Supplementary Information *for*

Dielectric function of layered $Ga_4Se_{3+\delta}Te_{1-\delta}$ and emergent all van der Waals optical components

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Supplementary Note 1: Atomic-force microscopy of a representative vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ sample

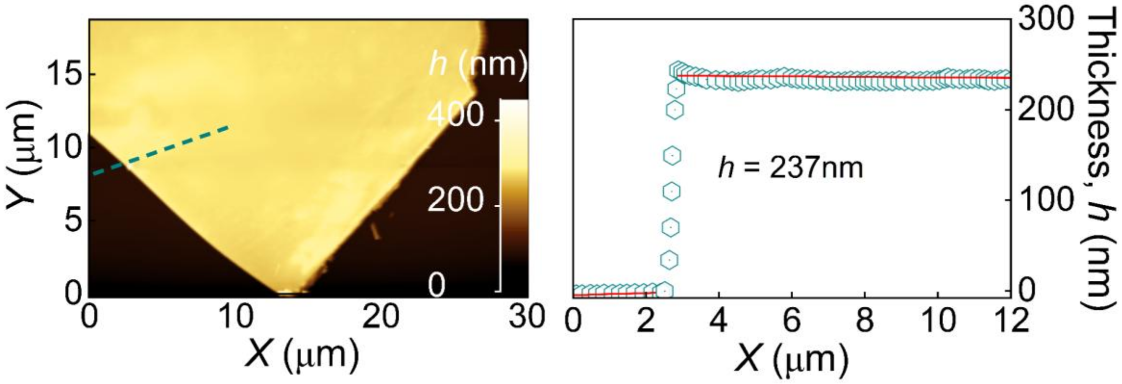

Fig. S1. Atomic force microscopy (AFM) data of a representative $Ga_4Se_{3+\delta}Te_{1-\delta}$ sample and the corresponding height profile extracted along the dashed line, indicating a thickness of 237 nm.

Supplementary Note 2: The electronic bandstructures of vdW $GaSe_xTe_{1-x}$ for various ternary compositions.

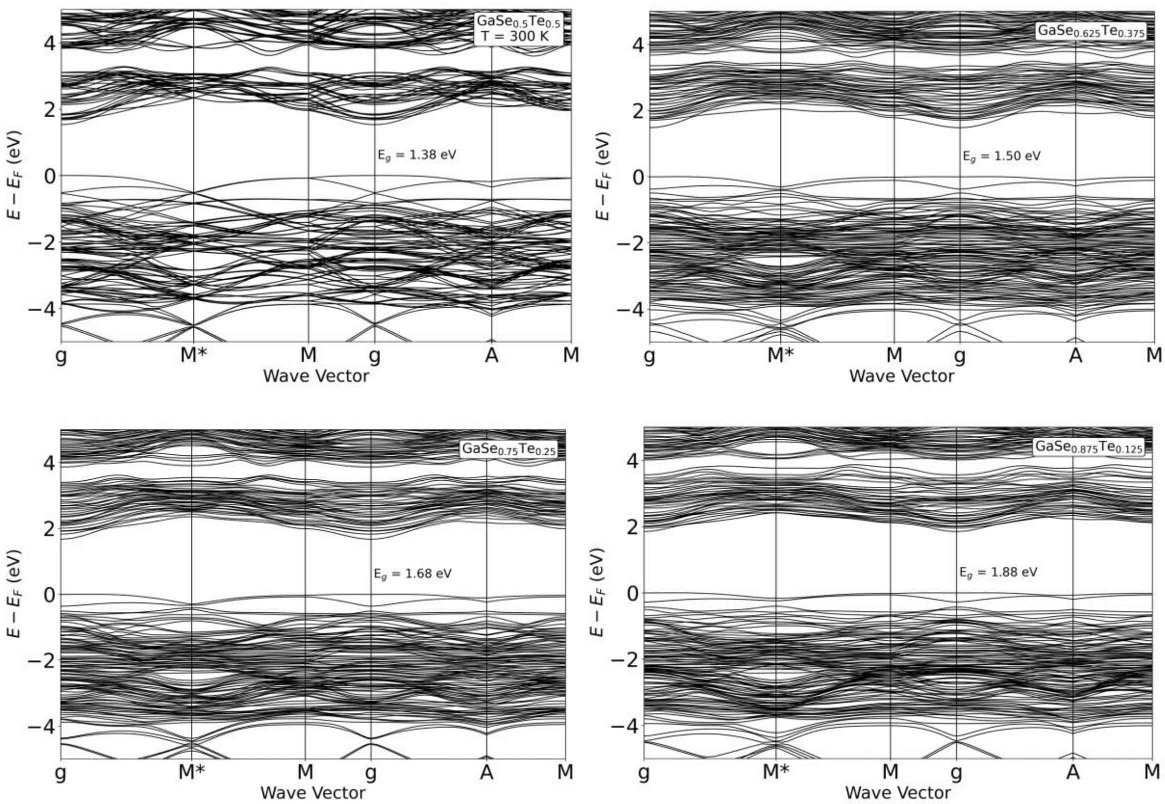

Fig. S2. DFT-calculated bandstructures of $GaSe_{0.5}Te_{0.5}$, $GaSe_{0.625}Te_{0.375}$, $GaSe_{0.75}Te_{0.25}$ and $GaSe_{0.875}Te_{0.125}$, with corresponding bandgaps of 1.38 eV, 1.50 eV, 1.68 eV and 1.88 eV (all at 300 K).

Supplementary Note 3: Ellipsometric parameters (*Ψ*, *Δ*) of representative vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ samples on different substrates.

