Electronic Supplementary Information

Enhanced Optical Anisotropy via Dimensional Control in the Alkali-metal Chalcogenides

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Table S1. The obtained bond length and bond populations.

Table S1.1. The bond length and bond population of $KPSe_6$.

Bond	Population	Length(Å)
P01-Se09	0.55	2.13916
P03-Se21	0.55	2.13916
P04-Se15	0.55	2.13916
P02-Se03	0.55	2.13916
P04-Se19	0.59	2.14585
P02-Se07	0.59	2.14585
P03-Se13	0.59	2.14585
P01-Se01	0.59	2.14585
P02-Se22	0.25	2.273
P03-Se04	0.25	2.273
P04-Se10	0.25	2.273
P01-Se16	0.25	2.273
P04-Se24	0.28	2.30731
P02-Se12	0.28	2.30731
P01-Se06	0.28	2.30731
P03-Se18	0.28	2.30731
Se06-Se17	-0.41	2.34662
Se11-Se24	-0.41	2.34662
Se12-Se23	-0.41	2.34662
Se05-Se18	-0.41	2.34662
Se02-Se04	-0.84	2.37277
Se14-Se16	-0.84	2.37277
Se08-Se10	-0.84	2.37277

Se20-Se22	-0.84	2.37277
Se05-Se14	0.11	2.41859
Se02-Se17	0.11	2.41859
Se11-Se20	0.11	2.41859
Se08-Se23	0.11	2.41859

Table S1.2. The bond length and bond population of $Na_2Ge_2Se_5$.

Bond	Population	Length(Å)
Ge03-Se10	1.37	2.30259
Ge05-Se15	1.37	2.30259
Ge01-Se05	1.37	2.30259
Ge07-Se20	1.37	2.30259
Ge06-Se09	0.98	2.31077
Ge02-Se19	0.98	2.31077
Ge04-Se14	0.98	2.31077
Ge08-Se04	0.98	2.31077
Ge02-Se17	0.59	2.3986
Ge08-Se02	0.59	2.3986
Ge06-Se07	0.59	2.3986
Ge04-Se12	0.59	2.3986
Ge04-Se11	0.45	2.40287
Ge08-Se01	0.45	2.40287
Ge02-Se16	0.45	2.40287
Ge06-Se06	0.45	2.40287
Ge07-Se16	0.36	2.41412

Ge03-Se06	0.36	2.41412
Ge05-Se11	0.36	2.41412
Ge01-Se01	0.36	2.41412
Ge05-Se07	0.12	2.4149
Ge01-Se17	0.12	2.4149
Ge07-Se02	0.12	2.4149
Ge03-Se12	0.12	2.4149
Ge02-Se18	0.63	2.42085
Ge08-Se03	0.63	2.42085
Ge04-Se13	0.63	2.42085
Ge06-Se08	0.63	2.42085
Ge03-Se13	0.56	2.42385
Ge01-Se18	0.56	2.42385
Ge05-Se08	0.56	2.42385
Ge07-Se03	0.56	2.42385
Na03-Se19	0.03	2.95359
Na07-Se09	0.03	2.95359
Na05-Se04	0.03	2.95359
Na01-Se14	0.03	2.95359
Na07-Se04	-0.06	2.98468
Na03-Se14	-0.06	2.98468
Na05-Se09	-0.06	2.98468
Na01-Se19	-0.06	2.98468

Table S1.3. The bond length and bond population of $Li_2In_2GeSe_6$.

Bond	Population	Length(Å)
Ge02-Se12	0.63	2.33841
Ge01-Se06	0.63	2.33841
Ge02-Se10	1	2.34413
Ge01-Se04	1	2.34413
Ge01-Se03	0.54	2.34698
Ge02-Se09	0.54	2.34698
Ge02-Se11	0.15	2.35546
Ge01-Se05	0.15	2.35546
Se06-In02	0.2	2.52937
Se12-In04	0.2	2.52937
Se01-In01	0.35	2.53347
Se07-In03	0.35	2.53347
Se07-In02	0	2.53463
Se01-In04	0	2.53463
Se05-In01	0.43	2.53599
Se11-In03	0.43	2.53599
Se02-In04	0.43	2.54028
Se08-In02	0.43	2.54028
Se08-In01	0.61	2.54526
Se02-In03	0.61	2.54526
Se09-In03	0.4	2.54645
Se03-In01	0.4	2.54645
Se04-In04	0.75	2.55322
Se10-In02	0.75	2.55322

Table S2. The obtained Born effective charges.

