Intermixing during epitaxial growth of van der Waals bonded nominal $GeTe/Sb_2Te_3$ superlattices

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SUPPLEMENTARY INFORMATION

Experimental details:

All the substrates are cleaned and the surfaces are prepared using the methods described in Boschker et al.[5] and Wang et al.[6] The growth of CSLs is described by J. Momand *et al.* [9] A nominal superlattice structure was built with alternating layers of GeTe and Sb_2Te_3. During the growth, substrate (227.5 °C) and cell temperatures ($T(Ge)_{base}$ = 1120°C and $T(Ge)_{tip}$ = 1140°C for the Ge cell, $T(Sb)_{base}$ = 450°C and $T(Sb)_{tip}$ = 600°C for the Sb cell, $T(Te)_{base}$ = 340°C and $T(Te)_{tip}$ = 476°C for the Te cell) were kept constant. The shutter of the Te cell remained open while the shutters of the Ge and Sb cells were alternatively opened and closed depending on the desired layer. The used flux ratio Ge/Sb/Te was ~2/2/5. After the deposition of the CSL, the sample was cooled down to room temperature, and *in-situ* capped with Si_3N_4 by sputtering to prevent oxidation of the last GeTe sublayer.

The diffractometer used for XRD characterization of the samples is a Panalytical X' Pert PRO MRD system with Ge (220) hybrid monochromator, employing a $CuK\alpha_1$ (λ = 1.540598 Å) X-ray radiation.

The Raman spectra were excited with the 632.8 nm line of a He-Ne laser and the scattered light was analyzed using a spectrometer equipped with an LN_2-cooled charge-coupled device detector. Raman spectra were obtained in backscattering z(x,xy)-z geometry, meaning that incident light is polarized in the x direction, while the polarization of the backscattered light is not selected.