47	0.0912	2.9872	3.5166	3.1579
H-10 \rightarrow L+3	1.80	52.22	4.41	0.0720	2.9730	3.5015	3.1431
H-10 \rightarrow L+4	1.86	41.70	6.86	0.1039	2.9738	3.5019	3.1444
H-10 \rightarrow L+5	1.81	56.34	2.58	0.0237	2.9724	3.4996	3.143

H-10 \rightarrow L+6	1.80	63.33	4.06	0.1079	2.9699	3.4988	3.1411
H-10 \rightarrow L+7	2.04	86.42	9.13	0.0496	2.9700	3.5001	3.1411
H-10 \rightarrow L+8	2.29	24.79	6.52	0.0270	2.9723	3.5015	3.1439
H-10 \rightarrow L+9	2.39	32.49	10.23	0.0706	2.9712	3.5001	3.1433
H-10 \rightarrow L+10	4.82	61.82	24.90	0.0174	2.9730	3.5012	3.1447
H-10 \rightarrow L+11	2.18	50.68	0.97	0.0439	2.9738	3.5006	3.1457
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H-11 \rightarrow L	2.60	85.79	15.31	0.1013	2.9804	3.5084	3.1498
H-11 \rightarrow L+1	1.75	61.02	1.74	0.4739	2.9734	3.5033	3.1434
H-11 \rightarrow L+2	1.93	73.08	10.51	0.1660	2.9774	3.5085	3.1483
H-11 \rightarrow L+3	1.79	84.07	3.45	0.1900	2.9802	3.5106	3.1524
H-11 \rightarrow L+4	1.84	78.18	2.39	0.1504	2.9784	3.508	3.1508
H-11 \rightarrow L+5	2.05	100.28	7.56	0.1168	2.9779	3.5078	3.1497
H-11 \rightarrow L+6	1.95	112.50	8.58	0.0107	2.9769	3.5079	3.1486
H-11 \rightarrow L+7	3.42	62.30	17.58	0.2248	2.9751	3.5051	3.1462
H-11 \rightarrow L+8	2.03	71.19	3.68	0.0971	2.9744	3.5043	3.147
H-11 \rightarrow L+9	2.23	105.28	8.38	0.0273	3.0035	3.5371	3.1774
H-11 \rightarrow L+10	2.20	38.25	6.60	0.0089	2.9744	3.5029	3.1466
H-11 \rightarrow L+11	2.51	73.93	10.24	0.0500	2.9751	3.5048	3.1485
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Ground state	-	68.47	0.62	-	2.9768	3.4832	3.1137
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Table S2: Particle-hole state, excitation energy (E_{ex} , magnetic anisotropy energy (MAE), Dipole moment, Transition dipole moment(TDM), Average spin charge on Cr sites for CrBr_3 for singly excited states spin conserving particle-hole state.

Transition	E (eV)	MAE (K)	Dipole (D)	TDM	Spin charge (e)		
					Integrated	Mulliken	Lowdin
H \rightarrow L	1.86	10.79	2.24	0.0336	2.8848	3.3234	3.0177
H \rightarrow L+1	1.79	14.78	2.13	0.1707	2.8841	3.3208	3.0167
H \rightarrow L+2	1.73	13.05	2.62	0.0398	2.8835	3.3202	3.017
H \rightarrow L+3	1.99	14.04	2.21	0.1083	2.8845	3.3213	3.0177
H \rightarrow L+4	1.75	14.64	0.84	0.0288	2.8825	3.3209	3.0174
H \rightarrow L+5	1.79	13.44	2.54	0.0441	2.8836	3.3214	3.0174
H \rightarrow L+6	1.80	16.68	1.98	0.0016	2.8820	3.3211	3.017
H \rightarrow L+7	2.09	18.18	5.16	0.0037	2.8798	3.3176	3.0142
H \rightarrow L+8	2.44	12.51	6.54	0.1683	2.8821	3.3194	3.0169
H \rightarrow L+9	2.20	12.94	6.16	0.1723	2.8763	3.3128	3.0107
H \rightarrow L+10	1.98	11.90	5.90	0.1642	2.8767	3.3137	3.012
H \rightarrow L+11	2.10	21.93	1.82	0.0973	2.8744	3.3118	3.0092
H-1 \rightarrow L	1.62	8.89	0.81	0.0185	2.8847	3.3228	3.0172
H-1 \rightarrow L+1	1.84	14.61	4.67	0.1393	2.8853	3.3222	3.0186
H-1 \rightarrow L+2	1.97	13.88	4.87	0.0129	2.8851	3.3221	3.0188
H-1 \rightarrow L+3	1.71	16.36	1.25	0.0789	2.8836	3.32	3.0164
H-1 \rightarrow L+4	1.77	14.73	2.20	0.0663	2.8824	3.3205	3.0172
H-1 \rightarrow L+5	1.98	15.09	5.17	0.0314	2.8831	3.3209	3.017
H-1 \rightarrow L+6	1.84	15.87	2.04	0.0804	2.8808	3.3188	3.0158
H-1 \rightarrow L+7	2.12	14.30	0.44	0.1132	2.8814	3.3195	3.016
H-1 \rightarrow L+8	1.76	12.03	1.22	0.0859	2.8791	3.3154	3.0139
H-1 \rightarrow L+9	2.07	13.63	1.81	0.2554	2.8787	3.3147	3.0121

H-1 \rightarrow L+10	2.99	10.07	7.43	0.1000	2.8777	3.3143	3.0129
H-1 \rightarrow L+11	5.48	25.11	15.80	0.0604	2.8816	3.3194	3.0168
H-2 \rightarrow L	1.86	10.01	1.00	0.0135	2.8700	3.3055	3.002
H-2 \rightarrow L+1	1.76	11.41	0.93	0.0602	2.8715	3.3048	3.0032
H-2 \rightarrow L+2	1.81	11.67	0.63	0.0495	2.8702	3.3039	3.003
H-2 \rightarrow L+3	1.89	11.16	1.36	0.0356	2.8695	3.303	3.0016
H-2 \rightarrow L+4	1.93	11.17	1.88	0.1521	2.8681	3.3032	3.0019
H-2 \rightarrow L+5	1.89	11.46	2.09	0.0681	2.8689	3.3036	3.0021
H-2 \rightarrow L+6	1.87	12.42	0.18	0.0307	2.8667	3.302	3.0007
H-2 \rightarrow L+7	1.95	12.40	2.00	0.0571	2.8659	3.3012	2.9999
H-2 \rightarrow L+8	2.09	9.03	1.96	0.0268	2.8644	3.2982	2.9988
H-2 \rightarrow L+9	2.04	12.39	2.82	0.0349	2.8631	3.2972	2.997
H-2 \rightarrow L+10	2.08	11.00	1.29	0.0074	2.8633	3.2972	2.9978
H-2 \rightarrow L+11	1.91	19.97	2.31	0.0473	2.8706	3.3038	3.0036
H-3 \rightarrow L	2.27	15.49	5.79	0.0902	2.8756	3.31	3.0075
H-3 \rightarrow L+1	2.73	12.93	12.73	0.0454	2.8716	3.3074	3.0051
H-3 \rightarrow L+2	2.30	13.80	8.62	0.0218	2.8712	3.3062	3.005
H-3 \rightarrow L+3	2.04	15.77	3.39	0.0345	2.8740	3.3067	3.0062
H-3 \rightarrow L+4	2.16	15.58	1.43	0.0289	2.8732	3.3075	3.0068
H-3 \rightarrow L+5	2.72	16.29	11.27	0.0804	2.8739	3.3082	3.0068
H-3 \rightarrow L+6	1.93	17.78	1.05	0.1244	2.8717	3.3061	3.0053
H-3 \rightarrow L+7	2.15	15.61	2.00	0.1160	2.8694	3.3032	3.0031
H-3 \rightarrow L+8	2.41	16.37	2.04	0.0098	2.8712	3.304	3.0049
H-3 \rightarrow L+9	2.02	15.20	1.83	0.0572	2.8661	3.2991	2.9999
H-3 \rightarrow L+10	2.29	13.57	4.11	0.0386	2.8678	3.3008	3.0022
H-3 \rightarrow L+11	5.52	18.74	23.49	0.0471	2.8666	3.301	3.0015

H-4 → L	2.49	15.65	3.52	0.1091	2.6434	3.0537	2.7495
H-4 → L+1	2.48	9.04	4.29	0.0155	2.6437	3.0544	2.7504
H-4 → L+2	2.39	15.72	1.83	0.0481	2.6401	3.0523	2.7462
H-4 → L+3	2.43	13.69	0.80	0.0248	2.6409	3.0537	2.7476
H-4 → L+4	2.41	14.67	0.77	0.0122	2.6295	3.0393	2.7329
H-4 → L+5	2.45	12.68	0.79	0.0446	2.6426	3.055	2.7517
H-4 → L+6	2.40	16.05	0.21	0.0978	2.6324	3.0452	2.7403
H-4 → L+7	3.57	15.05	0.86	0.0520	2.6397	3.0525	2.7459
H-4 → L+8	2.63	22.51	3.70	0.0046	2.6287	3.0437	2.7384
H-4 → L+9	2.52	19.51	0.80	0.0236	2.6390	3.053	2.751
H-4 → L+10	2.72	9.74	1.33	0.0422	2.6172	3.0322	2.7283
H-4 → L+11	2.82	19.42	2.99	0.0168	2.6324	3.0507	2.7453
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H-5 → L	1.90	15.16	2.62	0.1571	2.8734	3.308	3.0048
H-5 → L+1	2.02	14.32	6.07	0.0378	2.8737	3.3068	3.0054
H-5 → L+2	2.17	15.22	6.53	0.0240	2.8733	3.3069	3.0058
H-5 → L+3	2.12	15.25	5.87	0.0059	2.8728	3.3054	3.0042
H-5 → L+4	1.91	15.44	1.91	0.0258	2.8711	3.3055	3.0046
H-5 → L+5	2.08	16.41	4.69	0.0581	2.8722	3.3062	3.0047
H-5 → L+6	2.00	16.07	3.19	0.0939	2.8689	3.3032	3.0027
H-5 → L+7	2.36	16.63	3.40	0.0803	2.8696	3.3039	3.0029
H-5 → L+8	2.06	13.97	0.40	0.0021	2.8676	3.3004	3.0014
H-5 → L+9	2.43	15.63	2.67	0.0149	2.8674	3.3	3.0002
H-5 → L+10	4.02	14.32	10.81	0.0610	2.8698	3.3034	3.0028
H-5 → L+11	6.00	26.21	16.71	0.1083	2.8702	3.3044	3.0041
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H-6 → L	1.98	15.18	2.95	0.0711	2.8668	3.3011	2.9987
H-6 → L+1	1.91	16.00	0.41	0.1598	2.8680	3.3005	2.9998