Table S2.1. The Born effective charge of $KPSe_6$.

P1	1.98494	-0.06681	-0.95555
	0.09458	0.86054	0.12785
	0.64406	-0.23213	3.08519
P2	1.98494	-0.06681	0.95555
	0.09458	0.86054	-0.12785
	-0.64406	0.23213	3.08519
P3	1.98494	0.06681	-0.95555
	-0.09458	0.86054	-0.12785
	0.64406	0.23213	3.08519
P4	1.98494	0.06681	0.95555
	-0.09458	0.86054	0.12785
	-0.64406	-0.23213	3.08519
K1	1.48273	0.05897	-0.0025
	0.14612	0.52914	-0.14975
	0.02929	0.01645	1.47079
K2	1.48273	0.05897	0.0025
	0.14612	0.52914	0.14975
	-0.02929	-0.01645	1.47079
K3	1.48273	-0.05897	-0.0025
	-0.14612	0.52914	0.14975
	0.02929	-0.01645	1.47079
K4	1.48273	-0.05897	0.0025
	-0.14612	0.52914	-0.14975
	-0.02929	0.01645	1.47079

Se1	-1.01893	0.0509	0.36038
	-0.04837	-0.84683	-0.35969
	-0.26703	0.01626	-1.6801
Se2	-0.51676	-0.10067	-0.15051
	-0.69211	1.01059	1.41338
	-0.08687	0.54726	2.61045
Se3	-0.86891	-0.06752	0.59618
	0.19276	-0.82824	0.05492
	0.26212	0.09621	-2.53449
Se4	-0.95017	0.11731	0.08031
	0.06332	-0.18813	-0.05329
	-0.0327	0.04511	-0.92525
Se5	-0.54381	-0.24223	0.27713
	0.11974	-0.1497	-0.30164
	0.37004	-0.14775	-1.16915
Se6	0.4309	0.30414	0.06519
	0.05366	-0.38738	0.9326
	0.40043	0.42714	-0.85744
Se7	-1.01893	0.0509	-0.36038
	-0.04837	-0.84683	0.35969
	0.26703	-0.01626	-1.6801
Se8	-0.51676	-0.10067	0.15051
	-0.69211	1.01059	-1.41338
	0.08687	-0.54726	2.61045
Se9	-0.86891	-0.06752	-0.59618
	0.19276	-0.82824	-0.05492

	-0.26212	-0.09621	-2.53449
Se10	-0.95017	0.11731	-0.08031
	0.06332	-0.18813	0.05329
	0.0327	-0.04511	-0.92525
Se11	-0.54381	-0.24223	-0.27713
	0.11974	-0.1497	0.30164
	-0.37004	0.14775	-1.16915
Se12	0.4309	0.30414	-0.06519
	0.05366	-0.38738	-0.9326
	-0.40043	-0.42714	-0.85744
Se13	-1.01893	-0.0509	0.36038
	0.04837	-0.84683	0.35969
	-0.26703	-0.01626	-1.6801
Se14	-0.51676	0.10067	-0.15051
	0.69211	1.01059	-1.41338
	-0.08687	-0.54726	2.61045
Se15	-0.86891	0.06752	0.59618
	-0.19276	-0.82824	-0.05492
	0.26212	-0.09621	-2.53449
Se16	-0.95017	-0.11731	0.08031
	-0.06332	-0.18813	0.05329
	-0.0327	-0.04511	-0.92525
Se17	-0.54381	0.24223	0.27713
	-0.11974	-0.1497	0.30164
	0.37004	0.14775	-1.16915
Se18	0.4309	-0.30414	0.06519

	-0.05366	-0.38738	-0.9326
	0.40043	-0.42714	-0.85744
Se19	-1.01893	-0.0509	-0.36038
	0.04837	-0.84683	-0.35969
	0.26703	0.01626	-1.6801
Se20	-0.51676	0.10067	0.15051
	0.69211	1.01059	1.41338
	0.08687	0.54726	2.61045
Se21	-0.86891	0.06752	-0.59618
	-0.19276	-0.82824	0.05492
	-0.26212	0.09621	-2.53449
Se22	-0.95017	-0.11731	-0.08031
	-0.06332	-0.18813	-0.05329
	0.0327	0.04511	-0.92525
Se23	-0.54381	0.24223	-0.27713
	-0.11974	-0.1497	-0.30164
	-0.37004	-0.14775	-1.16915
Se24	0.4309	-0.30414	-0.06519
	-0.05366	-0.38738	0.9326
	-0.40043	0.42714	-0.85744

Table S2.2 The Born effective charge of $Na_2Ge_2Se_5$.

