# Controlled Growth of 3D Topological Insulator $BiSb(Te_{1-y}Se_y)_3$ Nanocrystals by Chemical Vapor Transport

Nour Abdelrahman [abcd]*, Titouan Charvin [a], Samuel Froeschke [a], Romain Giraud [ae], Joseph Dufouleur [a], Alexey Popov [a], Sandra Schiemenz [a], Daniel Wolf [a], Bernd Büchner [af], Michael Mertig [bg], and Silke Hampel [a]*

[a]Leibniz Institute for Solid State and Materials Research Dresden, Helmholtzstraße 20, 01069 Dresden, Germany. E-mail: n.abdelrahman@ifw-dresden.de; Tel: +4935146591116.
[b]Institute of Physical Chemistry, Technische Universität Dresden, 01062 Dresden, Germany
[c]Department of Chemistry, Faculty of Applied Sciences, Palestine Technical University-Kadoorie, Jaffa Street, Tulkarm, P.O. Box 7, Palestine.
[d] Helmholtz-Zentrum Dresden-Rossendorf e.V. D-01328 Dresden, Germany.
[e]Université Grenoble Alpes, CNRS, CEA, Grenoble-INP, Spintec, 38054 Grenoble, France
[f]Institute of Solid State and Materials Physics, Technische Universität Dresden, Dresden, 01062, Germany
[g]Kurt-Schwabe-Institut für Mess- und Sensortechnik Meinsberg e.V., Kurt-Schwabe-Straße 4, Waldheim 04736, Germany

*Contact E-mail: n.abdelrahman@ifw-dresden.de. (Nour Abdelrahman)

# Supplemntary data

**SI. 1.** Thermodynamic data for the condensed species used in the simulation

| Formula | State | T | H(T) in $J\cdot mol^{-1}$ | S(T) in $J\cdot mol^{-1}\cdot K^{-1}$ | a | b | c | d | e |
|---|---|---|---|---|---|---|---|---|---|
| **Bi** | S | 298 | 0 | 56.735 | 11.849 | 3.05E-02 | 4.105E+05 | | |
| **Bi (l)** | L | 544 | 9271.870 | 71.997 | 19.016 | 1.04E-02 | 2.07E+06 | -3.979E-06 | |
| **$Bi_2Te$** | S | 298 | -27614.4 | 175.728 | 80.7512 | | | | |
| **BiTe** | S | 298 | -27196 | 110.876 | 53.9736 | | | | |
| **BiSe** | S | 298 | -53136.8 | 102.508 | 53.5552 | | | | |
| **$Bi_2O_3$** | S | 298 | -578010 | 149.81 | 96.780 | 0.04633747 | 249291.3 | -6.626E-09 | |
| **$Bi_2O_3$ (beta)** | S | 1003 | -563676.81 | 149.257021 | 149.7 | | | | |
| **$Bi_2O_3$ (l)** | L | 1098 | -590808.97 | 94.464161 | 202 | | | | |
| **Sb** | S | 298 | 0 | 45.522 | 30.472 | -0.015384568 | -199995.2 | 1.794E-05 | |
| **Sb (l)** | L | 904 | 17530.47 | 62.707929 | 31.38 | 0.06611 | | | |
| **$Sb_2O_3$** | S | 298 | -720305 | 110.449 | 134.7 | | -802800 | | -419.2 |
| **$Sb_2O_3$ (beta)** | S | 298 | -703623.52 | 134.62452 | 92.05 | 0.06611 | | | |
| **$Sb_2O_3$ (l)** | L | 298 | -662675.9 | 172.405918 | 156.9 | 0.1454735 | 2454752.8 | | |
| **$Sb_2O_4$** | S | 298 | -907509.6 | 127.1936 | 47.019792 | | -2182000 | | 151.4 |
| **$Sb_2O_5$** | S | 298 | -971901 | 125.102 | 133.4 | 0.034088 | | | |
| **$SbO_2$** | S | 298 | -453754.8 | 63.597 | 47.279 | 0.022676 | | -0.000012865 | |
| **Se** | S | 298 | 0 | 42.2584 | 11.7535 | 0.047225 | 85927.9 | | |
| **Se (l)** | L | 210 | 3661.58 | 48.349019 | 134.7248 | | | | |
| **$Se_2O_5$** | S | 298 | -413379.15 | 158.992 | 110.7 | | | | -726 |
| **$SeO_2$** | S | 298 | -225350 | 66.693 | 76.5672 | 0.07223 | | | |
| **$SeO_3$** | S | 298 | -170288.79 | 96.232 | 35.6687 | -0.031668718 | -310029 | 3.14425E-05 | |
| **Te** | S | 298 | 0 | 49.221 | 126.031802 | -0.44388672 | -1655855.3 | 0.000565245 | |
| **Te(l)** | L | 298 | 10925.22 | 62.246118 | 12.75016 | 0.26083056 | 1748912 | | |
| **$H_2TeO_4$** | S | 298 | -715000 | 99.759999 | 65.18672 | 0.01456032 | -502080 | | |
| **$TeO_2$** | S | 298 | -323423 | 74.057 | 65.1872 | 0.01456032 | -502080 | | |
| **$TeO_2$ (l)** | L | 298 | -298181.04 | 98.930644 | 167.36 | | | | |
| **$Bi_2Te_3$ (l)** | L | 870 | 24661.72 | 364.409986 | 112.88432 | 0.0531368 | | | |
| **$Sb_2Te_3$ (l)** | L | 298 | 34506.38 | 347.269875 | 188.28 | | | | |
| **$Bi_2Se_3$ (l)** | L | 995 | -102830.14 | 237.782448 | 118.74192 | 0.02092 | | | |
| **$Sb_2Se_3$ (l)** | L | 298 | -78770.27 | 266.642811 | 118.74192 | 0.02092 | | | |
| **$Sb_2Se_3$** | S | 298 | -127612 | 212.129 | 86.818 | 0.0489528 | | | |
| **$Bi_2Se_3$** | S | 298 | -139954.8 | 239.7432 | 112.88432 | 0.0531368 | | | |
| **$Sb_2Te_3$** | S | 298 | -56484 | 246.438 | 107.989 | 0.055229 | | | |
| **$Bi_2Te_3$** | S | 298 | -78659 | 261.082 | | | | | |