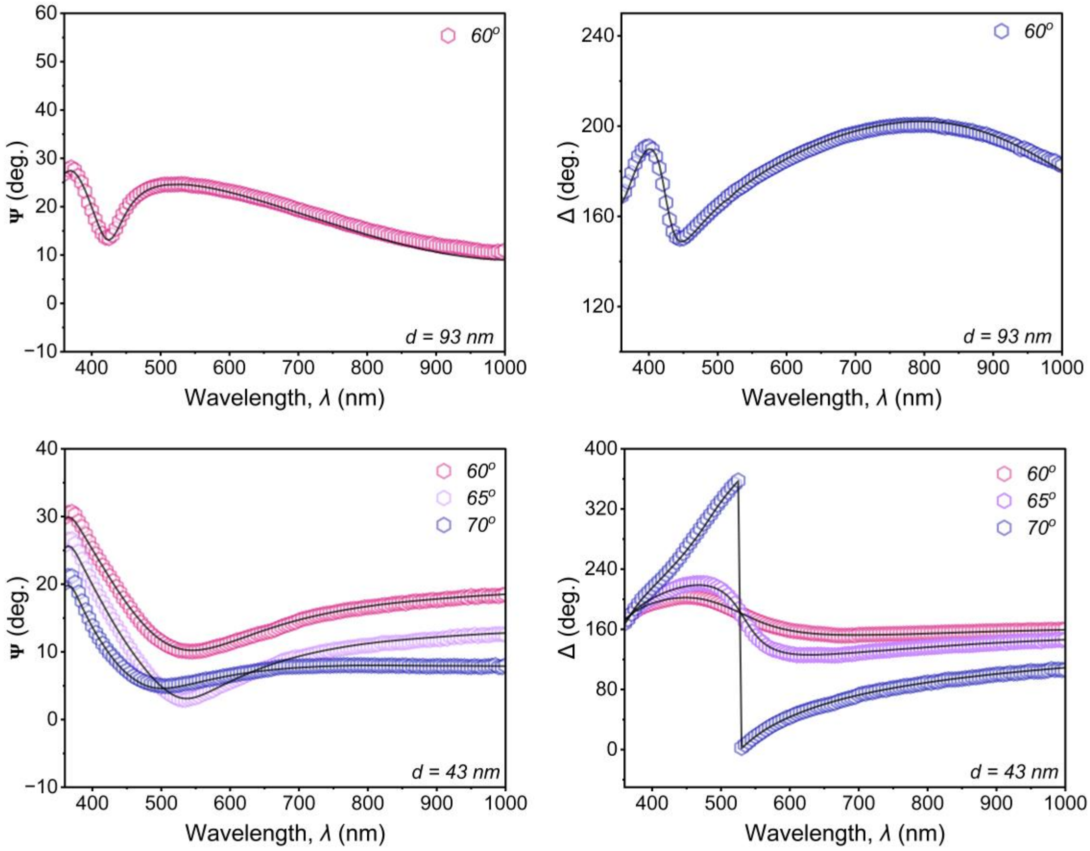

Fig. S3. Ellipsometric *Ψ* and *Δ* data acquired at different angles of incidence (AoI) for vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ samples of varying thicknesses on a Si substrate.

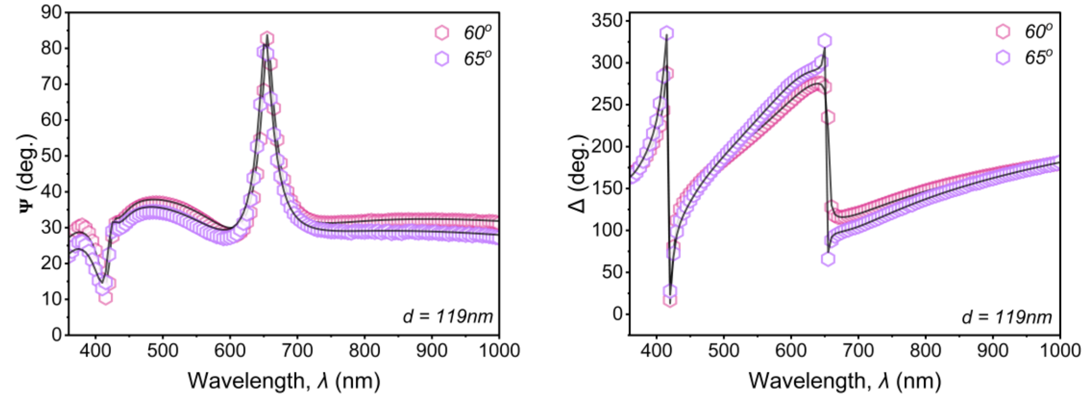

Fig. S4. Ellipsometric *Ψ* and *Δ* data acquired for a 119 nm thick vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ sample on a Si/SiO_2 (90 nm) substrate.

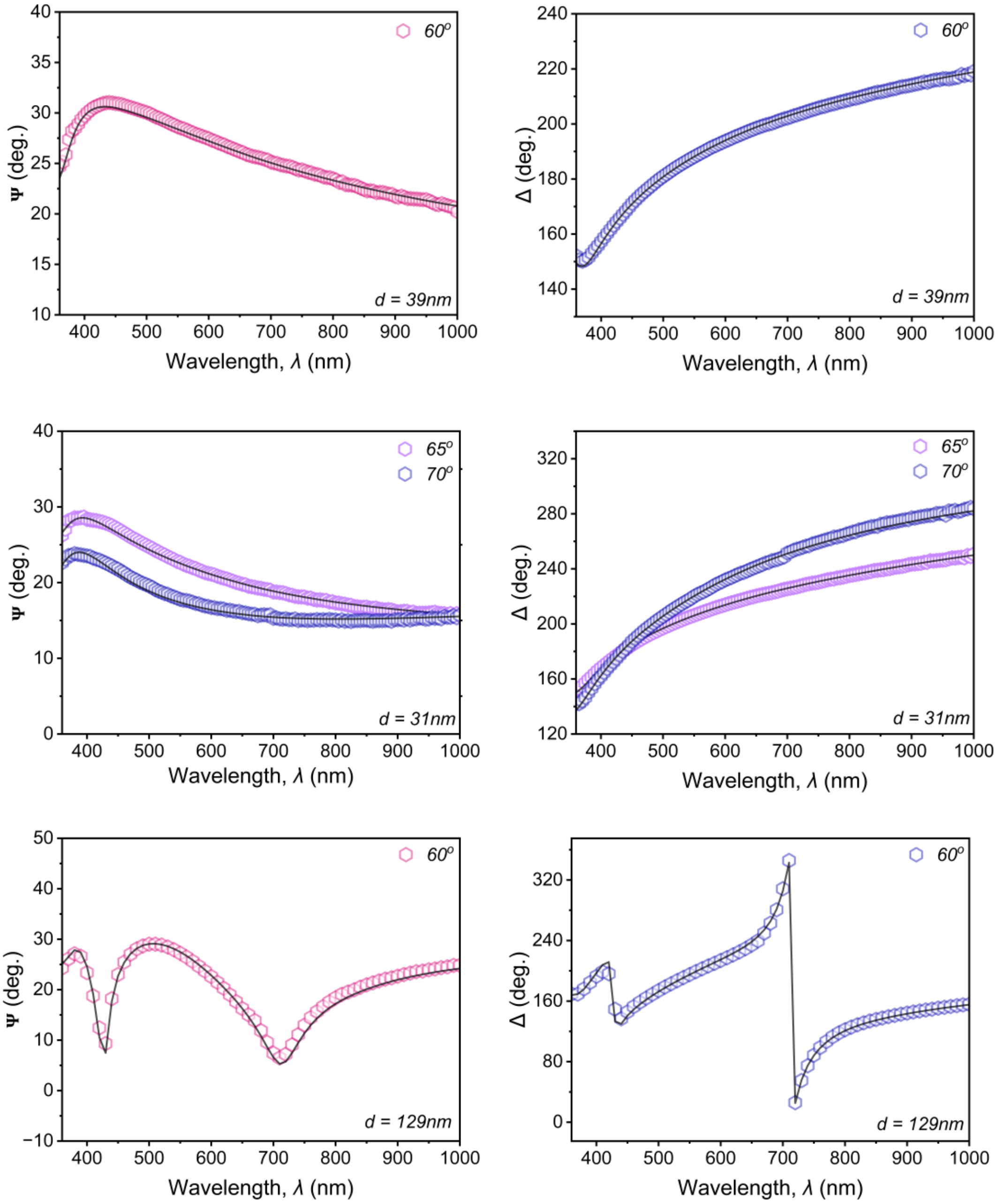

Fig. S5. Ellipsometric Ψ and Δ data acquired for vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ samples of varying thicknesses on a fused silica substrate.