Na1	1.14876	-0.10752	-0.01857
	-0.07148	1.20312	-0.00868
	-0.39065	0.01732	1.2741

Na2	1.42019	0.14595	0.08763
	0.03942	1.2959	0.03788
	0.30663	0.00311	1.2201
Na3	1.14876	-0.10752	0.01857
	-0.07148	1.20312	0.00868
	0.39065	-0.01732	1.2741
Na4	1.42019	0.14595	-0.08763
	0.03942	1.2959	-0.03788
	-0.30663	-0.00311	1.2201
Na5	1.14876	0.10752	-0.01857
	0.07148	1.20312	0.00868
	-0.39065	-0.01732	1.2741
Na6	1.42019	-0.14595	0.08763
	-0.03942	1.2959	-0.03788
	0.30663	-0.00311	1.2201
Na7	1.14876	0.10752	0.01857
	0.07148	1.20312	-0.00868
	0.39065	0.01732	1.2741
Na8	1.42019	-0.14595	-0.08763
	-0.03942	1.2959	0.03788
	-0.30663	0.00311	1.2201
Ge1	2.54904	0.3865	0.50826
	-0.35204	2.7167	-0.83849
	0.03789	0.67947	2.82527
Ge2	2.22334	-0.14784	0.85833
	0.25492	2.6215	0.84964

	-0.35593	-0.83292	3.18367
Ge3	2.54904	0.3865	-0.50826
	-0.35204	2.7167	0.83849
	-0.03789	-0.67947	2.82527
Ge4	2.22334	-0.14784	-0.85833
	0.25492	2.6215	-0.84964
	0.35593	0.83292	3.18367
Ge5	2.54904	-0.3865	0.50826
	0.35204	2.7167	0.83849
	0.03789	-0.67947	2.82527
Ge6	2.22334	0.14784	0.85833
	-0.25492	2.6215	-0.84964
	-0.35593	0.83292	3.18367
Ge7	2.54904	-0.3865	-0.50826
	0.35204	2.7167	-0.83849
	-0.03789	0.67947	2.82527
Ge8	2.22334	0.14784	-0.85833
	-0.25492	2.6215	0.84964
	0.35593	-0.83292	3.18367
Se1	-1.20494	0.29929	-0.59337
	0.34626	-1.18812	-0.11112
	-0.52207	-0.03186	-2.3568
Se2	-1.05823	0.27289	0.26916
	0.21498	-1.95893	-0.42185
	0.05057	-0.53286	-1.64374
Se3	-1.33679	-0.00414	0.45323

	0.0371	-2.0132	0.49374
	0.19084	0.67966	-1.5594
Se4	-1.88614	-0.0282	0.05923
	0.0605	-1.375	-0.15675
	-0.01904	0.08147	-1.39823
Se5	-1.85524	-0.1411	0.1895
	-0.00048	-1.30197	0.10219
	0.52966	-0.02991	-1.54498
Se6	-1.20494	0.29929	0.59337
	0.34626	-1.18812	0.11112
	0.52207	0.03186	-2.3568
Se7	-1.05823	0.27289	-0.26916
	0.21498	-1.95893	0.42185
	-0.05057	0.53286	-1.64374
Se8	-1.33679	-0.00414	-0.45323
	0.0371	-2.0132	-0.49374
	-0.19084	-0.67966	-1.5594
Se9	-1.88614	-0.0282	-0.05923
	0.0605	-1.375	0.15675
	0.01904	-0.08147	-1.39823
Se10	-1.85524	-0.1411	-0.1895
	-0.00048	-1.30197	-0.10219
	-0.52966	0.02991	-1.54498
Se11	-1.20494	-0.29929	-0.59337
	-0.34626	-1.18812	0.11112
	-0.52207	0.03186	-2.3568