*Cp function d

The units of th

# Supplemntary data

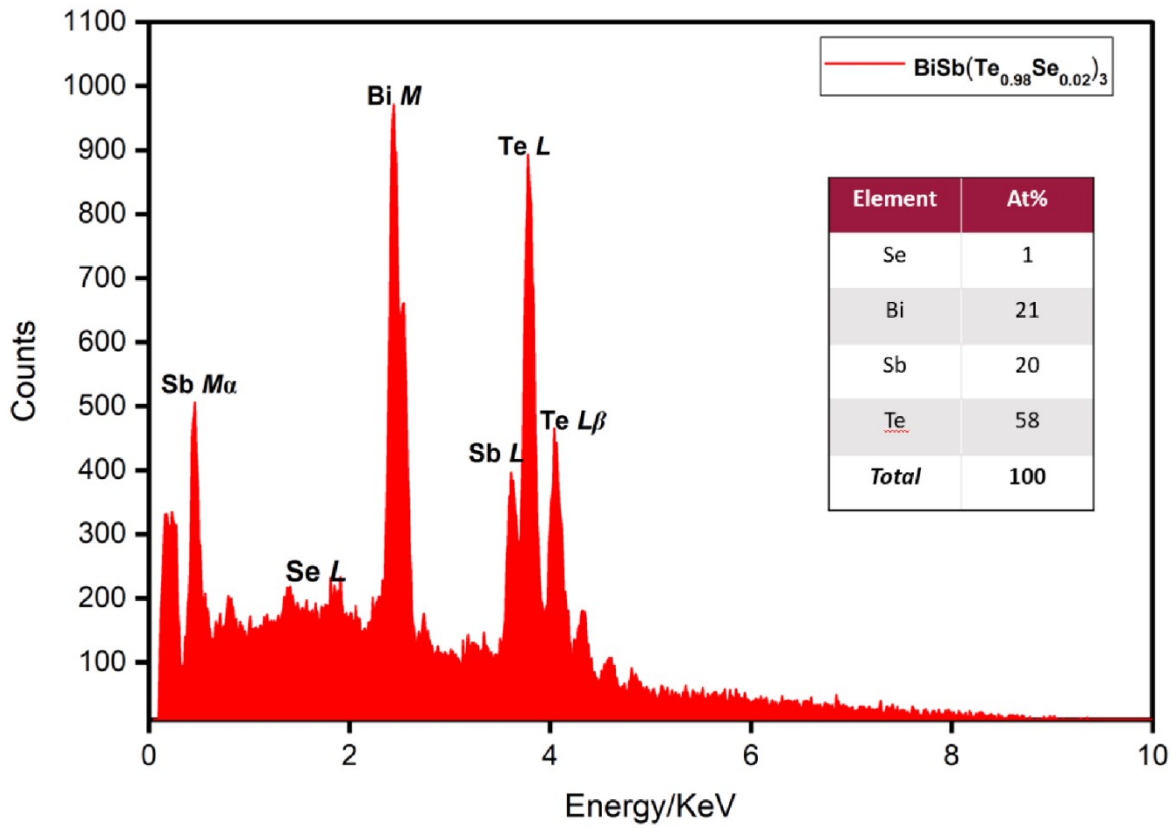
**SI. 2.** Thermodynamic data for the gaseous species used in the simulations