Supplementary Note 4: The optical model, birefringence and tabulated constants of vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$

The in-plane and out-of-plane dielectric responses of vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ are modeled as a sum of Tauc–Lorentz oscillators, including the high-frequency contribution,

$$\varepsilon_i\,(E) = \varepsilon_{\infty,i} + \sum_n \varepsilon_{TL,i}^{(n)}\,(E), \qquad i = ab, c, \tag{1}$$

where $\varepsilon_i\ (E)$ denotes the diagonal components of the complex dielectric tensor as a function of photon energy E (in eV), n is the oscillator index.

The first term, $\varepsilon_{\infty,i}$, represents the real part of the dielectric permittivity in the high-frequency limit. The Tauc–Lorentz contribution is expressed as:

$$\varepsilon_{TL}^{(n)}(E) = \varepsilon_{TL1}^{(n)}(E) + i\varepsilon_{TL2}^{(n)}(E), \tag{2}$$

where $\varepsilon_{TL1}^{(n)}(E)$ and $\varepsilon_{TL2}^{(n)}(E)$ are the real and imaginary parts of the dielectric function associated with interband transitions, describing refraction and extinction, respectively. The imaginary part is given by

$$\varepsilon_{TL2}^{(n)}(E) = \begin{cases} 0,\ for\ E \leq E_{g,n} \\ \frac{(E-E_{g,n})^2}{E^2} \frac{A_n E_{0,n} \Gamma_n E}{(E_{0,n}^2 - E^2)^2 - \Gamma_n^2 E^2},\ for\ E > E_{g,n} \end{cases}, \tag{3}$$

where A_n is the oscillator strength, $E_{0,n}$ is the resonance energy, Γ_n is the damping parameter, and $E_{g,n}$ is the optical band gap. The real part ($\varepsilon_{TL1}(E)$) is obtained from the imaginary part *via* Kramers-Kronig integration.

Table S1. In-plane and out-of-plane dielectric function parameters extracted from the fit.

Contribution	Oscillator	Component	Parameter	Symbol	Value	Unit
Background	—	*ab*	High-frequency permittivity	$\varepsilon_{\infty,ab}$	2.444 ± 0.046	—
Background	—	*c*	High-frequency permittivity	$\varepsilon_{\infty,c}$	1.51 ± 0.17	—
TL	TL_1	*ab*	Oscillator strength	A_1	16.938 ± 2.98	eV
TL	TL_1	*ab*	Resonance energy	$E_{0,1}$	1.209 ± 0.057	eV
TL	TL_1	*ab*	Damping	Γ_1	0.299 ± 0.011	eV
TL	TL_1	*ab*	Optical band gap	$E_{g,1}$	1.869 ± 0.006	eV
TL	TL_2	*ab*	Oscillator strength	A_2	64.469 ± 1.5	eV
TL	TL_2	*ab*	Resonance energy	$E_{0,2}$	4.571 ± 0.01	eV
TL	TL_2	*ab*	Damping	Γ_2	0.554 ± 0.017	eV
TL	TL_2	*ab*	Optical band gap	$E_{g,2}$	1.879 ± 0.012	eV
TL	TL_3	*ab*	Oscillator strength	A_3	34.794 ± 0.97	eV
TL	TL_3	*ab*	Resonance energy	$E_{0,3}$	3.523 ± 0.065	eV
TL	TL_3	*ab*	Damping	Γ_3	0.591 ± 0.009	eV
TL	TL_3	*ab*	Optical band gap	$E_{g,3}$	2.664 ± 0.01	eV
TL	TL_1	*c*	Oscillator strength	A_1	85.514 ± 1.76	eV
TL	TL_1	*c*	Resonance energy	$E_{0,1}$	5.759 ± 0.024	eV
TL	TL_1	*c*	Damping	Γ_1	2.269 ± 0.058	eV
TL	TL_1	*c*	Optical band gap	$E_{g,1}$	2.957 ± 0.005	eV
TL	TL_2	*c*	Oscillator strength	A_2	18.831 ± 2.5	eV
TL	TL_2	*c*	Resonance energy	$E_{0,2}$	3.957 ± 0.038	eV
TL	TL_2	*c*	Damping	Γ_2	1.346 ± 0.015	eV
TL	TL_2	*c*	Optical band gap	$E_{g,2}$	2.444 ± 0.01	eV
TL	TL_3	*c*	Oscillator strength	A_3	20.986 ± 1.56	eV
TL	TL_3	*c*	Resonance energy	$E_{0,3}$	6.341 ± 0.062	eV
TL	TL_3	*c*	Damping	Γ_3	0.05 ± 0.007	eV
TL	TL_3	*c*	Optical band gap	$E_{g,3}$	1.582 ± 0.016	eV

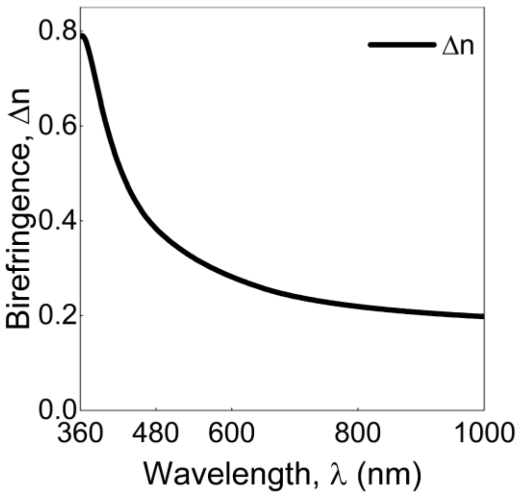

Fig. S6. Birefringence of vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ in the measured spectral range.

Table S2. Tabulated dielectric constants of vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$.

λ (nm)	**Re[ε_c]**	**Im[ε_c]**	**Re[ε_{ab}]**	**Im[ε_{ab}]**
360.0	10.2064	0.9688	15.6089	4.2741
365.0	10.0778	0.8363	15.5145	3.6412
370.0	9.9484	0.7194	15.2962	3.0438
375.0	9.8201	0.6169	14.9941	2.5226
380.0	9.6942	0.5275	14.6473	2.0882
385.0	9.5716	0.4498	14.2872	1.7354
390.0	9.4533	0.3826	13.932	1.4514
395.0	9.3394	0.3246	13.5921	1.224
400.0	9.2303	0.2749	13.2731	1.0402
405.0	9.1264	0.2325	12.9758	0.8916
410.0	9.0276	0.1966	12.701	0.7702
415.0	8.9339	0.1664	12.4475	0.6705
420.0	8.8458	0.1413	12.2132	0.5877
425.0	8.7632	0.1199	11.9966	0.5194
430.0	8.6856	0.1012	11.7964	0.4614
435.0	8.6123	0.0849	11.6107	0.4131
440.0	8.5429	0.0707	11.4393	0.3715
445.0	8.4773	0.0585	11.2791	0.3371
450.0	8.4151	0.0479	11.1309	0.3071
455.0	8.3561	0.0387	10.9926	0.2818
460.0	8.3001	0.0309	10.864	0.2604
465.0	8.2468	0.0243	10.7437	0.2418
470.0	8.1961	0.0188	10.6327	0.2259
475.0	8.1478	0.0141	10.5291	0.2108
480.0	8.102	0.0104	10.4316	0.1964
485.0	8.0585	0.0074	10.3395	0.1836
490.0	8.0169	0.0051	10.253	0.171
495.0	7.9772	0.0034	10.1708	0.1597