H-6 \rightarrow L+2	1.94	15.91	2.37	0.0541	2.8667	3.2993	2.9995
H-6 \rightarrow L+3	2.08	16.39	3.93	0.1600	2.8678	3.3002	2.9999
H-6 \rightarrow L+4	1.96	15.52	3.53	0.0310	2.8653	3.2995	2.9993
H-6 \rightarrow L+5	1.99	15.98	2.04	0.0452	2.8660	3.3002	2.9989
H-6 \rightarrow L+6	1.93	17.20	1.18	0.0377	2.8645	3.299	2.9982
H-6 \rightarrow L+7	2.18	16.03	2.32	0.0070	2.8637	3.2981	2.9977
H-6 \rightarrow L+8	2.63	14.40	6.91	0.0633	2.8627	3.2954	2.9968
H-6 \rightarrow L+9	2.25	15.41	2.90	0.0349	2.8604	3.2934	2.9942
H-6 \rightarrow L+10	2.17	11.95	5.85	0.0163	2.8628	3.2962	2.9975
H-6 \rightarrow L+11	2.12	19.81	4.06	0.0116	2.8646	3.3006	2.9996
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H-7 \rightarrow L	1.89	15.36	0.59	0.0284	2.8661	3.3004	2.998
H-7 \rightarrow L+1	1.96	15.76	4.31	0.1974	2.8655	3.2977	2.997
H-7 \rightarrow L+2	2.01	16.84	3.85	0.0057	2.8651	3.298	2.9978
H-7 \rightarrow L+3	1.91	15.77	2.06	0.0742	2.8653	3.2978	2.9974
H-7 \rightarrow L+4	1.97	16.11	1.62	0.0585	2.8642	3.2979	2.9979
H-7 \rightarrow L+5	2.05	16.27	4.73	0.0390	2.8658	3.2994	2.999
H-7 \rightarrow L+6	1.99	15.95	1.28	0.0220	2.8616	3.2956	2.9957
H-7 \rightarrow L+7	2.17	16.18	1.78	0.0350	2.8611	3.295	2.9949
H-7 \rightarrow L+8	2.01	14.15	1.75	0.0055	2.8641	3.2964	2.9977
H-7 \rightarrow L+9	2.23	15.41	1.35	0.0407	2.8618	3.295	2.9956
H-7 \rightarrow L+10	2.99	12.91	7.91	0.0351	2.8593	3.2924	2.9942
H-7 \rightarrow L+11	5.74	24.30	16.20	0.0748	2.8621	3.2958	2.9965
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H-8 \rightarrow L	2.20	10.23	0.28	0.0125	2.8946	3.3303	3.0286
H-8 \rightarrow L+1	2.41	9.85	6.21	0.0086	2.8866	3.3188	3.0189
H-8 \rightarrow L+2	2.05	0.00	3.06	0.0035	2.8839	3.3193	3.0196
H-8 \rightarrow L+3	2.34	11.46	1.47	0.0273	2.8949	3.3293	3.0293

H-8 \rightarrow L+4	2.11	12.40	2.60	0.0081	2.8910	3.3274	3.0275
H-8 \rightarrow L+5	2.21	11.24	3.25	0.0091	2.8880	3.3255	3.0234
H-8 \rightarrow L+6	2.14	13.53	3.39	0.0171	2.8831	3.3191	3.0196
H-8 \rightarrow L+7	2.13	10.32	1.89	0.0149	2.8845	3.3185	3.0194
H-8 \rightarrow L+8	2.64	10.88	1.67	0.0014	2.8838	3.3169	3.0196
H-8 \rightarrow L+9	2.20	9.02	1.06	0.0015	2.8829	3.316	3.0179
H-8 \rightarrow L+10	2.40	9.20	5.49	0.0017	2.8844	3.3178	3.0205
H-8 \rightarrow L+11	5.87	17.58	22.05	0.0008	2.8783	3.313	3.0143
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H-9 \rightarrow L	2.05	9.67	0.83	0.0127	2.8925	3.328	3.0264
H-9 \rightarrow L+1	2.52	15.11	10.59	0.0037	2.8927	3.3279	3.0278
H-9 \rightarrow L+2	2.33	13.67	6.08	0.0022	2.8945	3.3289	3.0297
H-9 \rightarrow L+3	2.17	10.25	3.13	0.0390	2.8877	3.3217	3.0221
H-9 \rightarrow L+4	2.10	11.65	3.00	0.0034	2.8911	3.327	3.0277
H-9 \rightarrow L+5	2.59	10.31	8.73	0.0195	2.8912	3.3264	3.0262
H-9 \rightarrow L+6	2.10	12.05	1.09	0.0072	2.8911	3.3265	3.0273
H-9 \rightarrow L+7	2.21	11.69	2.53	0.0081	2.8908	3.3252	3.0259
H-9 \rightarrow L+8	2.42	10.22	2.16	0.0006	2.8893	3.3226	3.0252
H-9 \rightarrow L+9	2.23	14.53	2.07	0.0002	2.8860	3.3191	3.0208
H-9 \rightarrow L+10	2.67	7.13	6.23	0.0013	2.8858	3.3191	3.0218
H-9 \rightarrow L+11	5.47	24.63	18.63	0.0028	2.8918	3.3269	3.0282
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H-10 \rightarrow L	2.01	13.55	3.90	0.0108	2.8640	3.3031	2.9963
H-10 \rightarrow L+1	2.32	14.49	3.91	0.0012	2.8645	3.3013	2.997
H-10 \rightarrow L+2	2.39	14.84	9.28	0.0213	2.8628	3.3012	2.996
H-10 \rightarrow L+3	2.31	14.22	7.80	0.0175	2.8640	3.3013	2.9966
H-10 \rightarrow L+4	1.99	14.36	1.44	0.0117	2.8621	3.3014	2.9965
H-10 \rightarrow L+5	2.61	14.96	9.34	0.0071	2.8629	3.3017	2.9966

H-10 \rightarrow L+6	2.11	14.99	4.32	0.0151	2.8598	3.2994	2.9944
H-10 \rightarrow L+7	2.07	14.97	1.83	0.0096	2.8594	3.2992	2.9941
H-10 \rightarrow L+8	2.25	11.78	0.87	0.0004	2.8567	3.2954	2.992
H-10 \rightarrow L+9	2.19	12.65	1.70	0.0011	2.8570	3.2955	2.9916
H-10 \rightarrow L+10	2.43	10.14	4.59	0.0010	2.8560	3.2945	2.9918
H-10 \rightarrow L+11	6.62	24.10	24.29	0.0028	2.8598	3.2976	2.9946
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H-11 \rightarrow L	2.14	13.96	5.04	0.1238	2.8790	3.3131	3.0127
H-11 \rightarrow L+1	4.51	13.83	23.07	0.0062	2.8830	3.3149	3.0158
H-11 \rightarrow L+2	4.92	14.03	23.68	0.0600	2.8829	3.3152	3.0168
H-11 \rightarrow L+3	2.23	14.44	7.87	0.0509	2.8755	3.3094	3.0097
H-11 \rightarrow L+4	2.73	14.55	11.72	0.0565	2.8722	3.3047	3.0061
H-11 \rightarrow L+5	2.10	13.56	5.72	0.0792	2.8775	3.3118	3.0131
H-11 \rightarrow L+6	4.02	14.46	16.38	0.0510	2.8790	3.3132	3.0143
H-11 \rightarrow L+7	4.39	13.31	13.49	0.0227	2.8790	3.3131	3.0135
H-11 \rightarrow L+8	2.19	15.49	4.76	0.0151	2.8728	3.3074	3.007
H-11 \rightarrow L+9	5.54	12.54	25.82	0.0122	2.8765	3.3101	3.0118
H-11 \rightarrow L+10	4.91	11.77	13.29	0.0298	2.8774	3.3102	3.0124
H-11 \rightarrow L+11	6.51	21.79	15.72	0.0105	2.8759	3.3097	3.0117
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Ground state	-	14.51	0.15	-	2.8582	3.3037	2.9957
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Table S3: Particle-hole state, excitation energy (E_{ex} , magnetic anisotropy energy (MAE), Dipole moment, Transition dipole moment (TDM), Average spin charge on Cr sites for CrCl_3 for singly excited states spin conserving particle-hole state.