| Formula | T | H(T) inJ·mol$^{-1}$ | S(T) inJ·mol$^{-1}$·K$^{-1}$ | a | b | c | d | e | f | g |
|---|---|---|---|---|---|---|---|---|---|---|
| **Ar** | 298.0 | 0.0 | 154.845 | 2.079E+01 | | | | | | |
| **H** | 298.0 | 217999.0 | 114.716 | 2.079E+01 | | | | | | |
| **$H_2$** | 298.0 | 0.000 | 130.680 | 1.983E+01 | 3.078E-03 | -2.952E+05 | 1.430E-06 | 1.949E+02 | | |
| **$H_2O$** | 298.0 | -241834.0 | 188.834 | 2.578E+01 | 1.495E-02 | -2.800E+04 | -5.524E-07 | | 1.107E+03 | |
| **O** | 298.0 | 249173 | 161.058 | 1.996E+01 | 2.639E-04 | 5.865E+04 | -3.695E-08 | 2.095E+01 | | |
| **$O_2$** | 298.0 | 0 | 205.147 | 2.692E+01 | 1.698E-02 | 2.293E+05 | -6.766E-06 | -7.916E+01 | | |
| **Bi** | 298 | 210873.6 | 187.009 | 2.079E+01 | | | | | | |
| **$Bi_2$** | 298.0 | 220078.4 | 273.743 | 3.679E+01 | 7.615E-04 | | | | | |
| **BiSe** | 298 | 166523.2 | 269.559044 | 3.670E+01 | 7.950E-04 | | | | | |
| **BiTe** | 298.0 | 190372.0 | 273.006 | 3.688E+01 | 9.120E-04 | | | | | |
| **BiO** | 298.0 | 113595.6 | 246.547 | 3.611E+01 | 8.368E-04 | -3.305E+05 | | | | |
| **Sb** | 298.0 | 264554.320 | 180.264 | 2.079E+01 | | | | | | |
| **$Sb_2$** | 500.0 | 235879.23 | 254.648 | 3.724E+01 | | | | | | |
| **$Sb_4$** | 500.0 | 208775.96 | 353.228 | 8.264E+01 | | | | | | |
| **SbSe** | 298 | 212129 | 255.877364 | 3.635E+01 | 1.172E-03 | | | | | |
| **SbTe** | 298.0 | 202087.2 | 262.488 | 3.630E+01 | 1.239E-03 | 3.820E+03 | -1.948E-08 | | | |
| **SbOH** | 298 | -133523.5 | 133.812444 | 9.018E+01 | | | | | | |
| **$Sb(OH)_2$** | 298 | -493667.4 | 156.091444 | 134.8493 | | | | | | |
| **$Sb_4O_6$** | 500.0 | -125565.39 | 429.455 | 2.234E+02 | | | | | | |
| **$SbH_3$** | 298.0 | 145101 | 233.075 | 5.050E+01 | 1.870E+00 | -1.318E+06 | | | | |
| **SbO** | 298.0 | 89297 | 238.346 | 5.463E+01 | -5.428E-03 | | | -3.655E+02 | | |
| **Se** | 298 | 235350 | 176.715 | 21.464 | 0.001506 | -92000 | | | | |
| **$Se_2$** | 298 | 136699 | 243.618 | 44.601 | -0.002657 | -251000 | | | | |
| **$Se_3$** | 298 | 173518 | 315.038 | 58.145 | 0.003038 | -222000 | | | | |
| **$Se_4$** | 298 | 180631 | 379.204 | 83.082 | 0.000033 | -251000 | | | | |