500.0	7.9399	0.0024	10.0932	0.1492
505.0	7.9044	0.0018	10.019	0.1388
510.0	7.8704	0.0017	9.9486	0.1296
515.0	7.8389	0.0016	9.8815	0.1205
520.0	7.8086	0.0015	9.8176	0.1122
525.0	7.7798	0.0014	9.7568	0.1039
530.0	7.7523	0.0013	9.6983	0.0963
535.0	7.7259	0.0013	9.6426	0.0893
540.0	7.7008	0.0012	9.5887	0.0824
545.0	7.6764	0.0011	9.5373	0.0762
550.0	7.6533	0.0011	9.488	0.07
555.0	7.6308	0.001	9.4409	0.0645
560.0	7.6095	0.001	9.395	0.059
565.0	7.5887	0.0009	9.3507	0.0537
570.0	7.5689	0.0009	9.3085	0.0488
575.0	7.5496	0.0008	9.268	0.0439
580.0	7.5311	0.0008	9.2282	0.0396
585.0	7.5132	0.0007	9.1904	0.0354
590.0	7.4958	0.0007	9.1534	0.0311
595.0	7.4793	0.0006	9.1174	0.0275
600.0	7.463	0.0006	9.0833	0.024
605.0	7.4474	0.0005	9.0492	0.0204
610.0	7.4323	0.0005	9.017	0.0174
615.0	7.4175	0.0005	8.9853	0.0145
620.0	7.4035	0.0004	8.955	0.0115
625.0	7.3897	0.0004	8.9249	0.0092
630.0	7.3762	0.0004	8.8961	0.0072
635.0	7.3634	0.0004	8.8678	0.0053
640.0	7.3508	0.0003	8.8406	0.0036
645.0	7.3385	0.0003	8.8138	0.0024
650.0	7.3268	0.0003	8.7883	0.0012
655.0	7.3153	0.0003	8.7634	0.0006
660.0	7.304	0.0002	8.7392	0.0
665.0	7.2931	0.0002	8.7161	0.0
670.0	7.2826	0.0002	8.6937	0.0
675.0	7.2723	0.0002	8.6725	0.0
680.0	7.2621	0.0002	8.6518	0.0
685.0	7.2525	0.0001	8.6317	0.0
690.0	7.243	0.0001	8.6121	0.0
695.0	7.2338	0.0001	8.5934	0.0
700.0	7.2246	0.0001	8.5754	0.0
705.0	7.2159	0.0001	8.5577	0.0
710.0	7.2074	0.0001	8.5405	0.0
715.0	7.199	0.0001	8.5234	0.0

720.0	7.1908	0.0001	8.5075	0.0
725.0	7.1829	0.0	8.4915	0.0
730.0	7.1752	0.0	8.4767	0.0
735.0	7.1676	0.0	8.4616	0.0
740.0	7.1602	0.0	8.447	0.0
745.0	7.1529	0.0	8.4328	0.0
750.0	7.1459	0.0	8.4192	0.0
755.0	7.1391	0.0	8.4056	0.0
760.0	7.1324	0.0	8.3926	0.0
765.0	7.1257	0.0	8.38	0.0
770.0	7.1192	0.0	8.3675	0.0
775.0	7.113	0.0	8.3554	0.0
780.0	7.1069	0.0	8.3437	0.0
785.0	7.1009	0.0	8.3318	0.0
790.0	7.095	0.0	8.3205	0.0
795.0	7.0892	0.0	8.3098	0.0
800.0	7.0835	0.0	8.2991	0.0
805.0	7.0781	0.0	8.2884	0.0
810.0	7.0727	0.0	8.2783	0.0
815.0	7.0675	0.0	8.2681	0.0
820.0	7.0623	0.0	8.2586	0.0
825.0	7.0572	0.0	8.2491	0.0
830.0	7.0521	0.0	8.2396	0.0
835.0	7.0473	0.0	8.2304	0.0
840.0	7.0426	0.0	8.2214	0.0
845.0	7.038	0.0	8.2124	0.0
850.0	7.0334	0.0	8.2039	0.0
855.0	7.0289	0.0	8.1955	0.0
860.0	7.0245	0.0	8.1871	0.0
865.0	7.0201	0.0	8.1793	0.0
870.0	7.0158	0.0	8.1714	0.0
875.0	7.0117	0.0	8.1635	0.0
880.0	7.0077	0.0	8.1561	0.0
885.0	7.0038	0.0	8.1485	0.0
890.0	6.9999	0.0	8.1412	0.0
895.0	6.996	0.0	8.134	0.0
900.0	6.9922	0.0	8.1273	0.0
905.0	6.9884	0.0	8.12	0.0
910.0	6.9847	0.0	8.1133	0.0
915.0	6.9811	0.0	8.1066	0.0
920.0	6.9777	0.0	8.1006	0.0
925.0	6.9743	0.0	8.0939	0.0
930.0	6.971	0.0	8.0878	0.0
935.0	6.9676	0.0	8.0817	0.0
940.0	6.9644	0.0	8.0756	0.0

945.0	6.9611	0.0	8.0695	0.0
950.0	6.9579	0.0	8.064	0.0
955.0	6.9548	0.0	8.0585	0.0
960.0	6.9517	0.0	8.053	0.0
965.0	6.9487	0.0	8.0474	0.0
970.0	6.9458	0.0	8.0419	0.0
975.0	6.943	0.0	8.0364	0.0
980.0	6.9401	0.0	8.0315	0.0
985.0	6.9374	0.0	8.0262	0.0
990.0	6.9346	0.0	8.0212	0.0
995.0	6.9319	0.0	8.0162	0.0
1000.0	6.9292	0.0	8.0112	0.0

Table S3. Tabulated optical constants of vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$.

λ (nm)	n_c	k_c	n_{ab}	k_{ab}
360.0	3.1983	0.1514	3.987	0.536
365.0	3.1773	0.1316	3.9655	0.4591
370.0	3.1562	0.114	3.9302	0.3872
375.0	3.1352	0.0984	3.8858	0.3246
380.0	3.1147	0.0847	3.8368	0.2721
385.0	3.0947	0.0727	3.7868	0.2291
390.0	3.0752	0.0622	3.7376	0.1942
395.0	3.0565	0.0531	3.6905	0.1658
400.0	3.0385	0.0452	3.646	0.1426
405.0	3.0212	0.0385	3.6043	0.1237
410.0	3.0048	0.0327	3.5655	0.108
415.0	2.9891	0.0278	3.5294	0.095
420.0	2.9743	0.0237	3.4958	0.0841
425.0	2.9603	0.0202	3.4644	0.075
430.0	2.9472	0.0172	3.4352	0.0672
435.0	2.9347	0.0145	3.408	0.0606
440.0	2.9229	0.0121	3.3826	0.0549
445.0	2.9116	0.01	3.3588	0.0502
450.0	2.9009	0.0082	3.3366	0.046
455.0	2.8907	0.0067	3.3158	0.0425
460.0	2.881	0.0054	3.2963	0.0395
465.0	2.8717	0.0042	3.278	0.0369
470.0	2.8629	0.0033	3.261	0.0346
475.0	2.8544	0.0025	3.245	0.0325
480.0	2.8464	0.0018	3.2299	0.0304
485.0	2.8387	0.0013	3.2156	0.0286
490.0	2.8314	0.0009	3.2021	0.0267
495.0	2.8244	0.0006	3.1893	0.025
500.0	2.8178	0.0004	3.1771	0.0235

