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Electronic Supplementary Information

Monotonously Alloying-Driven Band Edge Emission in Two-Dimensional Hexagonal $GaSe_{1-x}Te_x$ Semiconductors for Visible to Near-Infrared Photodetection

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Table S1. *Solid Te composition extracted from XRD-2Theta scans and EDX, and lattice parameters of $GaSe_{1-x}Te_x$*

Nominal Te composition (x^0)	**Solid Te composition (x) extracted from**			**Lattice parameters (Å)**	
	XRD_*a*	**XRD_*c*/4**	**EDX**	**a**	**c**
0	0	0	0	3.755	15.884
0.2	0.073	0.095	0.069	3.776	15.976
0.33	0.134	0.174	0.142	3.795	16.062
0.5	0.247	0.290	0.229	3.833	16.199
0.65	0.292	0.353	0.251	3.843	16.258
0.75	0.328	0.401	0.352	3.854	16.309
0.85	0.370	0.448	0.395	3.867	16.361

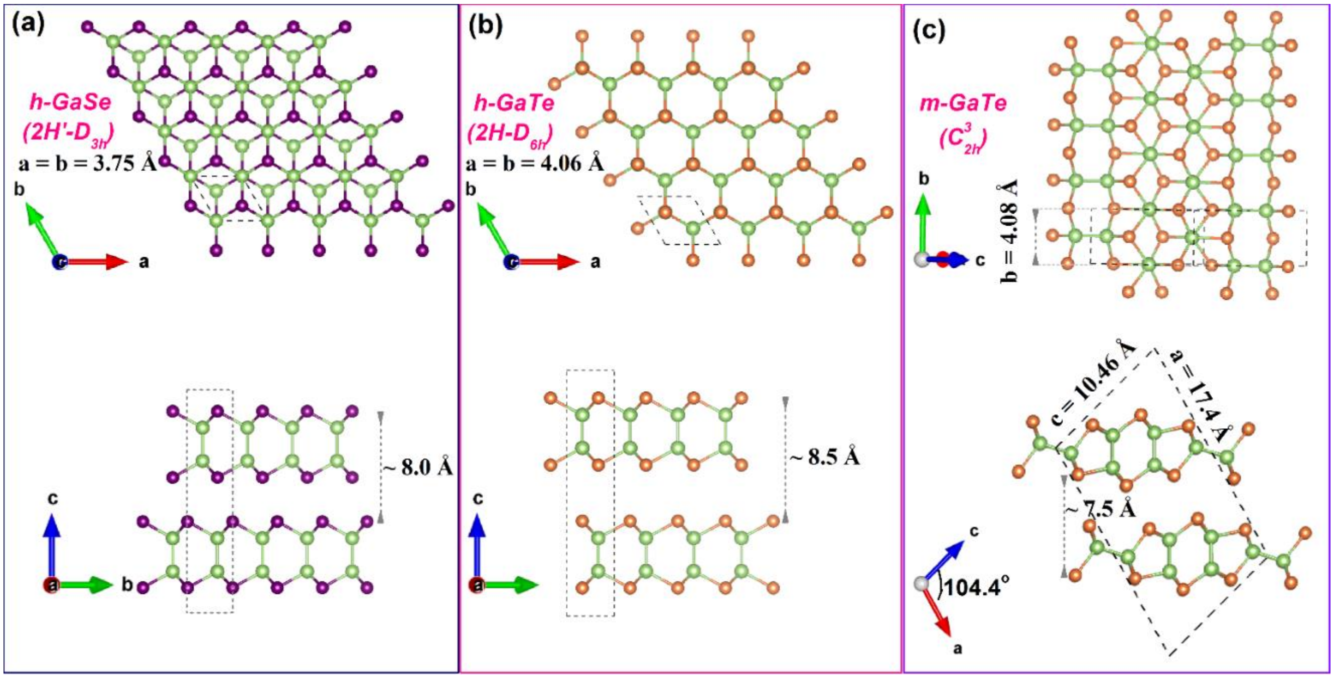

Figure S1. *Visualization of the crystal structures of 2D GaSe and GaTe materials by VESTA software.*[1]