Se12	-1.05823	-0.27289	0.26916
	-0.21498	-1.95893	0.42185
	0.05057	0.53286	-1.64374
Se13	-1.33679	0.00414	0.45323
	-0.0371	-2.0132	-0.49374
	0.19084	-0.67966	-1.5594
Se14	-1.88614	0.0282	0.05923
	-0.0605	-1.375	0.15675
	-0.01904	-0.08147	-1.39823
Se15	-1.85524	0.1411	0.1895
	0.00048	-1.30197	-0.10219
	0.52966	0.02991	-1.54498
Se16	-1.20494	-0.29929	0.59337
	-0.34626	-1.18812	-0.11112
	0.52207	-0.03186	-2.3568
Se17	-1.05823	-0.27289	-0.26916
	-0.21498	-1.95893	-0.42185
	-0.05057	-0.53286	-1.64374
Se18	-1.33679	0.00414	-0.45323
	-0.0371	-2.0132	0.49374
	-0.19084	0.67966	-1.5594
Se19	-1.88614	0.0282	-0.05923
	-0.0605	-1.375	-0.15675
	0.01904	0.08147	-1.39823
Se20	-1.85524	0.1411	-0.1895
	0.00048	-1.30197	0.10219

	-0.52966	-0.02991	-1.54498

Table S2.3. The Born effective charge of $Li_2In_2GeSe_6$.

Li1	1.23601	0.33619	0.09174
	0.20012	1.25867	0.04804
	0.03768	-0.03782	1.16862
Li2	1.2681	0.22605	0.02274
	-0.14565	1.43077	-0.1251
	0.09195	-0.03224	1.10749
Li3	1.23601	-0.33619	0.09174
	-0.20012	1.25867	-0.04804
	0.03768	0.03782	1.16862
Li4	1.2681	-0.22605	0.02274
	0.14565	1.43077	0.1251
	0.09195	0.03224	1.10749
Ge1	2.45672	0.29643	0.02463
	0.68865	2.49519	0.25574
	-0.32697	-0.13327	2.95468
Ge2	2.45672	-0.29643	0.02463
	-0.68865	2.49519	-0.25574
	-0.32697	0.13327	2.95468
Se1	-1.93171	-0.34357	0.25084
	-0.2832	-1.55581	-0.17453
	0.22465	-0.22634	-2.07966
Se2	-1.80445	0.14186	-0.11537

	0.11181	-1.40453	0.11314
	-0.08578	0.01094	-2.06561
Se3	-1.58111	-0.32341	-0.21185
	-0.34817	-1.22983	0.19828
	-0.02818	0.24753	-1.44123
Se4	-1.23627	-0.17205	-0.25417
	-0.30727	-2.00144	-0.39245
	-0.22211	-0.54474	-1.74404
Se5	-1.736	-0.43676	0.54425
	-0.12397	-2.01358	0.24951
	0.43093	0.29479	-1.18402
Se6	-1.72879	-0.25783	0.10522
	-0.17687	-0.89669	0.25233
	0.04578	0.21973	-2.61053
Se7	-1.93171	0.34357	0.25084
	0.2832	-1.55581	0.17453
	0.22465	0.22634	-2.07966
Se8	-1.80445	-0.14186	-0.11537
	-0.11181	-1.40453	-0.11314
	-0.08578	-0.01094	-2.06561
Se9	-1.58111	0.32341	-0.21185
	0.34817	-1.22983	-0.19828
	-0.02818	-0.24753	-1.44123
Se10	-1.23627	0.17205	-0.25417
	0.30727	-2.00144	0.39245
	-0.22211	0.54474	-1.74404

Se11	-1.736	0.43676	0.54425
	0.12397	-2.01358	-0.24951
	0.43093	-0.29479	-1.18402
Se12	-1.72879	0.25783	0.10522
	0.17687	-0.89669	-0.25233
	0.04578	-0.21973	-2.61053
In1	2.70167	0.55969	-0.06433
	0.12989	1.94474	0.04784
	-0.19873	-0.01491	2.8815
In2	2.35584	-0.07659	-0.39369
	0.16756	1.97251	0.27868
	0.03077	-0.12032	3.01278
In3	2.70167	-0.55969	-0.06433
	-0.12989	1.94474	-0.04784
	-0.19873	0.01491	2.8815
In4	2.35584	0.07659	-0.39369
	-0.16756	1.97251	-0.27868
	0.03077	0.12032	3.01278

Figure S1. Birefringence of $KPSe_6$ (a), $Na_2Ge_2Se_5$ (b), and $Li_2In_2GeSe_6$ (c)