505.0	2.8115	0.0003	3.1654	0.0219
510.0	2.8054	0.0003	3.1542	0.0205
515.0	2.7998	0.0003	3.1435	0.0192
520.0	2.7944	0.0003	3.1333	0.0179
525.0	2.7892	0.0003	3.1236	0.0166
530.0	2.7843	0.0002	3.1143	0.0155
535.0	2.7795	0.0002	3.1053	0.0144
540.0	2.775	0.0002	3.0966	0.0133
545.0	2.7706	0.0002	3.0883	0.0123
550.0	2.7665	0.0002	3.0803	0.0114
555.0	2.7624	0.0002	3.0726	0.0105
560.0	2.7585	0.0002	3.0651	0.0096
565.0	2.7548	0.0002	3.0579	0.0088
570.0	2.7512	0.0002	3.051	0.008
575.0	2.7477	0.0001	3.0444	0.0072
580.0	2.7443	0.0001	3.0378	0.0065
585.0	2.741	0.0001	3.0316	0.0058
590.0	2.7379	0.0001	3.0255	0.0051
595.0	2.7348	0.0001	3.0195	0.0046
600.0	2.7319	0.0001	3.0139	0.004
605.0	2.729	0.0001	3.0082	0.0034
610.0	2.7262	0.0001	3.0028	0.0029
615.0	2.7235	0.0001	2.9976	0.0024
620.0	2.7209	0.0001	2.9925	0.0019
625.0	2.7184	0.0001	2.9875	0.0015
630.0	2.7159	0.0001	2.9826	0.0012
635.0	2.7136	0.0001	2.9779	0.0009
640.0	2.7112	0.0001	2.9733	0.0006
645.0	2.709	0.0001	2.9688	0.0004
650.0	2.7068	0.0001	2.9645	0.0002
655.0	2.7047	0.0	2.9603	0.0001
660.0	2.7026	0.0	2.9562	0.0
665.0	2.7006	0.0	2.9523	0.0
670.0	2.6986	0.0	2.9485	0.0
675.0	2.6967	0.0	2.9449	0.0
680.0	2.6948	0.0	2.9414	0.0
685.0	2.693	0.0	2.938	0.0
690.0	2.6913	0.0	2.9346	0.0
695.0	2.6896	0.0	2.9315	0.0
700.0	2.6879	0.0	2.9284	0.0
705.0	2.6863	0.0	2.9254	0.0
710.0	2.6847	0.0	2.9224	0.0
715.0	2.6831	0.0	2.9195	0.0
720.0	2.6816	0.0	2.9168	0.0

725.0	2.6801	0.0	2.914	0.0
730.0	2.6787	0.0	2.9115	0.0
735.0	2.6772	0.0	2.9089	0.0
740.0	2.6759	0.0	2.9064	0.0
745.0	2.6745	0.0	2.9039	0.0
750.0	2.6732	0.0	2.9016	0.0
755.0	2.6719	0.0	2.8992	0.0
760.0	2.6706	0.0	2.897	0.0
765.0	2.6694	0.0	2.8948	0.0
770.0	2.6682	0.0	2.8927	0.0
775.0	2.667	0.0	2.8906	0.0
780.0	2.6659	0.0	2.8885	0.0
785.0	2.6648	0.0	2.8865	0.0
790.0	2.6636	0.0	2.8845	0.0
795.0	2.6626	0.0	2.8827	0.0
800.0	2.6615	0.0	2.8808	0.0
805.0	2.6605	0.0	2.879	0.0
810.0	2.6595	0.0	2.8772	0.0
815.0	2.6585	0.0	2.8754	0.0
820.0	2.6575	0.0	2.8738	0.0
825.0	2.6565	0.0	2.8721	0.0
830.0	2.6556	0.0	2.8705	0.0
835.0	2.6547	0.0	2.8689	0.0
840.0	2.6538	0.0	2.8673	0.0
845.0	2.6529	0.0	2.8657	0.0
850.0	2.6521	0.0	2.8643	0.0
855.0	2.6512	0.0	2.8628	0.0
860.0	2.6504	0.0	2.8613	0.0
865.0	2.6496	0.0	2.8599	0.0
870.0	2.6487	0.0	2.8586	0.0
875.0	2.648	0.0	2.8572	0.0
880.0	2.6472	0.0	2.8559	0.0
885.0	2.6465	0.0	2.8546	0.0
890.0	2.6457	0.0	2.8533	0.0
895.0	2.645	0.0	2.852	0.0
900.0	2.6443	0.0	2.8508	0.0
905.0	2.6436	0.0	2.8496	0.0
910.0	2.6429	0.0	2.8484	0.0
915.0	2.6422	0.0	2.8472	0.0
920.0	2.6415	0.0	2.8462	0.0
925.0	2.6409	0.0	2.845	0.0
930.0	2.6403	0.0	2.8439	0.0
935.0	2.6396	0.0	2.8428	0.0
940.0	2.639	0.0	2.8418	0.0
945.0	2.6384	0.0	2.8407	0.0

950.0	2.6378	0.0	2.8397	0.0
955.0	2.6372	0.0	2.8388	0.0
960.0	2.6366	0.0	2.8378	0.0
965.0	2.636	0.0	2.8368	0.0
970.0	2.6355	0.0	2.8358	0.0
975.0	2.635	0.0	2.8349	0.0
980.0	2.6344	0.0	2.834	0.0
985.0	2.6339	0.0	2.8331	0.0
990.0	2.6334	0.0	2.8322	0.0
995.0	2.6328	0.0	2.8313	0.0
1000.0	2.6323	0.0	2.8304	0.0

Supplementary Note 5: Micro-transmittance and micro-reflectance spectra of representative vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ samples (the extended dataset)