Transition	E (eV)	MAE (K)	Dipole (D)	TDM	Spin charge (e)		
					Integrated	Mulliken	Lowdin
H \rightarrow L	1.91	6.05	2.30	0.0093	2.8270	3.2172	2.9395
H \rightarrow L+1	2.15	7.56	3.76	0.0959	2.8270	3.2152	2.9394
H \rightarrow L+2	2.01	6.77	2.15	0.0757	2.8262	3.2148	2.9395
H \rightarrow L+3	2.27	8.60	1.49	0.0341	2.8253	3.2143	2.9389
H \rightarrow L+4	2.01	5.34	2.64	0.0234	2.8253	3.2147	2.9387
H \rightarrow L+5	2.34	3.65	2.14	0.0114	2.8245	3.2145	2.9391
H \rightarrow L+6	2.69	5.92	1.78	0.0147	2.8245	3.2133	2.9384
H \rightarrow L+7	2.28	3.78	2.20	0.0223	2.8248	3.2139	2.9389
H \rightarrow L+8	2.37	6.21	1.48	0.0841	2.8219	3.2105	2.9362
H \rightarrow L+9	4.51	10.24	11.80	0.0252	2.8245	3.2131	2.9385
H \rightarrow L+10	2.49	9.59	5.38	0.0530	2.8213	3.2095	2.9360
H \rightarrow L+11	2.19	9.46	2.39	0.0068	2.8197	3.2075	2.9353
H-1 \rightarrow L	1.88	3.19	3.20	0.0296	2.8264	3.2164	2.9389
H-1 \rightarrow L+1	2.08	6.94	1.27	0.0411	2.8266	3.2149	2.9385
H-1 \rightarrow L+2	2.09	8.45	3.27	0.0421	2.8261	3.2147	2.9386
H-1 \rightarrow L+3	2.14	9.35	2.98	0.0064	2.8253	3.2143	2.9381
H-1 \rightarrow L+4	2.26	2.69	4.13	0.0090	2.8253	3.2149	2.9391
H-1 \rightarrow L+5	2.42	4.10	5.05	0.0047	2.8239	3.2135	2.9383
H-1 \rightarrow L+6	2.27	6.33	3.92	0.0693	2.8231	3.2118	2.9371
H-1 \rightarrow L+7	2.48	4.58	4.28	0.0625	2.8236	3.2124	2.9378
H-1 \rightarrow L+8	2.44	7.10	5.76	0.0772	2.8211	3.2102	2.9337
H-1 \rightarrow L+9	2.40	9.00	8.37	0.0917	2.8217	3.2099	2.936

H-1 \rightarrow L+10	2.54	7.69	5.89	0.0107	2.8198	3.2086	2.9342
H-1 \rightarrow L+11	3.36	10.85	6.32	0.0125	2.8209	3.2092	2.9362
H-2 \rightarrow L	1.96	5.20	1.47	0.0442	2.8209	3.2093	2.9329
H-2 \rightarrow L+1	1.97	7.28	1.91	0.0756	2.8210	3.2074	2.9326
H-2 \rightarrow L+2	2.01	8.41	2.10	0.0672	2.8209	3.2076	2.9333
H-2 \rightarrow L+3	2.14	9.12	2.37	0.0499	2.8198	3.2072	2.9326
H-2 \rightarrow L+4	2.02	4.42	2.56	0.0543	2.8194	3.2071	2.9326
H-2 \rightarrow L+5	2.03	3.58	2.01	0.0093	2.8178	3.2058	2.9318
H-2 \rightarrow L+6	2.25	5.54	2.17	0.0604	2.8174	3.2045	2.9310
H-2 \rightarrow L+7	2.03	3.83	0.98	0.0142	2.8181	3.2053	2.9318
H-2 \rightarrow L+8	2.41	6.70	5.04	0.0040	2.8153	3.2020	2.9292
H-2 \rightarrow L+9	2.65	7.39	2.68	0.0306	2.8160	3.2023	2.9300
H-2 \rightarrow L+10	2.19	8.09	2.82	0.0100	2.8134	3.1997	2.9275
H-2 \rightarrow L+11	2.18	9.38	0.95	0.0409	2.8137	3.1998	2.9288
H-3 \rightarrow L	2.36	5.71	7.13	0.0427	2.8165	3.2064	2.9270
H-3 \rightarrow L+1	3.08	7.27	12.01	0.0124	2.8186	3.2064	2.9291
H-3 \rightarrow L+2	3.16	5.24	8.37	0.0171	2.8234	3.2117	2.9366
H-3 \rightarrow L+3	2.79	8.76	8.47	0.0200	2.8157	3.2046	2.9268
H-3 \rightarrow L+4	2.95	3.67	11.60	0.0191	2.8170	3.2060	2.9290
H-3 \rightarrow L+5	2.27	3.18	6.32	0.0181	2.8150	3.2039	2.9277
H-3 \rightarrow L+6	2.07	6.48	3.49	0.0135	2.8151	3.2032	2.9274
H-3 \rightarrow L+7	2.20	4.19	4.81	0.0102	2.8135	3.2022	2.9257
H-3 \rightarrow L+8	2.31	7.16	2.53	0.0005	2.8100	3.1983	2.9226
H-3 \rightarrow L+9	4.88	10.89	19.05	0.0038	2.8143	3.2026	2.9265
H-3 \rightarrow L+10	2.39	8.36	0.56	0.0017	2.8093	3.1972	2.9225
H-3 \rightarrow L+11	2.24	8.41	1.79	0.0131	2.8085	3.1962	2.9226

H-4 → L	2.53	5.90	6.29	0.0010	2.8143	3.2019	2.9260
H-4 → L+1	2.76	6.13	11.43	0.0164	2.8168	3.2024	2.9280
H-4 → L+2	2.36	6.99	7.76	0.0411	2.8208	3.2078	2.9327
H-4 → L+3	2.35	8.32	5.58	0.0327	2.8180	3.2047	2.9311
H-4 → L+4	2.24	2.18	4.13	0.0449	2.8133	3.2003	2.9260
H-4 → L+5	2.12	3.50	4.33	0.0700	2.8107	3.1974	2.9241
H-4 → L+6	2.37	5.91	2.46	0.0115	2.8101	3.1961	2.9232
H-4 → L+7	2.10	5.08	1.98	0.0062	2.8100	3.1959	2.9234
H-4 → L+8	2.19	7.43	3.45	0.0119	2.8082	3.1944	2.9221
H-4 → L+9	4.19	9.96	18.04	0.0189	2.8115	3.1972	2.9247
H-4 → L+10	4.05	8.10	13.15	0.0855	2.8084	3.1940	2.9222
H-4 → L+11	2.49	8.28	1.78	0.0245	2.8066	3.1918	2.9217
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H-5 → L	2.04	4.21	2.78	0.0064	2.8154	3.2030	2.9269
H-5 → L+1	2.33	7.42	5.14	0.0209	2.8152	3.2011	2.9267
H-5 → L+2	3.54	8.39	14.25	0.0085	2.8148	3.2013	2.9269
H-5 → L+3	2.25	8.15	0.49	0.0230	2.8123	3.1981	2.9252
H-5 → L+4	2.51	3.34	8.65	0.0399	2.8130	3.2000	2.9258
H-5 → L+5	2.56	3.29	8.57	0.0750	2.8111	3.1987	2.9248
H-5 → L+6	2.37	5.62	5.30	0.0889	2.8120	3.1986	2.9254
H-5 → L+7	2.38	4.94	0.53	0.0279	2.8121	3.1990	2.9257
H-5 → L+8	2.53	11.22	5.38	0.0049	2.8101	3.1954	2.9233
H-5 → L+9	3.26	8.15	3.59	0.0052	2.8093	3.1953	2.9229
H-5 → L+10	2.25	8.58	2.10	0.0355	2.8072	3.1932	2.9216
H-5 → L+11	2.18	9.22	1.52	0.0146	2.8072	3.1934	2.9226
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H-6 → L	2.27	4.14	4.08	0.0897	2.8117	3.1993	2.9233
H-6 → L+1	2.21	6.43	5.55	0.0486	2.8128	3.1995	2.9241

H-6 \rightarrow L+2	2.13	8.02	3.42	0.0410	2.8121	3.1993	2.9240
H-6 \rightarrow L+3	2.26	8.72	1.68	0.0043	2.8093	3.1959	2.9213
H-6 \rightarrow L+4	2.12	3.46	2.10	0.0484	2.8093	3.1959	2.9219
H-6 \rightarrow L+5	2.13	2.65	3.06	0.0142	2.8090	3.1959	2.9228
H-6 \rightarrow L+6	2.43	5.30	6.22	0.0050	2.8084	3.1949	2.9215
H-6 \rightarrow L+7	2.33	3.29	6.45	0.0436	2.8070	3.1930	2.9206
H-6 \rightarrow L+8	2.31	5.85	0.84	0.0068	2.8045	3.1906	2.9182
H-6 \rightarrow L+9	4.31	9.54	15.92	0.0542	2.8078	3.1937	2.9211
H-6 \rightarrow L+10	2.95	8.68	6.70	0.0128	2.8032	3.1892	2.9174
H-6 \rightarrow L+11	2.97	11.88	4.13	0.0160	2.8121	3.1966	2.9234
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H-7 \rightarrow L	2.17	7.04	5.27	0.0751	2.8111	3.1987	2.9229
H-7 \rightarrow L+1	2.48	6.77	6.33	0.0726	2.8125	3.1981	2.9240
H-7 \rightarrow L+2	2.00	7.74	1.29	0.0250	2.8095	3.1949	2.9212
H-7 \rightarrow L+3	2.16	8.75	0.66	0.0215	2.8102	3.1975	2.9226
H-7 \rightarrow L+4	2.26	5.50	5.07	0.0057	2.8110	3.1987	2.9239
H-7 \rightarrow L+5	2.46	2.88	4.91	0.0034	2.8078	3.1950	2.9215
H-7 \rightarrow L+6	2.47	5.17	2.70	0.0068	2.8066	3.1922	2.9193
H-7 \rightarrow L+7	2.24	3.21	1.64	0.0969	2.8079	3.1942	2.9213
H-7 \rightarrow L+8	2.37	6.73	1.81	0.0782	2.8054	3.1915	2.9188
H-7 \rightarrow L+9	5.20	9.59	16.98	0.0323	2.8073	3.1931	2.9205
H-7 \rightarrow L+10	3.52	8.79	10.64	0.0040	2.8063	3.1919	2.9201
H-7 \rightarrow L+11	2.47	8.49	5.61	0.0136	2.8033	3.1886	2.9183
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H-8 \rightarrow L	2.65	6.92	9.40	0.0488	2.8139	3.2052	2.9263
H-8 \rightarrow L+1	3.41	6.95	13.17	0.0138	2.8149	3.2044	2.9271
H-8 \rightarrow L+2	2.84	8.50	8.16	0.0226	2.8150	3.2053	2.9279
H-8 \rightarrow L+3	2.88	8.74	7.22	0.0146	2.8138	3.2047	2.9269

