## Supporting Information for

## Reduction of carrier density and enhancement of the bulk Rashba spin-orbit coupling strength in Bi<sub>2</sub>Te<sub>3</sub>/GeTe superlattices

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**Fig. S1.** The cross-section STEM images of the symmetric  $([1|3]_{25}, [2|6]_{13}, [3|9]_8, [4|12]_6$ , and  $[6|18]_4$ ) and the asymmetric  $([2|18]_6)$  [BT|GT] SLs., where  $[x|y]_z$  notation represents an SL composed of *x* (u.c.) of the layer BT and *y* (u.c.) of the GT layer with *z* representing the repetitions of the layers. Fast Fourier transformation of the images is shown as an inset together with each STEM image.



**Fig. S2.** The *c*-lattice constant of GT (0003) (a) and BT (0003) (b), which are calculated from the XRD peaks in Fig. 1c of the main text.



**Fig. S3.** (a)-(e) XRD reciprocal lattice map around Si (224) of the symmetric [BT|GT] SLs. (f) The changes of  $Q_x$  of asymmetric (0,1,-1,n) reflections according to increasing period.



**Fig. S4.** AFM topography of the GeTe 2 nm template layer on Si (111) substrate (a),  $[2|6]_6$  (b),  $[4|12]_3$  (c), and  $[6|18]_2$  (d) [BT|GT] SLs.



**Fig. S5.** The sheet resistance  $(R_{sh})$  as a function of the temperature (T) of [BT|GT] SLs.



Fig. S6. The temperature-dependent carrier density (n) of [BT|GT] SLs. The sign of n means the carrier types; the positive (the negative) means hole (electron). n was calculated by the Hall measurement with the external magnetic field of 4 T.



**Fig. S7.** The Hall resistance  $(R_{xy})$  as a function of the magnetic field (*H*) of [BT|GT] SLs at T=1.8 K.



**Fig. S8.** The magneto-resistance (*MR*) of  $[t_{BT}|t_{GT}]$  SLs ( $t_{GT}=1, 2, 3, 4, \text{ and } 6$ ) as  $t_{BT} = 1$  (a), 2 (b), 3 (c), 4 (d), and 6 (e) at T=1.8 K. The *MR*<sub>ord</sub> is proportional to the square of mobility ( $\mu$ ) and magnetic field (*H*) (*MR*<sub>ord</sub> ~ ( $\mu$ *H*)<sup>2</sup>) because it is related to the scattering induced by the electron cyclone motion in a vertical magnetic field. Therefore, *MR*<sub>ord</sub> can be expressed as  $MR(H) = k(H/\rho_0)^2$  through Kohler's rule, and the constant *k* is a temperature invariant constant assuming that the carrier density is independent of temperature [1]. *k* has been extracted from *MR*(*H*) of relatively high temperature (100 K) at which *MR*<sub>ord</sub> dominates *MR*(*H*), and *MR*<sub>ord</sub> at 1.8 K has been reproduced with this *k* (Graphical representation of this process is shown in Fig. S10).



Fig. S9. Temperature dependence of the MR vs. H curve of an SL ([BT<sub>1</sub>|GT<sub>3</sub>]<sub>25</sub>) as a representative sample



**Fig. S10.** Process of extraction of  $MR_{WAL}$ . (a) MR vs.  $H / \rho_0$  curve at 100 K (black open circles), which can be fully expressed by the classical MR ( $MR_{ord}$ ) with the form of  $k(H / \rho_0)^2$  (red line). With k extracted from (a), the  $MR_{ord}$  at 1.8 K can be reproduced (blue line in (b)). And, the  $MR_{WAL}$  (red line in (b)) can be obtained by subtracting the  $MR_{ord}$  from raw MR (black open circles in (b)).



**Fig. S11.** The fitting parameters of  $B_{\phi}$  (a) and  $B_{so}$  (b) as a function of carrier mobility.

## Reference

[1] Y. Nakazawa, M. Uchida, S. Nishihaya, M. Ohno, S. Sato, and M. Kawasaki, Physical Review B **103** (2021).