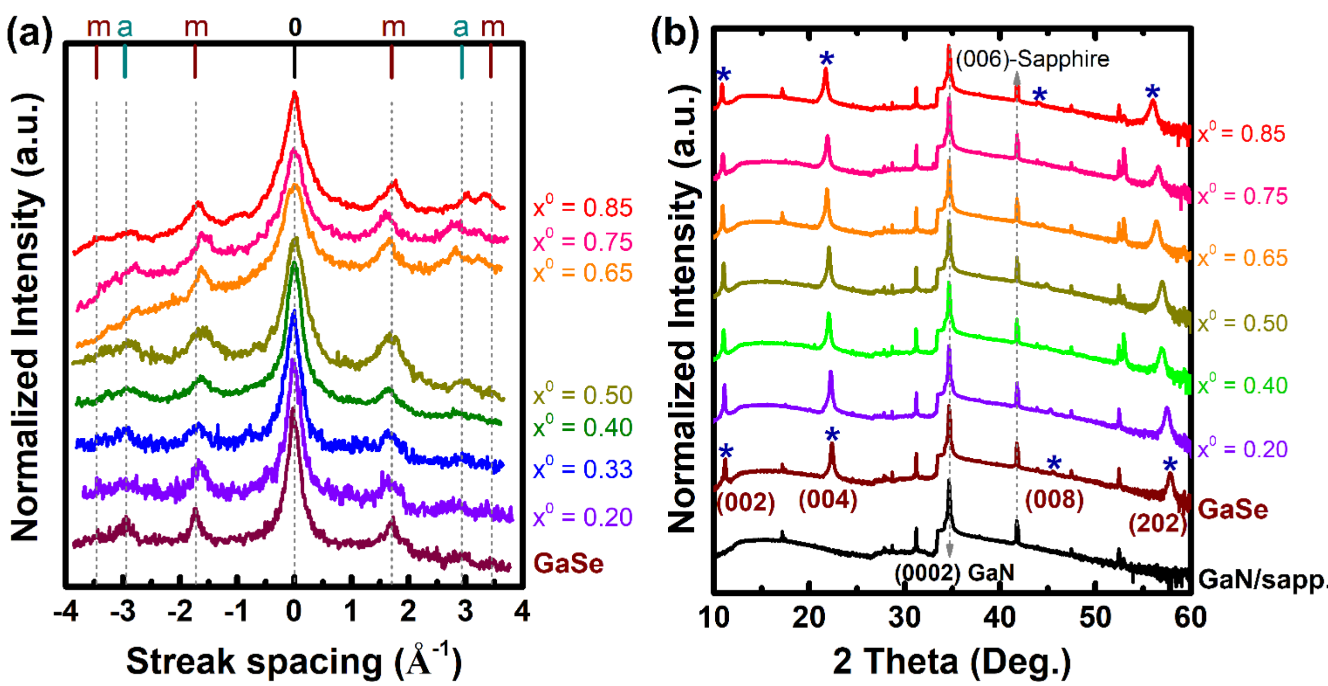

Figure S2. *(a) RHEED intensity profiles and (b) 2θ-XRD scans of the upward GaSeTe alloys versus nominal Te composition.*

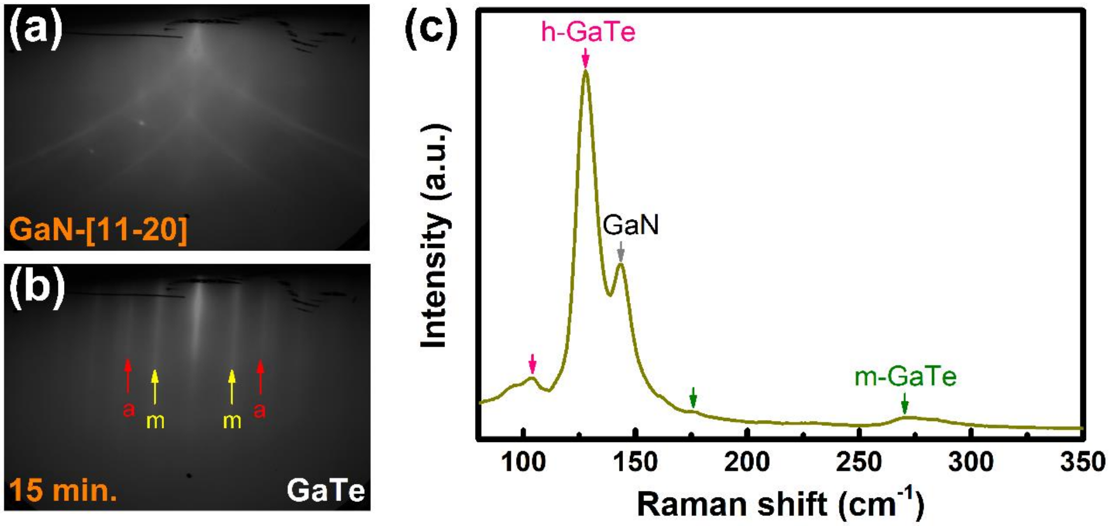

Figure S3. *RHEED pattern of (a) GaN/sapphire platform and (b) 15-min GaTe grown layer on GaN/sapphire at 525 °C, (c) 532-nm Raman spectrum of the 2hr-GaTe grown layer on GaN/Sapphire platform. (Reproduced from S.H. Huynh et al., ACS Appl. Nano Mater., 2021, 4, 8913-8921)*