H-8 \rightarrow L+4	4.31	4.95	17.55	0.0197	2.8123	3.2019	2.9256
H-8 \rightarrow L+5	2.38	3.08	5.59	0.0604	2.8105	3.1992	2.9243
H-8 \rightarrow L+6	2.82	4.12	8.05	0.0053	2.8116	3.2016	2.9255
H-8 \rightarrow L+7	2.39	3.05	4.36	0.0107	2.8116	3.2024	2.9259
H-8 \rightarrow L+8	2.46	6.31	1.50	0.0007	2.8081	3.1982	2.9226
H-8 \rightarrow L+9	4.83	9.64	17.67	0.0009	2.8120	3.2013	2.9259
H-8 \rightarrow L+10	2.43	7.58	1.50	0.0073	2.8073	3.1970	2.9225
H-8 \rightarrow L+11	2.29	6.84	0.15	0.0760	2.8063	3.1957	2.9222
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H-9 \rightarrow L	3.30	7.18	9.49	0.0465	2.8196	3.2057	2.9319
H-9 \rightarrow L+1	2.26	6.63	4.02	0.0197	2.8251	3.2097	2.9371
H-9 \rightarrow L+2	3.08	8.08	10.09	0.0107	2.8217	3.2065	2.9340
H-9 \rightarrow L+3	2.59	8.26	2.94	0.0638	2.8200	3.2050	2.9331
H-9 \rightarrow L+4	2.53	2.44	1.69	0.0224	2.8213	3.2072	2.9345
H-9 \rightarrow L+5	2.59	5.38	4.79	0.0489	2.8192	3.2047	2.9330
H-9 \rightarrow L+6	2.69	5.51	3.08	0.0270	2.8184	3.2034	2.9323
H-9 \rightarrow L+7	3.89	7.38	9.19	0.0117	2.8201	3.2058	2.9335
H-9 \rightarrow L+8	4.34	6.61	16.77	0.0206	2.8169	3.2016	2.9309
H-9 \rightarrow L+9	2.22	9.75	1.00	0.0541	2.8147	3.2004	2.9285
H-9 \rightarrow L+10	4.67	9.57	10.86	0.0026	2.8166	3.2009	2.9312
H-9 \rightarrow L+11	4.25	8.34	10.89	0.0822	2.8151	3.1994	2.9303
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H-10 \rightarrow L	2.57	5.37	5.49	0.0791	2.8195	3.2054	2.9313
H-10 \rightarrow L+1	2.28	6.30	2.83	0.0111	2.8192	3.2032	2.9307
H-10 \rightarrow L+2	2.62	6.03	4.96	0.0681	2.8188	3.2036	2.9319
H-10 \rightarrow L+3	3.61	9.21	11.83	0.0370	2.8183	3.2034	2.9308
H-10 \rightarrow L+4	2.50	1.95	3.70	0.0073	2.8180	3.2035	2.9310
H-10 \rightarrow L+5	2.42	3.83	0.54	0.0093	2.8158	3.2016	2.9299

H-10 \rightarrow L+6	2.37	5.80	0.85	0.0193	2.8155	3.2000	2.9289
H-10 \rightarrow L+7	2.53	3.32	3.17	0.0127	2.8158	3.2006	2.9295
H-10 \rightarrow L+8	4.21	7.96	11.64	0.0123	2.8153	3.1995	2.9287
H-10 \rightarrow L+9	3.91	10.13	13.03	0.0078	2.8147	3.1989	2.9280
H-10 \rightarrow L+10	3.93	6.58	10.10	0.0200	2.8136	3.1977	2.9279
H-10 \rightarrow L+11	2.45	8.37	2.71	0.0385	2.8106	3.1945	2.9259
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H-11 \rightarrow L	2.39	2.96	6.08	0.0072	2.8227	3.2087	2.9347
H-11 \rightarrow L+1	2.95	6.25	9.43	0.0268	2.8235	3.2077	2.9349
H-11 \rightarrow L+2	3.59	6.12	14.12	0.0103	2.8222	3.2077	2.9351
H-11 \rightarrow L+3	2.97	8.35	8.52	0.0291	2.8218	3.2066	2.9342
H-11 \rightarrow L+4	2.48	8.28	1.77	0.0143	2.8066	3.1918	2.9217
H-11 \rightarrow L+5	2.81	4.33	6.57	0.0314	2.8180	3.2034	2.9321
H-11 \rightarrow L+6	2.69	5.65	5.25	0.0712	2.8190	3.2035	2.9326
H-11 \rightarrow L+7	2.94	4.10	7.58	0.0172	2.8194	3.2041	2.9329
H-11 \rightarrow L+8	2.39	7.55	2.44	0.0078	2.8161	3.2003	2.9299
H-11 \rightarrow L+9	2.44	5.29	2.42	0.0226	2.8183	3.2036	2.9312
H-11 \rightarrow L+10	2.81	11.22	3.59	0.0416	2.8122	3.1991	2.9251
H-11 \rightarrow L+11	2.57	3.72	2.78		2.8152	3.2032	2.9285
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Ground state	-	1.58	0.26	-	2.8196	3.2079	2.9297
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Table S4: Particle-hole state, excitation energy (E_{ex} , magnetic anisotropy energy (MAE), Dipole moment, Average spin-charge on Cr sites for CrI_3 for singly excited states spin flip particle-hole state.

Transition	E (eV)	MAE (K)	Dipole (D)	Spin charge (e)		
				Integrated	Mulliken	Lowdin
H \rightarrow L	2.14	64.31	1.51	2.7565	3.2594	2.893
H \rightarrow L+1	2.15	64.63	1.68	2.7607	3.2646	2.8992
H \rightarrow L+2	2.24	57.65	0.72	2.7640	3.2682	2.9034
H \rightarrow L+3	2.17	50.10	0.61	2.7654	3.2705	2.9058
H \rightarrow L+4	2.17	51.32	1.54	2.7624	3.2672	2.9023
H \rightarrow L+5	2.18	70.99	2.00	2.7521	3.2591	2.8981
H \rightarrow L+6	2.20	69.31	1.43	2.7449	3.2554	2.8903
H \rightarrow L+7	2.13	61.05	0.89	2.7535	3.2608	2.8964
H \rightarrow L+8	2.24	64.61	3.28	2.7478	3.2559	2.8925
H \rightarrow L+9	2.33	113.73	2.25	2.7576	3.2682	2.9049
H \rightarrow L+10	2.41	58.15	3.02	2.7404	3.2487	2.8847
H \rightarrow L+11	2.43	61.63	4.20	2.7420	3.2552	2.8917
H-1 \rightarrow L	2.17	73.52	0.41	2.7617	3.2637	2.898
H-1 \rightarrow L+1	2.26	69.42	2.01	2.7639	3.2669	2.9021
H-1 \rightarrow L+2	2.22	65.35	2.35	2.7663	3.2704	2.906
H-1 \rightarrow L+3	2.18	63.73	1.89	2.7653	3.2702	2.9073
H-1 \rightarrow L+4	2.18	65.54	1.56	2.7609	3.2652	2.9023
H-1 \rightarrow L+5	2.22	68.47	2.26	2.7637	3.268	2.907
H-1 \rightarrow L+6	2.27	79.55	2.75	2.7473	3.2569	2.8926
H-1 \rightarrow L+7	2.41	72.96	4.00	2.7603	3.2686	2.9035
H-1 \rightarrow L+8	2.18	76.99	1.70	2.7534	3.2599	2.8958
H-1 \rightarrow L+9	2.33	106.93	1.67	2.7585	3.2694	2.9069

H-1 \rightarrow L+10	2.34	65.26	2.30	2.7440	3.251	2.8874
H-1 \rightarrow L+11	2.76	74.08	6.79	2.7536	3.2653	2.9006
H-2 \rightarrow L	2.36	124.29	2.32	2.7778	3.2814	2.9182
H-2 \rightarrow L+1	2.46	114.06	6.70	2.7786	3.2835	2.9216
H-2 \rightarrow L+2	2.52	106.06	6.05	2.7806	3.2864	2.9245
H-2 \rightarrow L+3	2.41	112.39	4.58	2.7835	3.2896	2.9284
H-2 \rightarrow L+4	2.28	113.07	0.46	2.7760	3.2826	2.9222
H-2 \rightarrow L+5	2.25	120.22	3.62	2.7662	3.2747	2.9184
H-2 \rightarrow L+6	2.31	130.84	2.48	2.7568	3.2698	2.9096
H-2 \rightarrow L+7	2.49	126.22	4.15	2.7799	3.2877	2.9252
H-2 \rightarrow L+8	2.34	123.96	0.99	2.7656	3.2742	2.9135
H-2 \rightarrow L+9	2.37	181.33	0.50	2.7709	3.2838	2.9251
H-2 \rightarrow L+10	2.41	121.42	2.36	2.7551	3.2652	2.9053
H-2 \rightarrow L+11	3.03	128.06	8.49	2.7820	3.2932	2.9287
H-3 \rightarrow L	2.33	112.22	5.23	2.7721	3.277	2.9145
H-3 \rightarrow L+1	2.46	118.99	4.52	2.7804	3.2856	2.9239
H-3 \rightarrow L+2	2.44	107.23	3.82	2.7847	3.2905	2.9287
H-3 \rightarrow L+3	2.33	106.09	3.12	2.7806	3.2873	2.9262
H-3 \rightarrow L+4	2.28	106.74	3.11	2.7750	3.2821	2.9219
H-3 \rightarrow L+5	2.25	124.56	1.02	2.7678	3.276	2.9192
H-3 \rightarrow L+6	2.32	125.83	3.42	2.7550	3.2686	2.9086
H-3 \rightarrow L+7	2.48	123.38	1.91	2.7800	3.2888	2.9262
H-3 \rightarrow L+8	2.27	103.50	1.76	2.7602	3.2701	2.9102
H-3 \rightarrow L+9	2.39	176.99	2.14	2.7724	3.2852	2.9258
H-3 \rightarrow L+10	2.49	108.67	3.97	2.7550	3.2649	2.9046
H-3 \rightarrow L+11	2.72	108.06	8.19	2.7548	3.271	2.9121