# Supplemntary data

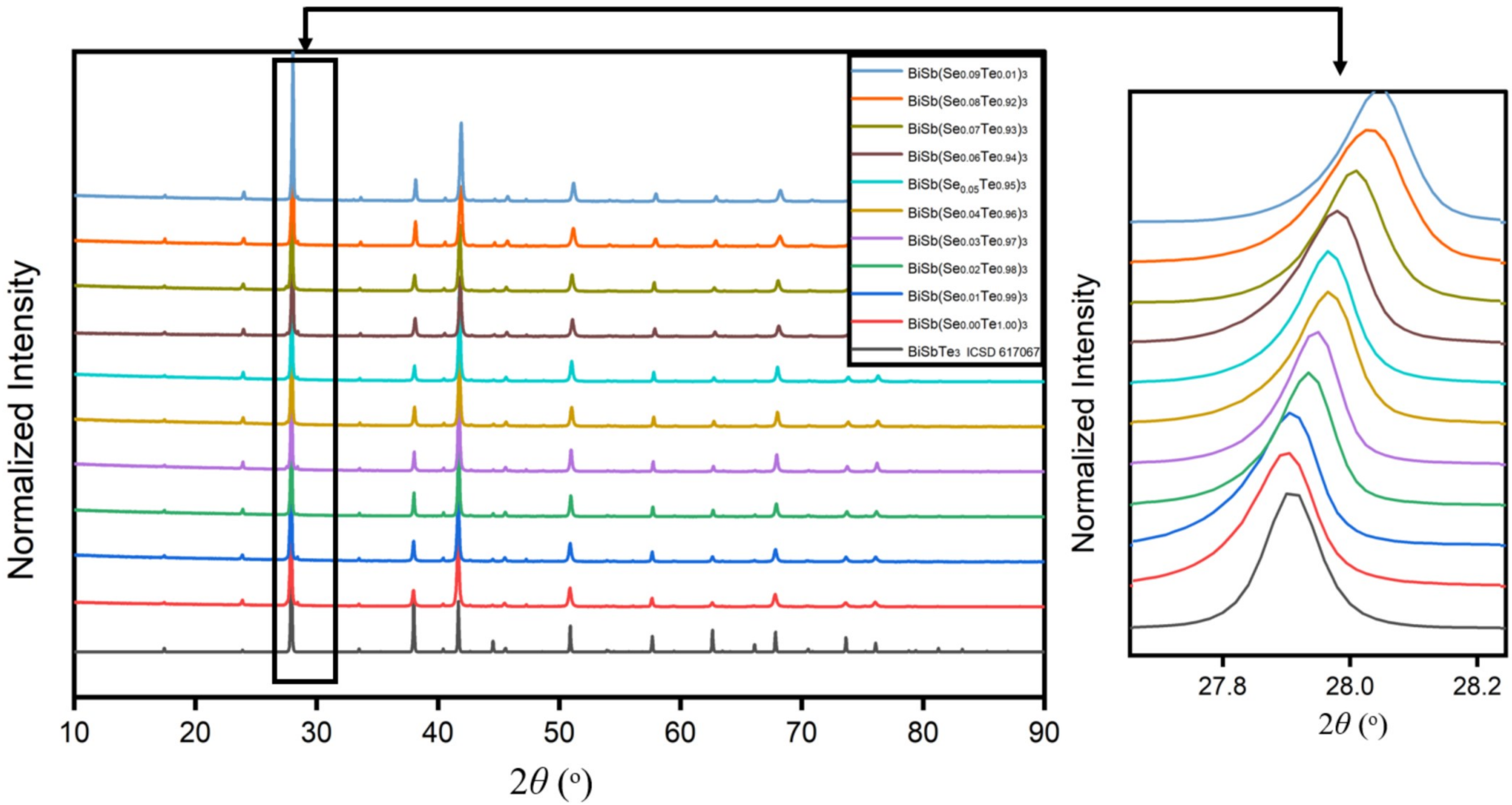
**SI. 2.** Thermodynamic data for the gaseous species used in the simulations – continued