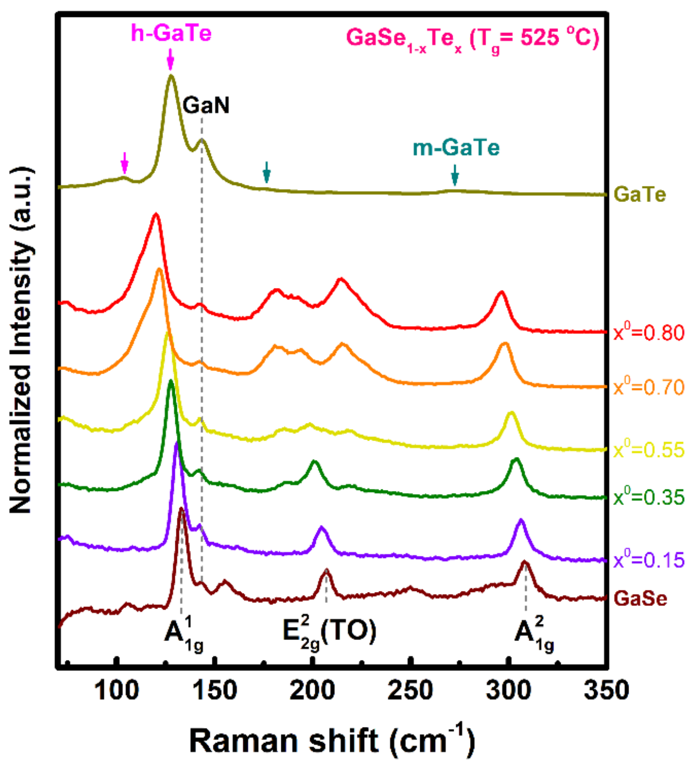

Figure S4. *532-nm Raman spectra of the downward GaSeTe alloys measured at room temperature versus nominal Te composition.*

Photoluminescence of GaSe grown on GaN/sapphire template at T_s = 560 °C

Temperature-dependent PL measurements of the GaSe (*x* = 0) sample were carried out to determine the optical bandedge emission energy of GaSe grown on GaN/sapphire as shown in Fig. S5a. The 10K-PL spectrum of GaSe is composed of two typical emission bands positioned at ~ 1.774 eV (peak I), and ~ 1.856 eV (peak II). Peak I was attributed to the emission of the exciton originated from the SDD growth as reported previously,[2] while peak II located at the higher energy is believed to be the emission of the exciton initiated from the LBL growth.[3] We noted a redshift as well as deterioration of all PL peaks as increasing the temperature, where the free excitonic emission (peak II) shifted an energy of ~90 meV (from ~1.856 eV to ~1.765 eV) when the temperature was raised from 10 K to 120 K (Fig. S5b). The temperature evolution of peak II shown in the inset of Fig. S5b is excellently fitted to the Varshni's empirical equation.[4] As a result, the bandgap energy of the GaSe sample grown on GaN/sapphire template in this study is estimated to be ~1.86 eV.