H-4 → L	2.32	46.18	2.38	2.7588	3.2639	2.9014
H-4 → L+1	2.53	57.52	5.00	2.7762	3.2816	2.9191
H-4 → L+2	2.53	64.83	4.33	2.7831	3.289	2.9263
H-4 → L+3	2.52	51.66	2.34	2.7841	3.2901	2.9274
H-4 → L+4	2.51	52.84	1.59	2.7824	3.2875	2.9239
H-4 → L+5	2.47	59.70	1.99	2.7750	3.281	2.9212
H-4 → L+6	2.47	64.34	3.11	2.7614	3.2732	2.9111
H-4 → L+7	2.87	61.15	4.46	2.7751	3.2838	2.92
H-4 → L+8	2.62	64.07	1.44	2.7702	3.2774	2.9141
H-4 → L+9	2.78	108.89	0.81	2.7764	3.2876	2.9254
H-4 → L+10	2.56	71.72	2.99	2.7503	3.2623	2.9024
H-4 → L+11	2.90	89.57	5.44	2.7485	3.2638	2.905
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H-5 → L	2.38	95.05	3.45	2.7630	3.2658	2.9004
H-5 → L+1	2.41	98.53	4.10	2.7638	3.2673	2.9028
H-5 → L+2	2.32	75.08	2.32	2.7602	3.2642	2.9001
H-5 → L+3	2.29	86.36	2.43	2.7641	3.2691	2.9056
H-5 → L+4	2.32	83.98	0.97	2.7637	3.268	2.9039
H-5 → L+5	2.43	96.53	3.96	2.7593	3.2645	2.9032
H-5 → L+6	2.39	101.69	2.24	2.7476	3.2571	2.8923
H-5 → L+7	2.45	99.31	1.86	2.7585	3.2658	2.9014
H-5 → L+8	2.43	93.80	2.24	2.7455	3.2531	2.8902
H-5 → L+9	2.46	151.28	2.03	2.7624	3.2723	2.909
H-5 → L+10	2.53	83.56	3.63	2.7327	3.2425	2.8815
H-5 → L+11	2.50	87.69	6.14	2.7296	3.2429	2.8822
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H-6 → L	2.64	78.00	8.87	2.7741	3.2779	2.9141
H-6 → L+1	2.67	72.99	8.43	2.7786	3.2836	2.921

H-6 → L+2	2.71	81.32	9.31	2.7769	3.2816	2.9189
H-6 → L+3	2.53	64.78	4.99	2.7689	3.2749	2.9135
H-6 → L+4	2.58	69.57	6.02	2.7744	3.2793	2.9169
H-6 → L+5	2.56	72.77	6.93	2.7703	3.2765	2.9167
H-6 → L+6	2.57	73.38	5.97	2.7580	3.27	2.9089
H-6 → L+7	2.76	85.52	8.64	2.7701	3.2782	2.9148
H-6 → L+8	2.60	76.97	4.64	2.7578	3.2669	2.9065
H-6 → L+9	2.60	113.75	4.55	2.7661	3.2771	2.9159
H-6 → L+10	2.79	72.22	6.65	2.7519	3.2624	2.9016
H-6 → L+11	2.88	83.06	7.68	2.7546	3.2687	2.9093
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H-7 → L	2.35	30.15	1.40	2.7467	3.248	2.8831
H-7 → L+1	2.41	27.92	2.57	2.7501	3.2514	2.8872
H-7 → L+2	2.30	29.28	1.09	2.7523	3.2548	2.8917
H-7 → L+3	2.44	32.51	4.17	2.7562	3.2597	2.8971
H-7 → L+4	2.38	34.50	1.76	2.7545	3.2568	2.8931
H-7 → L+5	2.70	38.99	7.07	2.7590	3.261	2.8981
H-7 → L+6	2.46	35.12	1.88	2.7375	3.2451	2.8808
H-7 → L+7	2.48	39.35	2.55	2.7482	3.2538	2.8903
H-7 → L+8	2.43	36.70	1.02	2.7399	3.2452	2.8822
H-7 → L+9	2.54	62.62	1.55	2.7443	3.2526	2.8908
H-7 → L+10	2.54	27.53	3.03	2.7373	3.2426	2.8784
H-7 → L+11	2.60	26.24	1.99	2.7345	3.2456	2.8834
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H-8 → L	2.54	75.14	2.76	2.7678	3.2699	2.9062
H-8 → L+1	2.62	71.24	2.83	2.7736	3.2764	2.9137
H-8 → L+2	2.62	67.98	4.60	2.7747	3.2778	2.9154
H-8 → L+3	2.44	62.84	1.52	2.7675	3.2713	2.9088

H-8 \rightarrow L+4	2.54	65.61	1.77	2.7713	3.2744	2.9116
H-8 \rightarrow L+5	2.50	76.23	1.73	2.7546	3.2602	2.9014
H-8 \rightarrow L+6	2.74	78.95	3.64	2.7577	3.266	2.9024
H-8 \rightarrow L+7	2.69	73.26	5.69	2.7676	3.2748	2.9122
H-8 \rightarrow L+8	2.64	78.01	3.90	2.7531	3.2596	2.8975
H-8 \rightarrow L+9	2.60	113.27	0.66	2.7601	3.2698	2.9091
H-8 \rightarrow L+10	2.85	70.70	5.89	2.7483	3.2554	2.8933
H-8 \rightarrow L+11	2.82	77.08	4.70	2.7494	3.2631	2.9043
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H-9 \rightarrow L	2.53	37.38	1.66	2.7662	3.2685	2.9043
H-9 \rightarrow L+1	2.48	34.02	1.85	2.7680	3.2708	2.9074
H-9 \rightarrow L+2	2.59	51.48	7.13	2.7710	3.2754	2.9125
H-9 \rightarrow L+3	2.45	29.20	4.17	2.7717	3.2765	2.9142
H-9 \rightarrow L+4	2.49	15.92	2.30	2.7708	3.2749	2.9119
H-9 \rightarrow L+5	2.49	40.42	6.13	2.7571	3.2638	2.9049
H-9 \rightarrow L+6	2.55	38.67	2.34	2.7566	3.2653	2.9013
H-9 \rightarrow L+7	2.67	34.14	4.91	2.7661	3.2726	2.9093
H-9 \rightarrow L+8	2.57	55.36	3.06	2.7551	3.2629	2.9012
H-9 \rightarrow L+9	2.56	93.45	2.76	2.7631	3.2734	2.9124
H-9 \rightarrow L+10	2.68	32.58	1.82	2.7465	3.2542	2.892
H-9 \rightarrow L+11	2.91	49.46	8.52	2.7573	3.2686	2.9049
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H-10 \rightarrow L	2.49	42.45	4.04	2.7530	3.2541	2.8904
H-10 \rightarrow L+1	2.69	52.71	6.59	2.7596	3.2625	2.8997
H-10 \rightarrow L+2	2.49	29.21	5.22	2.7601	3.2635	2.9005
H-10 \rightarrow L+3	2.45	29.83	1.84	2.7602	3.263	2.9004
H-10 \rightarrow L+4	2.43	24.38	3.05	2.7607	3.2627	2.8993
H-10 \rightarrow L+5	2.62	38.52	7.00	2.7443	3.249	2.8897

H-10 \rightarrow L+6	2.72	45.37	6.81	2.7408	3.2494	2.8869
H-10 \rightarrow L+7	2.95	41.72	9.81	2.7564	3.2613	2.8981
H-10 \rightarrow L+8	2.67	36.71	4.83	2.7461	3.2514	2.8879
H-10 \rightarrow L+9	2.51	82.06	2.79	2.7466	3.256	2.8956
H-10 \rightarrow L+10	2.79	34.10	6.94	2.7358	3.2422	2.88
H-10 \rightarrow L+11	3.67	33.68	15.57	2.7529	3.2622	2.8974
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H-11 \rightarrow L	2.47	55.85	4.02	2.7555	3.2581	2.8933
H-11 \rightarrow L+1	2.46	55.98	3.63	2.7596	3.2623	2.8992
H-11 \rightarrow L+2	2.50	43.61	2.67	2.7580	3.2617	2.8988
H-11 \rightarrow L+3	2.51	65.15	3.24	2.7673	3.2722	2.9093
H-11 \rightarrow L+4	2.47	48.41	1.25	2.7623	3.2663	2.9031
H-11 \rightarrow L+5	2.54	68.04	3.28	2.7518	3.2588	2.8994
H-11 \rightarrow L+6	2.57	65.27	2.19	2.7422	3.252	2.8891
H-11 \rightarrow L+7	2.51	59.96	1.44	2.7523	3.2593	2.8967
H-11 \rightarrow L+8	2.69	68.08	3.79	2.7528	3.2599	2.8984
H-11 \rightarrow L+9	2.68	114.93	1.31	2.7586	3.2683	2.9064
H-11 \rightarrow L+10	2.73	57.12	2.55	2.7442	3.2514	2.8884
H-11 \rightarrow L+11	2.64	54.52	0.64	2.7336	3.2458	2.8851
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Ground state	-	68.47	0.62	2.9768	3.4832	3.1137
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Table S5: Particle-hole state, excitation energy (E_{ex} , magnetic anisotropy energy (MAE), Dipole moment, Average spin-charge on Cr sites for CrBr_3 for singly excited states spin flip particle-hole state.