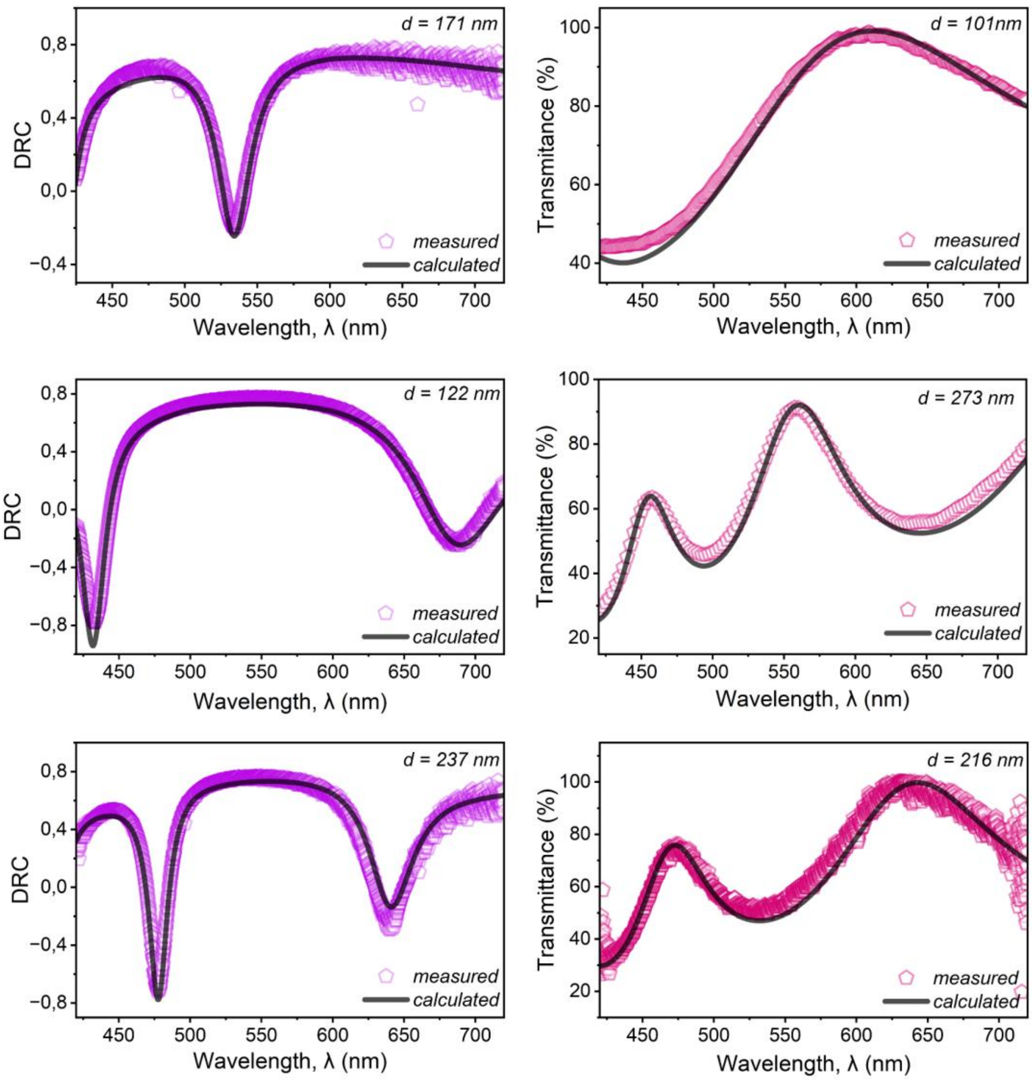

Fig. S7. Measured and calculated differential reflectance contrast (DRC) on Si/SiO_2 substrates and transmittance on fused silica substrates for vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ samples of varying thicknesses, demonstrating good agreement between experimental data and calculated spectra.

The transmittance and reflectance spectra of the vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ structure were calculated using the generalized transfer-matrix method (TMM) [1]. Within this framework, the electromagnetic wave propagation inside each layer is described by the propagation matrix, while the boundary conditions at the interfaces are accounted for by the interface (boundary-condition) matrices. These matrices are defined as follows:

$$P_i = \begin{bmatrix} e^{-i\frac{2\pi}{\lambda}q_{i1}d_i} & 0 & 0 & 0 \\ 0 & e^{-i\frac{2\pi}{\lambda}q_{i2}d_i} & 0 & 0 \\ 0 & 0 & e^{-i\frac{2\pi}{\lambda}q_{i3}d_i} & 0 \\ 0 & 0 & 0 & e^{-i\frac{2\pi}{\lambda}q_{i4}d_i} \end{bmatrix}, \quad (4)$$

$$A_i = \begin{bmatrix} \gamma_{i11} & \gamma_{i21} & \gamma_{i31} & \gamma_{i41} \\ \gamma_{i12} & \gamma_{i22} & \gamma_{i32} & \gamma_{i42} \\ \frac{q_{i1}\gamma_{i11}-\xi\gamma_{i13}}{\mu_i} & \frac{q_{i2}\gamma_{i21}-\xi\gamma_{i23}}{\mu_i} & \frac{q_{i3}\gamma_{i31}-\xi\gamma_{i33}}{\mu_i} & \frac{q_{i4}\gamma_{i41}-\xi\gamma_{i43}}{\mu_i} \\ \frac{q_{i1}\gamma_{i12}}{\mu_i} & \frac{q_{i2}\gamma_{i22}}{\mu_i} & \frac{q_{i3}\gamma_{i32}}{\mu_i} & \frac{q_{i4}\gamma_{i42}}{\mu_i} \end{bmatrix}, \quad (5)$$

where d_i is the thickness of layer i, λ is the free-space wavelength, and q_{ij} (j = 1, …, 4) are the eigenvalues associated with the four electromagnetic eigenmodes of the layer, γ_{ijk} are the components of the eigenmode matrix derived from the 4 × 4 electromagnetic eigenproblem and describe the coupling of the field components across interfaces. The parameter ξ denotes the conserved in-plane wave-vector component, and μ_i is the magnetic permeability of layer i. For the case of normal incidence considered here, $\xi = 0$. Since vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ is non-magnetic, the magnetic permeability is set to $\mu_i = 1$.

After defining the constituent matrices, the total transfer matrix of the system, Γ_{tot} (*Eq.* 6), is constructed by incorporating the boundary conditions at both the superstrate and substrate. The matrix is then transformed via the normalization $\Gamma \to \Gamma^*$, after which the polarization-dependent transmittance and reflectance coefficients are evaluated using *Eq*. (7) and *Eq*. (8).

$$\Gamma_{tot} = A_{sup}^{-1} \prod_{i=1}^{N} A_i P_i A_i^{-1} A_{sub}, \quad (6)$$

In hexagonal vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$, the in-plane dielectric response is isotropic; therefore, at normal incidence $t_{pp} = t_{ss} = t_{tot}$ and $r_{pp} = r_{ss} = r_{tot}$.

$$t_{pp} = \frac{\Gamma_{33}^*}{\Gamma_{11}^*\Gamma_{33}^* - \Gamma_{13}^*\Gamma_{31}^*}, \qquad t_{ss} = \frac{\Gamma_{11}^*}{\Gamma_{11}^*\Gamma_{33}^* - \Gamma_{13}^*\Gamma_{31}^*}, \quad (7)$$

$$r_{pp} = \frac{\Gamma_{21}^*\Gamma_{33}^* - \Gamma_{23}^*\Gamma_{31}^*}{\Gamma_{11}^*\Gamma_{33}^* - \Gamma_{13}^*\Gamma_{31}^*}, \qquad r_{ss} = \frac{\Gamma_{11}^*\Gamma_{43}^* - \Gamma_{41}^*\Gamma_{13}^*}{\Gamma_{11}^*\Gamma_{33}^* - \Gamma_{13}^*\Gamma_{31}^*}. \quad (8)$$

Differential reflectance contrast and transmittance were calculated by converting the total reflectance using the substrate reflectance as a reference (*Eq.* 9) and normalizing the total transmittance to an air–glass reference spectrum (*Eq.* 10), respectively.

$$T = \frac{T_{tot}}{T_{ref}}, \quad (9)$$

$$R_{DRC} = \frac{R_{tot} - R_{ref}}{R_{tot} + R_{ref}}. \quad (10)$$

Here, $T = |t_{tot}|^2$ and $R_{tot} = |r_{tot}|^2$ denote the total transmittance and reflectance spectra, respectively, while $T_{ref} = |t_{ref}|^2$, $R_{ref} = |r_{ref}|^2$ denote the corresponding reference spectra for transmittance and reflectance.