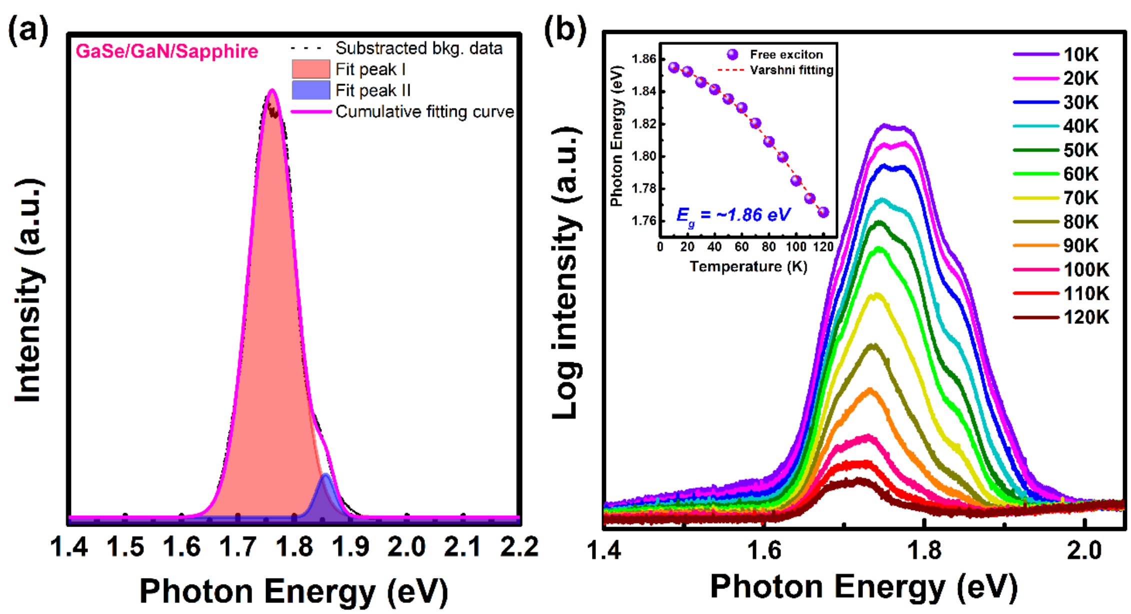

Figure S5. *(a) Low-temperature PL emission of GaSe grown at T_s=560 °C. (b) Temperature-dependent PL spectra of GaSe grown at 560 °C. Inset of (b) is Varshni fitting curve of the corresponding excitonic emission.*

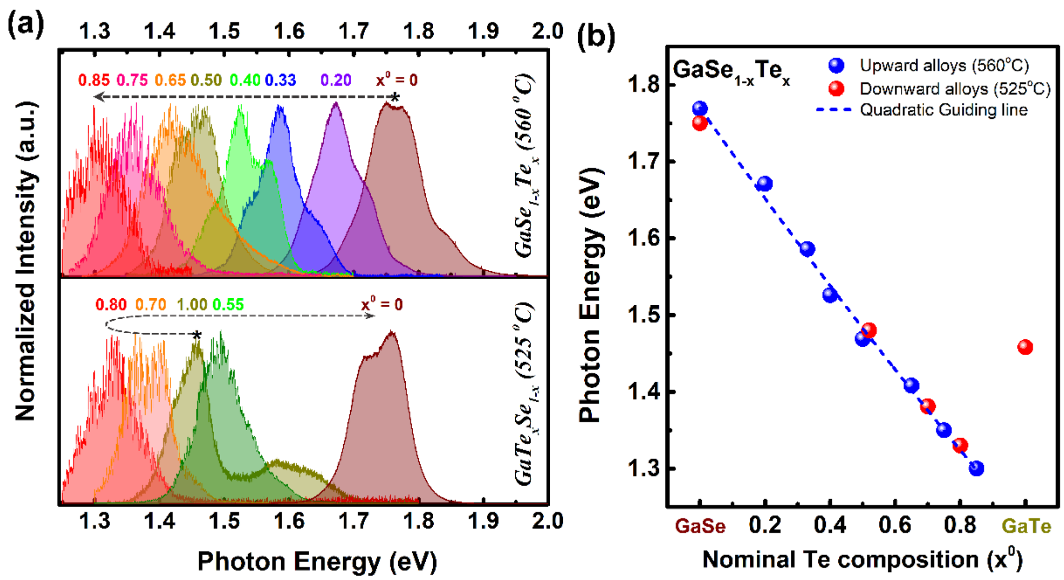

Figure S6. *Evolution of the central PL band emission versus nominal Te concentration of both upward and downward alloy series.*