Transition	E (eV)	MAE (K)	Dipole (D)	Spin charge (e)		
				Integrated	Mulliken	Lowdin
H \rightarrow L	2.24	13.90	0.47	2.6559	3.0695	2.7641
H \rightarrow L+1	2.29	9.80	1.34	2.6565	3.0704	2.7653
H \rightarrow L+2	2.30	12.82	1.01	2.6536	3.0692	2.7617
H \rightarrow L+3	2.33	14.84	1.91	2.6543	3.0704	2.7628
H \rightarrow L+4	2.20	12.92	1.67	2.6434	3.0563	2.7483
H \rightarrow L+5	2.33	12.04	1.07	2.6555	3.0713	2.7667
H \rightarrow L+6	2.34	14.30	2.01	2.6446	3.0612	2.7551
H \rightarrow L+7	2.46	13.99	2.50	2.6466	3.0642	2.7573
H \rightarrow L+8	2.52	21.84	2.49	2.6420	3.0602	2.7536
H \rightarrow L+9	2.49	19.38	0.82	2.6520	3.0695	2.7663
H \rightarrow L+10	2.69	7.78	1.73	2.6394	3.0573	2.7492
H \rightarrow L+11	2.61	18.53	1.00	2.6446	3.0664	2.7603
H-1 \rightarrow L	2.25	11.62	0.30	2.6556	3.0697	2.7649
H-1 \rightarrow L+1	2.24	12.26	0.65	2.6544	3.0681	2.7632
H-1 \rightarrow L+2	2.26	13.28	0.22	2.6533	3.0693	2.7626
H-1 \rightarrow L+3	2.32	13.89	1.23	2.6533	3.069	2.7617
H-1 \rightarrow L+4	2.18	13.81	1.35	2.6424	3.0556	2.7485
H-1 \rightarrow L+5	2.31	21.29	1.15	2.6536	3.0688	2.7646
H-1 \rightarrow L+6	2.31	13.86	1.54	2.6426	3.0591	2.7541
H-1 \rightarrow L+7	2.25	13.87	1.91	2.6388	3.0551	2.7506
H-1 \rightarrow L+8	2.44	21.22	1.65	2.6411	3.0596	2.7536
H-1 \rightarrow L+9	2.50	19.07	1.42	2.6539	3.0716	2.768

H-1 \rightarrow L+10	2.67	8.26	3.53	2.6339	3.0529	2.747
H-1 \rightarrow L+11	2.35	20.37	1.63	2.6445	3.0642	2.7608
H-2 \rightarrow L	2.27	13.42	0.36	2.6441	3.0549	2.7509
H-2 \rightarrow L+1	2.27	11.43	0.25	2.6443	3.0551	2.7512
H-2 \rightarrow L+2	2.28	11.91	0.33	2.6421	3.055	2.7486
H-2 \rightarrow L+3	2.29	12.53	0.52	2.6421	3.0552	2.7488
H-2 \rightarrow L+4	2.40	8.54	0.35	2.6327	3.0426	2.7356
H-2 \rightarrow L+5	2.43	10.36	0.28	2.6449	3.0577	2.7544
H-2 \rightarrow L+6	2.68	10.61	1.94	2.6343	3.0473	2.7423
H-2 \rightarrow L+7	2.66	10.42	1.78	2.6344	3.0475	2.7421
H-2 \rightarrow L+8	2.52	17.87	1.29	2.6316	3.0467	2.741
H-2 \rightarrow L+9	2.44	16.53	0.78	2.6432	3.0576	2.7551
H-2 \rightarrow L+10	2.76	2.74	0.29	2.6279	3.0429	2.7361
H-2 \rightarrow L+11	2.62	15.07	0.44	2.6369	3.0542	2.7491
H-3 \rightarrow L	2.48	13.11	2.99	2.6468	3.0565	2.7537
H-3 \rightarrow L+1	2.39	12.36	0.80	2.6460	3.056	2.7536
H-3 \rightarrow L+2	2.42	15.40	1.99	2.6441	3.0558	2.7509
H-3 \rightarrow L+3	2.54	16.31	3.70	2.6453	3.0573	2.7522
H-3 \rightarrow L+4	2.47	14.84	1.13	2.6335	3.0426	2.7375
H-3 \rightarrow L+5	2.51	15.22	2.35	2.6462	3.058	2.7558
H-3 \rightarrow L+6	3.46	13.11	3.28	2.6446	3.0575	2.7515
H-3 \rightarrow L+7	2.53	15.72	1.12	2.6369	3.0492	2.7449
H-3 \rightarrow L+8	2.72	23.48	4.22	2.6327	3.047	2.7428
H-3 \rightarrow L+9	2.46	20.73	0.64	2.6423	3.0556	2.7549
H-3 \rightarrow L+10	2.92	8.74	1.45	2.6302	3.0442	2.7386
H-3 \rightarrow L+11	2.91	20.49	2.97	2.6388	3.0557	2.7516

H-4 → L	2.49	15.55	3.52	2.6434	3.0537	2.7495
H-4 → L+1	2.48	8.99	4.29	2.6437	3.0544	2.7504
H-4 → L+2	2.39	15.71	1.83	2.6401	3.0523	2.7462
H-4 → L+3	2.43	13.62	0.80	2.6409	3.0537	2.7476
H-4 → L+4	2.41	14.58	0.77	2.6295	3.0393	2.7329
H-4 → L+5	2.45	12.56	0.79	2.6426	3.055	2.7517
H-4 → L+6	2.40	15.97	0.21	2.6324	3.0452	2.7403
H-4 → L+7	3.57	14.94	0.86	2.6397	3.0525	2.7459
H-4 → L+8	2.63	22.30	3.70	2.6287	3.0437	2.7384
H-4 → L+9	2.52	19.33	0.80	2.6390	3.053	2.751
H-4 → L+10	2.72	9.70	1.33	2.6172	3.0322	2.7283
H-4 → L+11	2.82	19.28	2.99	2.6324	3.0507	2.7453
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H-5 → L	2.37	10.74	1.42	2.6443	3.0547	2.7518
H-5 → L+1	2.55	13.78	5.22	2.6450	3.055	2.7518
H-5 → L+2	2.41	13.54	0.40	2.6424	3.0547	2.7498
H-5 → L+3	2.43	15.67	4.85	2.6407	3.0527	2.7476
H-5 → L+4	2.42	15.67	1.19	2.6313	3.0411	2.7358
H-5 → L+5	2.46	23.83	2.23	2.6413	3.0528	2.7508
H-5 → L+6	2.59	13.83	0.76	2.6355	3.0484	2.7446
H-5 → L+7	2.54	14.58	1.86	2.6355	3.0479	2.7437
H-5 → L+8	2.53	21.99	1.66	2.6301	3.0448	2.7405
H-5 → L+9	2.53	19.68	1.99	2.6424	3.0565	2.7546
H-5 → L+10	2.85	10.19	3.69	2.6189	3.0346	2.7315
H-5 → L+11	2.70	17.29	1.13	2.6383	3.0542	2.7508
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H-6 → L	2.35	11.62	2.11	2.6406	3.0503	2.7474
H-6 → L+1	2.46	15.05	2.86	2.6414	3.0513	2.7486

H-6 → L+2	2.41	17.58	2.19	2.6384	3.0502	2.745
H-6 → L+3	2.43	13.95	2.35	2.6385	3.0506	2.7454
H-6 → L+4	2.47	14.42	1.93	2.6289	3.0379	2.7321
H-6 → L+5	2.43	15.36	1.84	2.6411	3.0531	2.7508
H-6 → L+6	2.74	14.94	3.44	2.6303	3.0424	2.7387
H-6 → L+7	2.91	14.85	4.58	2.6313	3.0438	2.7397
H-6 → L+8	2.78	22.29	4.07	2.6275	3.0416	2.7372
H-6 → L+9	2.57	21.06	1.28	2.6386	3.0517	2.7508
H-6 → L+10	2.74	8.61	2.20	2.6243	3.0384	2.7327
H-6 → L+11	2.86	16.65	3.30	2.6321	3.0488	2.7448
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H-7 → L	2.40	17.74	0.76	2.6378	3.0473	2.7452
H-7 → L+1	2.34	10.58	1.51	2.6375	3.0469	2.7446
H-7 → L+2	2.37	13.72	0.81	2.6361	3.0477	2.7432
H-7 → L+3	2.41	18.44	1.01	2.6371	3.0489	2.7438
H-7 → L+4	2.55	14.56	1.49	2.6268	3.0355	2.7302
H-7 → L+5	2.51	15.34	2.38	2.6401	3.0516	2.7494
H-7 → L+6	2.65	15.02	1.84	2.6270	3.0382	2.7354
H-7 → L+7	2.62	14.69	0.66	2.6280	3.0397	2.7366
H-7 → L+8	2.64	21.95	1.53	2.6252	3.0389	2.735
H-7 → L+9	2.69	20.80	3.87	2.6368	3.05	2.7489
H-7 → L+10	2.80	9.27	1.29	2.6223	3.0361	2.7307
H-7 → L+11	2.71	17.65	1.51	2.6302	3.0459	2.7428
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H-8 → L	2.50	10.19	0.68	2.6639	3.0753	2.7749
H-8 → L+1	2.55	13.51	0.73	2.6617	3.0735	2.774
H-8 → L+2	2.56	16.07	0.24	2.6600	3.0735	2.7712
H-8 → L+3	2.61	12.14	1.41	2.6618	3.0755	2.7731