Supplementary Note 6: Optical constants of vdW *h*BN obtained by the spectroscopic micro-ellipsometry

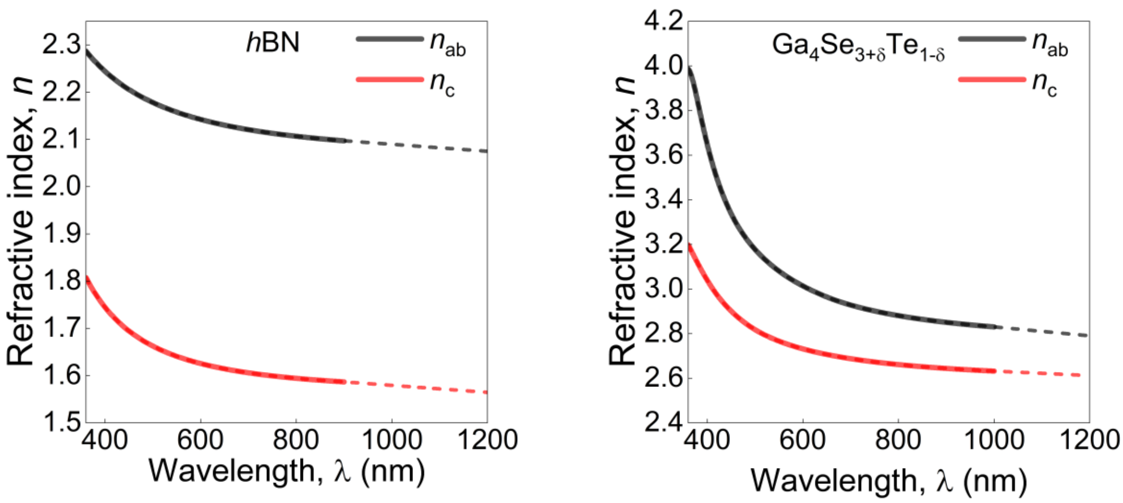

Fig. S8. The optical constants of vdW *h*BN (left) and vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ (right) used for unpolarized beam splitter computations. The interpolated range for each case is shown by dashed lines.

Supplementary Note 7: The performance characteristics of all-vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$/*h*BN unpolarized beam splitters

Table S4. Optimized all-vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$/*h*BN layer pairs for an unpolarized 50:50 plate beam splitter (AOI = 45°).

Overall performance: T_{un} = 50 ± 1 %, R_{un} = 50 ± 1 %
Total thickness: 642.3 nm
Number of layers: 7
Substrate: UV fused silica
Stack order: starts with low-refractive-index layer (*h*BN)
Spectral range: 800–1030 nm

Pair index	*h*BN (L) thickness (nm)	$Ga_4Se_{3+\delta}Te_{1-\delta}$ (H) thickness (nm)
1	131.19	63.59
2	126.74	80.93
3	119.99	60.42

Cap layer	54.91	*N.A.*

Table S5. Optimized all-vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$/*h*BN layer pairs for an unpolarized 30:70 plate beam splitter (AOI = 45°).

Overall performance: T_{un} = 70 ± 1 %, R_{un} = 30 ± 1 %
Total thickness: 638.9 nm
Number of layers: 7
Substrate: UV fused silica
Stack order: starts with low-refractive-index layer (*h*BN)
Spectral range: 800–1030 nm

Pair index	***h*BN (L) thickness (nm)**	**$Ga_4Se_{3+\delta}Te_{1-\delta}$ (H) thickness (nm)**
1	134.41	78.08
2	110.38	57.03
3	99.45	56.59
Cap layer	102.91	*N.A.*

Table S6. Optimized all-vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$/*h*BN layer pairs for an unpolarized 10:90 plate beam splitter (AOI = 45°).

Overall performance: T_{un} = 90 ± 1 %, R_{un} = 10 ± 1 %
Total thickness: 751.8 nm
Number of layers: 7
Substrate: UV fused silica
Stack order: starts with high-refractive-index layer ($Ga_4Se_{3+\delta}Te_{1-\delta}$)
Spectral range: 800–1030 nm

Pair index	***h*BN (L) thickness (nm)**	**$Ga_4Se_{3+\delta}Te_{1-\delta}$ (H) thickness (nm)**
1	120.72	84.1
2	112	16.28
3	76.89	249.05
Cap layer	92.76	*N.A.*

Supplementary Note 8: The optical constants of vdW $GaSe_xTe_{1-x}$ for various ternary compositions.