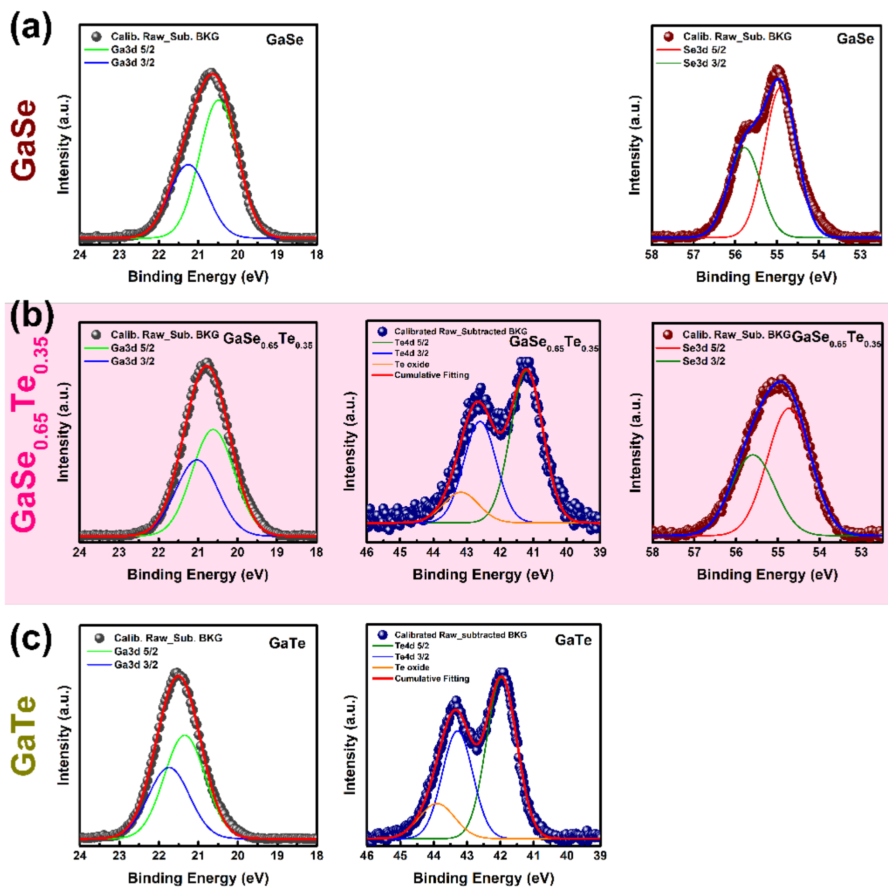

Figure S7. *Synchrotron XPS spectra of Ga 3d, Te 4d, and Se 3d core levels of (a) GaSe, (b) $GaSe_{0.65}Te_{0.35}$, and (c) GaTe.*

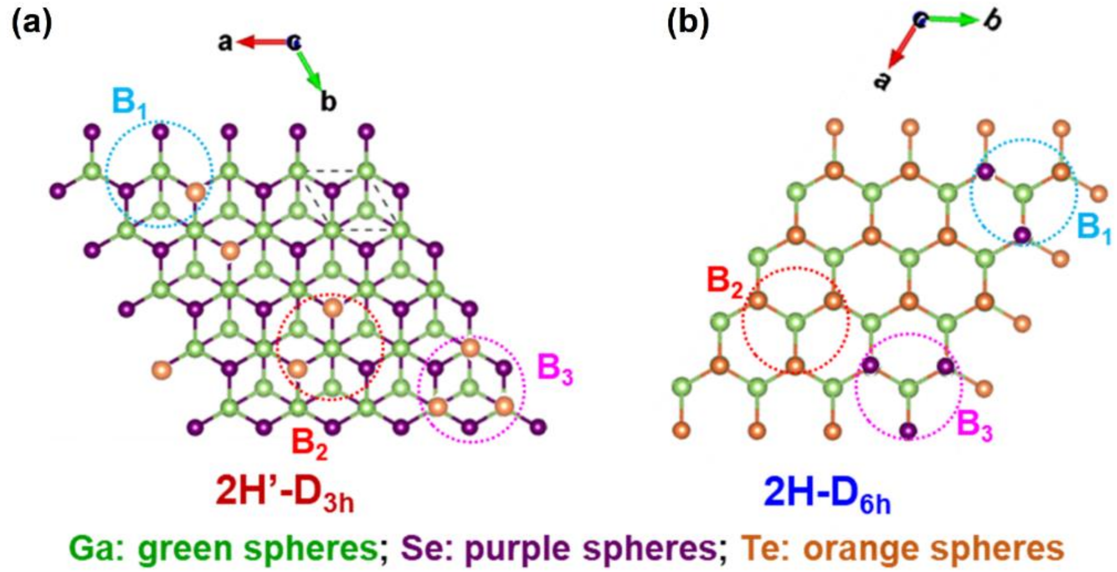

Figure S8. *Visualized exemplification of atomic arrangement observed from the top view of (a) 2H'-D_{3h} $GaSe_{1-x}Te_x$ crystal, and (b) 2H-D_{6h} $GaSe_{1-x}Te_x$ crystal.*

References

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