| Formula | T | H(T) inJ·mol$^{-1}$ | S(T) inJ·mol$^{-1}$·K$^{-1}$ | a | b | c | d | e | f | g |
|---|---|---|---|---|---|---|---|---|---|---|
| $Se_5$ | 298 | 135444 | 385.359 | 107.926 | 0.000088 | -590000 | | | | |
| $Se_6$ | 298 | 132515 | 433.613 | 132.905 | 0.000067 | -594000 | | | | |
| $Se_7$ | 298 | 141302 | 486.474 | 157.762 | 0.000113 | -828200 | | | | |
| $Se_8$ | 298 | 152176 | 531.159 | 182.74 | 0.000092 | -494000 | | | | |
| $SeO$ | 298 | 62341 | 233.994 | 3.494E+01 | 1.506E-03 | -3.680E+05 | | | | |
| $SeO_2$ | 298 | -107842 | 264.998 | 5.284E+01 | 3.088E-03 | -9.200E+05 | | | | |
| $H_2Se$ | 298 | 29288 | 218.933 | 3.176E+01 | 1.464E-02 | -1.297E+05 | | | | |
| $SeO_2$ | 298 | -107842 | 264.998 | 5.284E+01 | 3.088E-03 | | | | | |
| $H_2Se$ | 298 | 29288 | 218.933 | 3.176E+01 | 1.464E-02 | | | | | |
| $TeSe$ | 298 | 151464 | 256.882177 | 3.348E+01 | 3.743E-02 | -3.451E+05 | -4.053E-05 | | | 1.305E-08 |
| $Te$ | 298 | 209451 | 182.707 | 2.079E+01 | | | | | | |
| $Te_2$ | 298.0 | 162063 | 258.944 | 3.570E+01 | -1.507E-03 | 1.635E-05 | | | | |
| $Te_3$ | 298.0 | 203223.04 | 335.942 | 5.802E+01 | 4.022E-04 | -1.428E+05 | -3.159E-07 | | | 8.504E-11 |
| $Te_4$ | 298.0 | 217321.89 | 379.093 | 8.282E+01 | 7.007E-04 | -2.418E+05 | -5.506E-07 | | | 1.483E-10 |
| $Te_5$ | 298.0 | 220026.76 | 462.011 | 1.079E+02 | 3.929E-04 | -2.155E+05 | -3.096E-07 | | | 8.367E-11 |
| $Te_6$ | 298.0 | 226560.81 | 491.477 | 1.329E+02 | 3.529E-04 | -2.504E+05 | -2.798E-07 | | | 7.592E-11 |
| $Te_7$ | 298.0 | 254235.86 | 558.072 | 1.578E+02 | 4.434E-04 | -3.050E+05 | -3.503E-07 | | | 9.496E-11 |
| $Te_2O_2$ | 298.0 | -108784 | 327.298 | 8.211E+01 | 5.577E-03 | -1.180E+06 | | | | |
| $TeO$ | 298 | 744750 | 240.689 | 3.531E+01 | 1.339E-03 | -3.47E+05 | | | | |
| $TeO_2$ | 298 | -59413 | 274.998244 | 5.477E+01 | 2.414E-03 | -1.183E+06 | -5.336E-06 | | | |
| $Te(OH)_2$ | 298 | -374860 | 228.114 | 6.587E+01 | 4.215E-02 | -6.585E+04 | -2.668E-06 | 1.157E+02 | | |
| $TeOH$ | 298 | -102890 | 187.869 | 4.250E+01 | 3.212E-02 | -3.293E+04 | | 5.785E+01 | | |
| $H_2Te$ | 298.0 | 99579 | 228.974 | 3.548E+01 | 1.205E-02 | -3.096E+05 | | | | |
| $H_2TeO_3$ | 298.0 | 430000 | 322.586 | 7.933E+01 | 5.064E-02 | 4.881E+04 | -8.719E-06 | 7.612E+01 | | |




**SI. 3.** EDX spectrum (15 kV) of bulk BiSb ($Te_{0.98}Se_{0.02}$)$_3$ used as starting material for nanocrystals synthesis.