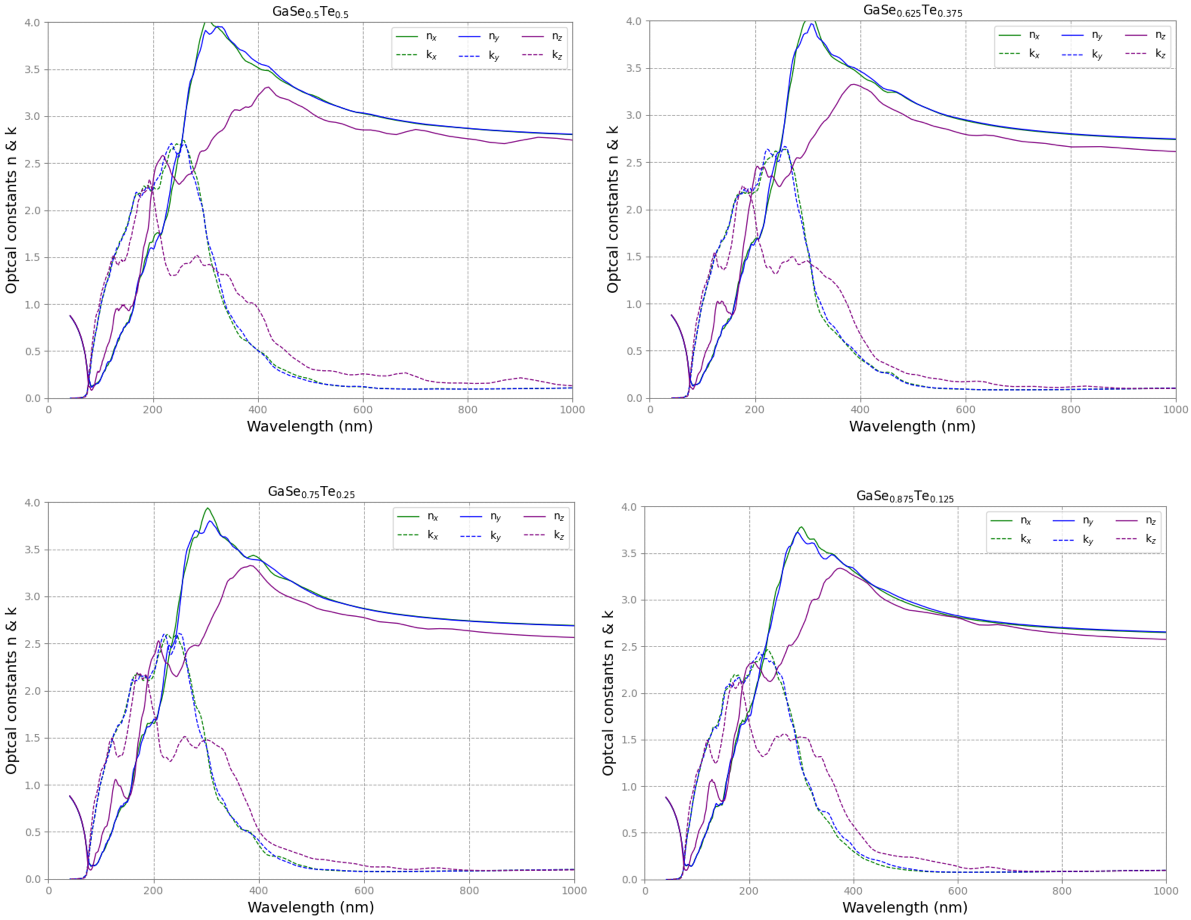

Fig. S9. DFT-calculated refractive indices and extinction coefficients of vdW $GaSe_{0.5}Te_{0.5}$, $GaSe_{0.625}Te_{0.375}$, $GaSe_{0.75}Te_{0.25}$ and $GaSe_{0.875}Te_{0.125}$ (all at 300 K).

Supplementary Note 9: Mueller-matrix ellipsometry of representative vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ samples

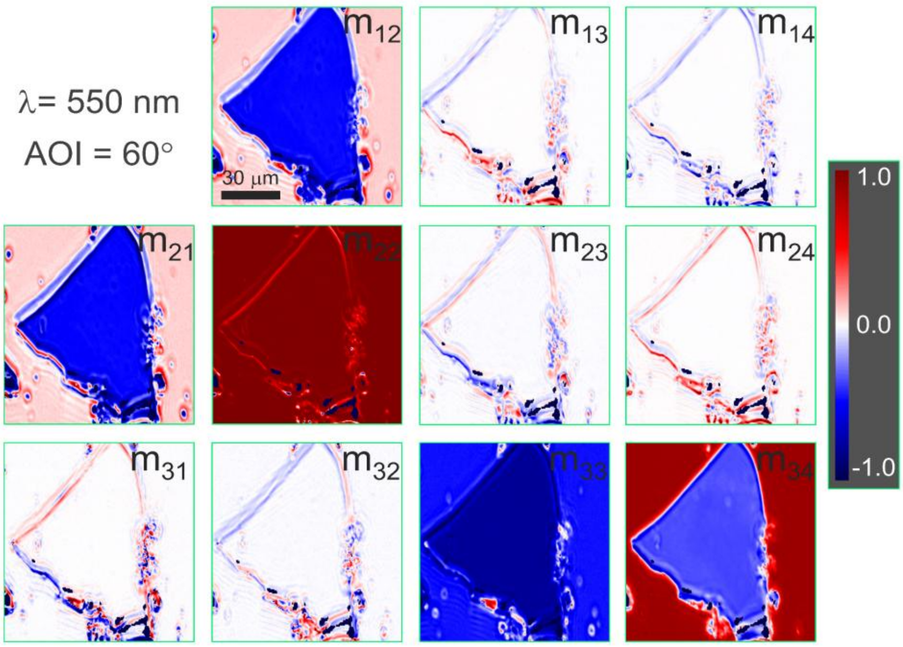

Fig. S10. Micro-maps of the normalized Mueller-matrix (MM) elements of vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ sample on a Si/SiO_2 substrate, measured at an angle of incidence (AOI) of 60° and λ = 550 nm.

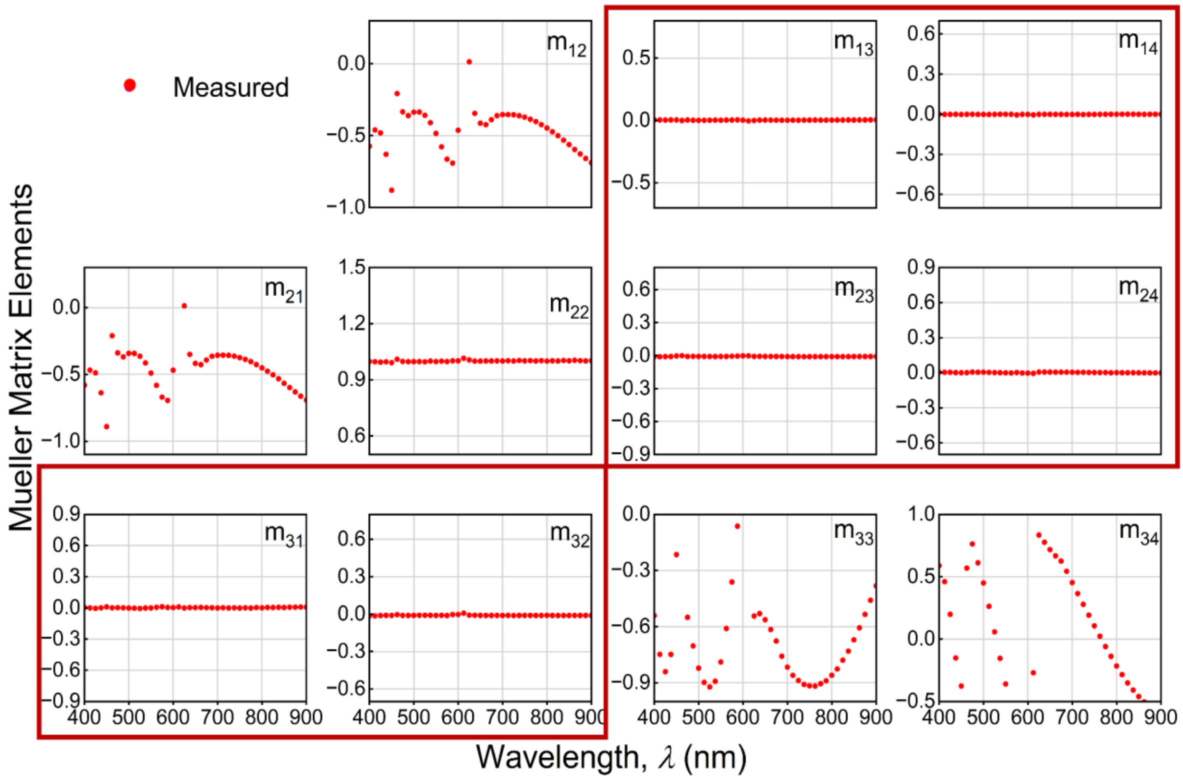

Fig. S11. Spectral dependence of the normalized 3 × 4 MM elements of vdW $Ga_4Se_{3+\delta}Te_{1-\delta}$ sample on a Si/SiO_2 substrate at an AOI of 60°, obtained from region-of-interest (ROI) evaluation of the corresponding MM micro-maps. The off-diagonal Mueller-matrix elements highlighted by the red rectangle remain close to zero across the measured spectral range.

References

1. Passler, N. C. & Paarmann, A. Generalized 4 × 4 matrix formalism for light propagation in anisotropic stratified media: study of surface phonon polaritons in polar dielectric heterostructures. *J. Opt. Soc. Am. B.* **34**, 2128–2139 (2017).