H-8 → L+4	2.45	10.74	1.05	2.6464	3.057	2.7547
H-8 → L+5	2.67	17.12	4.06	2.6427	3.0575	2.7599
H-8 → L+6	3.20	7.88	1.00	2.6570	3.0698	2.7651
H-8 → L+7	3.05	10.71	0.62	2.6599	3.0727	2.7677
H-8 → L+8	2.79	20.19	3.16	2.6468	3.063	2.7616
H-8 → L+9	2.78	20.39	5.03	2.6524	3.0673	2.7695
H-8 → L+10	2.92	8.85	0.93	2.6378	3.0545	2.7537
H-8 → L+11	3.01	18.64	3.80	2.6369	3.0584	2.7583
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H-9 → L	2.78	14.14	1.88	2.6550	3.0662	2.7684
H-9 → L+1	2.58	11.19	2.50	2.6576	3.0687	2.7702
H-9 → L+2	2.64	14.20	1.15	2.6535	3.0667	2.7664
H-9 → L+3	2.74	13.14	2.88	2.6601	3.0731	2.7711
H-9 → L+4	2.76	12.18	0.82	2.6453	3.0557	2.7546
H-9 → L+5	4.03	14.55	13.87	2.6552	3.0684	2.7709
H-9 → L+6	4.86	12.78	11.61	2.6245	3.0393	2.7401
H-9 → L+7	2.74	10.55	2.88	2.6603	3.0732	2.7695
H-9 → L+8	3.80	20.69	4.17	2.6386	3.0548	2.7551
H-9 → L+9	3.24	20.24	3.71	2.6570	3.0719	2.7743
H-9 → L+10	3.11	8.88	3.69	2.6317	3.0483	2.7488
H-9 → L+11	3.87	17.01	8.74	2.6413	3.0596	2.7609
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H-10 → L	2.57	13.49	1.53	2.6352	3.0501	2.7433
H-10 → L+1	2.71	11.58	1.91	2.6381	3.0535	2.7454
H-10 → L+2	2.67	12.84	2.19	2.6325	3.0493	2.7405
H-10 → L+3	2.52	16.22	1.29	2.6360	3.0536	2.743
H-10 → L+4	2.68	12.18	1.51	2.6240	3.0376	2.7286
H-10 → L+5	2.87	13.53	2.83	2.6400	3.0577	2.7496

H-10 \rightarrow L+6	3.02	14.21	3.39	2.6253	3.0423	2.7347
H-10 \rightarrow L+7	2.77	13.42	1.43	2.6333	3.0524	2.7408
H-10 \rightarrow L+8	2.84	20.85	4.12	2.6212	3.0404	2.7327
H-10 \rightarrow L+9	3.09	19.26	3.18	2.6366	3.0554	2.7491
H-10 \rightarrow L+10	3.01	8.86	2.48	2.6072	3.027	2.7208
H-10 \rightarrow L+11	3.01	18.46	2.85	2.6305	3.0521	2.7435
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H-11 \rightarrow L	3.71	13.63	15.36	2.6568	3.066	2.7643
H-11 \rightarrow L+1	2.48	8.36	6.92	2.6459	3.057	2.7569
H-11 \rightarrow L+2	3.76	13.59	14.45	2.6523	3.0633	2.7581
H-11 \rightarrow L+3	3.94	12.93	16.45	2.6543	3.0656	2.7613
H-11 \rightarrow L+4	2.73	14.73	3.45	2.6386	3.0488	2.7458
H-11 \rightarrow L+5	4.21	12.38	18.35	2.6550	3.0665	2.7652
H-11 \rightarrow L+6	5.31	12.49	15.77	2.6470	3.0588	2.7561
H-11 \rightarrow L+7	3.65	11.94	15.44	2.6439	3.0558	2.7532
H-11 \rightarrow L+8	4.06	19.60	14.68	2.6449	3.0581	2.7541
H-11 \rightarrow L+9	4.96	18.03	16.60	2.6551	3.0683	2.7681
H-11 \rightarrow L+10	4.03	5.57	15.11	2.6408	3.0539	2.7493
H-11 \rightarrow L+11	3.42	19.97	9.17	2.6206	3.0354	2.7367
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Ground state	-	14.51	0.15	2.8582	3.3037	2.9957
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Table S6: Particle-hole state, excitation energy (E_{ex} , magnetic anisotropy energy (MAE), Dipole moment, Average spin-charge on Cr sites for CrCl_3 for singly excited states spin flip particle-hole state.

Transition	E (eV)	MAE (K)	Dipole (D)	Spin charge (e)		
				Integrated	Mulliken	Lowdin
H \rightarrow L	2.45	10.72	0.93	2.5917	2.959	2.6838
H \rightarrow L+1	2.32	9.86	0.88	2.5915	2.9593	2.6848
H \rightarrow L+2	2.46	8.28	1.51	2.5892	2.9586	2.6818
H \rightarrow L+3	2.43	11.42	2.10	2.5903	2.9605	2.6838
H \rightarrow L+4	2.22	5.06	1.59	2.5785	2.9457	2.6688
H \rightarrow L+5	2.49	12.32	2.88	2.5885	2.9581	2.6854
H \rightarrow L+6	2.21	16.93	0.94	2.5752	2.9441	2.6708
H \rightarrow L+7	2.34	8.72	0.97	2.5829	2.9532	2.6776
H \rightarrow L+8	2.61	4.27	0.99	2.5898	2.9594	2.6878
H \rightarrow L+9	2.54	15.56	1.92	2.5769	2.9478	2.6727
H \rightarrow L+10	2.75	8.81	3.18	2.5717	2.9427	2.6692
H \rightarrow L+11	2.64	12.57	0.97	2.5804	2.9537	2.6791
H-1 \rightarrow L	2.36	9.87	1.07	2.5906	2.9579	2.683
H-1 \rightarrow L+1	2.32	10.97	1.00	2.5902	2.9576	2.6826
H-1 \rightarrow L+2	2.41	9.74	2.35	2.5893	2.9588	2.6823
H-1 \rightarrow L+3	2.40	12.63	1.40	2.5889	2.9587	2.6823
H-1 \rightarrow L+4	2.27	4.41	2.17	2.5788	2.9459	2.6686
H-1 \rightarrow L+5	2.50	15.31	1.99	2.5914	2.9605	2.687
H-1 \rightarrow L+6	2.48	15.86	2.15	2.5781	2.9465	2.6721
H-1 \rightarrow L+7	2.26	17.70	1.20	2.5773	2.9469	2.6725
H-1 \rightarrow L+8	2.77	5.35	2.38	2.5904	2.9604	2.6877
H-1 \rightarrow L+9	2.53	13.98	0.63	2.5754	2.9462	2.671

H-1 \rightarrow L+10	2.63	8.24	1.95	2.5678	2.9384	2.664
H-1 \rightarrow L+11	2.48	10.34	1.65	2.5861	2.9574	2.684
H-2 \rightarrow L	2.27	9.94	0.57	2.5860	2.9517	2.6777
H-2 \rightarrow L+1	2.26	10.33	0.75	2.5855	2.9512	2.6773
H-2 \rightarrow L+2	2.29	9.40	0.48	2.5842	2.952	2.6763
H-2 \rightarrow L+3	2.29	12.48	0.47	2.5843	2.9527	2.6768
H-2 \rightarrow L+4	2.42	3.99	0.44	2.5747	2.9402	2.6632
H-2 \rightarrow L+5	2.47	14.14	0.34	2.5869	2.955	2.6817
H-2 \rightarrow L+6	2.62	16.79	2.52	2.5759	2.9433	2.6689
H-2 \rightarrow L+7	2.56	14.47	1.05	2.5762	2.944	2.6693
H-2 \rightarrow L+8	2.46	3.60	0.99	2.5867	2.9549	2.6832
H-2 \rightarrow L+9	2.47	10.98	0.61	2.5727	2.9418	2.6667
H-2 \rightarrow L+10	2.69	5.27	0.50	2.5695	2.9386	2.6626
H-2 \rightarrow L+11	2.68	11.74	0.34	2.5814	2.9517	2.6781
H-3 \rightarrow L	2.48	10.32	4.28	2.5817	2.949	2.6719
H-3 \rightarrow L+1	2.49	10.15	2.56	2.5826	2.9502	2.6725
H-3 \rightarrow L+2	2.47	10.37	2.23	2.5822	2.9523	2.6724
H-3 \rightarrow L+3	2.40	11.44	2.16	2.5790	2.9485	2.6703
H-3 \rightarrow L+4	2.29	3.39	0.92	2.5660	2.9325	2.6541
H-3 \rightarrow L+5	2.46	14.27	1.46	2.5826	2.9519	2.676
H-3 \rightarrow L+6	2.36	16.42	1.29	2.5699	2.9381	2.6621
H-3 \rightarrow L+7	2.37	14.51	0.55	2.5715	2.9405	2.6631
H-3 \rightarrow L+8	2.59	3.54	1.74	2.5802	2.949	2.6757
H-3 \rightarrow L+9	2.58	10.36	2.07	2.5666	2.9368	2.6598
H-3 \rightarrow L+10	2.63	3.87	2.40	2.5636	2.9337	2.6561
H-3 \rightarrow L+11	2.56	12.43	0.95	2.5755	2.9469	2.671

H-4 \rightarrow L	2.37	9.64	1.88	2.5787	2.9433	2.6702
H-4 \rightarrow L+1	2.53	10.68	5.19	2.5792	2.944	2.6707
H-4 \rightarrow L+2	2.78	8.83	7.09	2.5776	2.9444	2.6694
H-4 \rightarrow L+3	2.32	12.80	1.82	2.5759	2.943	2.6683
H-4 \rightarrow L+4	2.54	3.83	2.92	2.5662	2.9304	2.6548
H-4 \rightarrow L+5	2.68	14.09	2.63	2.5815	2.949	2.6758
H-4 \rightarrow L+6	2.75	15.40	4.72	2.5669	2.933	2.6594
H-4 \rightarrow L+7	2.61	15.33	1.10	2.5695	2.9363	2.6623
H-4 \rightarrow L+8	2.76	4.12	4.37	2.5797	2.9469	2.6757
H-4 \rightarrow L+9	2.53	12.07	2.36	2.5640	2.932	2.6583
H-4 \rightarrow L+10	2.71	4.84	1.58	2.5609	2.929	2.6545
H-4 \rightarrow L+11	2.80	10.86	2.87	2.5744	2.9436	2.6707
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H-5 \rightarrow L	2.81	10.61	7.69	2.5790	2.9442	2.6703
H-5 \rightarrow L+1	2.28	10.25	1.61	2.5779	2.9432	2.6696
H-5 \rightarrow L+2	2.43	8.27	3.21	2.5765	2.9434	2.6684
H-5 \rightarrow L+3	2.68	11.48	7.07	2.5771	2.9449	2.6696
H-5 \rightarrow L+4	2.58	4.31	4.27	2.5669	2.9318	2.6555
H-5 \rightarrow L+5	2.86	13.41	6.13	2.5800	2.9476	2.6744
H-5 \rightarrow L+6	2.61	16.86	2.62	2.5699	2.9366	2.6626
H-5 \rightarrow L+7	2.96	12.97	6.43	2.5691	2.9366	2.6618
H-5 \rightarrow L+8	2.70	3.34	5.94	2.5784	2.9454	2.6749
H-5 \rightarrow L+9	2.64	12.26	4.71	2.5648	2.9334	2.6589
H-5 \rightarrow L+10	2.87	4.97	5.93	2.5597	2.9283	2.6538
H-5 \rightarrow L+11	3.01	11.62	6.09	2.5726	2.9427	2.669
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H-6 \rightarrow L	2.48	9.94	3.84	2.5752	2.9398	2.6661
H-6 \rightarrow L+1	2.41	11.61	2.48	2.5762	2.9411	2.6676

H-6 \rightarrow L+2	2.29	9.57	0.85	2.5736	2.9404	2.6652
H-6 \rightarrow L+3	2.44	11.99	2.63	2.5741	2.9415	2.6663
H-6 \rightarrow L+4	2.55	4.46	3.39	2.5646	2.9291	2.6535
H-6 \rightarrow L+5	2.63	14.73	4.47	2.5777	2.9452	2.6717
H-6 \rightarrow L+6	2.82	15.50	4.96	2.5661	2.9327	2.6586
H-6 \rightarrow L+7	2.74	12.73	4.13	2.5672	2.934	2.6597
H-6 \rightarrow L+8	2.52	3.41	2.19	2.5755	2.9421	2.6716
H-6 \rightarrow L+9	2.68	9.38	3.10	2.5629	2.9316	2.6574
H-6 \rightarrow L+10	2.74	4.12	2.91	2.5608	2.9292	2.6542
H-6 \rightarrow L+11	2.74	8.22	2.84	2.5679	2.9372	2.6644
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H-7 \rightarrow L	2.39	11.35	2.90	2.5758	2.9405	2.667
H-7 \rightarrow L+1	2.52	11.22	4.63	2.5760	2.9408	2.6677
H-7 \rightarrow L+2	2.46	9.14	2.62	2.5742	2.9416	2.666
H-7 \rightarrow L+3	2.43	11.49	1.75	2.5733	2.9405	2.6656
H-7 \rightarrow L+4	2.51	4.46	0.75	2.5633	2.9276	2.652
H-7 \rightarrow L+5	2.55	13.83	3.65	2.5758	2.9429	2.6704
H-7 \rightarrow L+6	2.71	15.34	1.80	2.5666	2.9332	2.6594
H-7 \rightarrow L+7	2.77	10.72	4.33	2.5659	2.9326	2.6584
H-7 \rightarrow L+8	2.72	3.24	3.58	2.5761	2.9433	2.6723
H-7 \rightarrow L+9	2.66	11.62	1.69	2.5622	2.9301	2.6562
H-7 \rightarrow L+10	2.79	4.79	1.10	2.5597	2.9279	2.654
H-7 \rightarrow L+11	2.90	11.92	3.24	2.5688	2.9385	2.6652
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H-8 \rightarrow L	2.45	11.83	1.49	2.5806	2.95	2.6725
H-8 \rightarrow L+1	2.73	11.91	6.35	2.5799	2.9487	2.6721
H-8 \rightarrow L+2	2.52	9.89	0.45	2.5790	2.9506	2.6713
H-8 \rightarrow L+3	2.49	11.55	2.42	2.5779	2.9494	2.671

H-8 \rightarrow L+4	2.50	5.65	1.24	2.5680	2.9362	2.6572
H-8 \rightarrow L+5	2.56	15.62	1.97	2.5804	2.9518	2.6758
H-8 \rightarrow L+6	2.68	15.38	3.05	2.5660	2.9342	2.6604
H-8 \rightarrow L+7	2.74	12.66	2.47	2.5716	2.9417	2.6651
H-8 \rightarrow L+8	2.74	1.72	2.42	2.5801	2.9515	2.6772
H-8 \rightarrow L+9	2.80	10.44	3.70	2.5661	2.9378	2.6608
H-8 \rightarrow L+10	2.77	4.18	2.71	2.5630	2.9344	2.6578
H-8 \rightarrow L+11	2.92	12.27	2.95	2.5744	2.9475	2.6717
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H-9 \rightarrow L	3.30	7.18	11.08	2.5862	2.9502	2.6791
H-9 \rightarrow L+1	2.26	11.13	4.02	2.5862	2.9501	2.6786
H-9 \rightarrow L+2	3.08	9.88	10.09	2.5859	2.9516	2.6786
H-9 \rightarrow L+3	2.59	11.56	2.94	2.5845	2.9507	2.6773
H-9 \rightarrow L+4	2.53	4.69	1.69	2.5761	2.9398	2.6654
H-9 \rightarrow L+5	2.59	14.80	4.79	2.5881	2.9542	2.6826
H-9 \rightarrow L+6	2.69	16.59	3.08	2.5785	2.9441	2.6711
H-9 \rightarrow L+7	3.89	11.96	9.19	2.5772	2.9428	2.6702
H-9 \rightarrow L+8	4.34	6.61	15.07	2.5837	2.9516	2.6766
H-9 \rightarrow L+9	2.22	9.94	1.00	2.5743	2.9421	2.6693
H-9 \rightarrow L+10	4.67	5.68	10.86	2.5714	2.9388	2.6652
H-9 \rightarrow L+11	4.25	6.15	10.89	2.5800	2.9483	2.6739
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H-10 \rightarrow L	2.57	10.48	5.49	2.5816	2.9444	2.6738
H-10 \rightarrow L+1	2.28	10.65	2.83	2.5812	2.944	2.6734
H-10 \rightarrow L+2	2.62	9.29	4.96	2.5795	2.9444	2.672
H-10 \rightarrow L+3	3.61	13.04	11.83	2.5779	2.9428	2.6712
H-10 \rightarrow L+4	2.50	4.49	3.70	2.5696	2.9324	2.6595
H-10 \rightarrow L+5	2.42	10.34	0.54	2.5874	2.9537	2.6814

H-10 \rightarrow L+6	2.37	11.35	0.85	2.5720	2.9369	2.6653
H-10 \rightarrow L+7	2.53	8.79	3.17	2.5657	2.9304	2.6606
H-10 \rightarrow L+8	4.21	2.93	11.64	2.5804	2.946	2.6772
H-10 \rightarrow L+9	3.91	11.04	13.03	2.5742	2.9412	2.6672
H-10 \rightarrow L+10	3.93	5.07	10.10	2.5681	2.9349	2.6609
H-10 \rightarrow L+11	2.45	10.48	2.71	2.5830	2.9512	2.6787
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H-11 \rightarrow L	2.61	10.20	0.90	2.5847	2.9477	2.6761
H-11 \rightarrow L+1	2.66	11.16	3.92	2.5867	2.9498	2.6783
H-11 \rightarrow L+2	2.58	11.13	3.86	2.5757	2.9406	2.6697
H-11 \rightarrow L+3	2.68	12.85	1.92	2.5843	2.9499	2.6772
H-11 \rightarrow L+4	2.77	4.20	4.78	2.5736	2.9367	2.6641
H-11 \rightarrow L+5	2.88	14.10	2.52	2.5904	2.9564	2.6845
H-11 \rightarrow L+6	3.71	16.38	3.45	2.5778	2.943	2.6705
H-11 \rightarrow L+7	2.72	12.68	2.59	2.5720	2.9368	2.6666
H-11 \rightarrow L+8	3.15	3.20	2.92	2.5901	2.9566	2.6852
H-11 \rightarrow L+9	2.94	10.19	4.52	2.5620	2.9303	2.6602
H-11 \rightarrow L+10	2.98	5.83	2.76	2.5686	2.9351	2.6623
H-11 \rightarrow L+11	3.03	11.57	3.02	2.5852	2.9531	2.681
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Ground state	-	1.58	0.26	2.8196	3.2079	2.9297
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