Supplemntary data



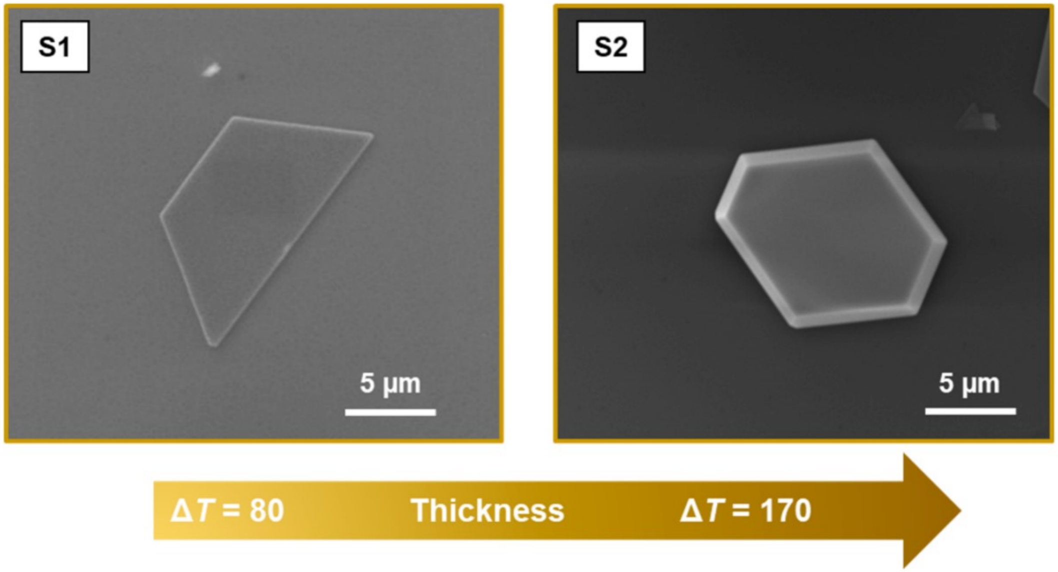

**SI. 4.** pXRD patterns of bulk $BiSb(Te_{1-y}Se_y)_3$ ($y$ = 0, 0.01, 0.02,…,0.09) and the reference pattern of $BiSbTe_3$, on the right side, main peak enlargement patterns (to show the corresponding peak shift).

## Supplemntary data

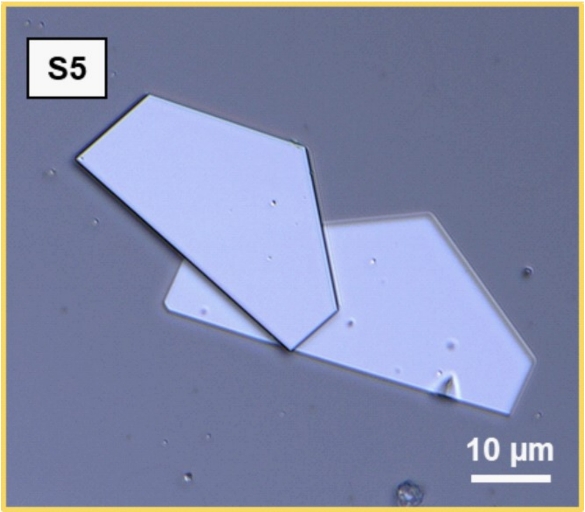
**SI. 5.**. Summary of the growth conditions for different samples of $BiSb(Te_{1-y}Se_{y})_3$ nanocrystals.

| Title | Substrate | $T_1$/ °C | $T_2$/ °C | $\Delta T$/ K | $D$ / cm | $t$ / h |
|---|---|---|---|---|---|---|
| S1 | $SiO_2$ | 500 | 420 | 80 | 12 | 8 |
| S2 | $SiO_2$ | 500 | 330 | 170 | 12 | 8 |
| S3 | $SiO_2$ | 500 | 330 | 170 | 12 | 4 |
| S4 | $SiO_2$ | 560 | 390 | 170 | 12 | 4 |
| S5 | $SiO_2$ | 630 | 460 | 170 | 12 | 4 |
| S6 | $SiO_2$ | 500 | 380 | 120 | 8 | 12 |
| S7 | $SiO_2$ | 500 | 380 | 120 | 16 | 12 |
| S8 | $SiO_2$ | 560 | 390 | 170 | 12 | 4 |
| S9 | $SiO_2$ | 560 | 390 | 170 | 12 | 8 |
| S10 | $SiO_2$ | 560 | 390 | 170 | 12 | 12 |
| S11 | $SiO_2$ | 560 | 390 | 170 | 12 | 16 |
| S12 | $Al_2O_3$ | 500 | 420 | 80 | 12 | 4 |
| S13 | Si | 500 | 380 | 120 | 12 | 8 |
| S14 | $BaF_2$ | 500 | 420 | 80 | 12 | 4 |

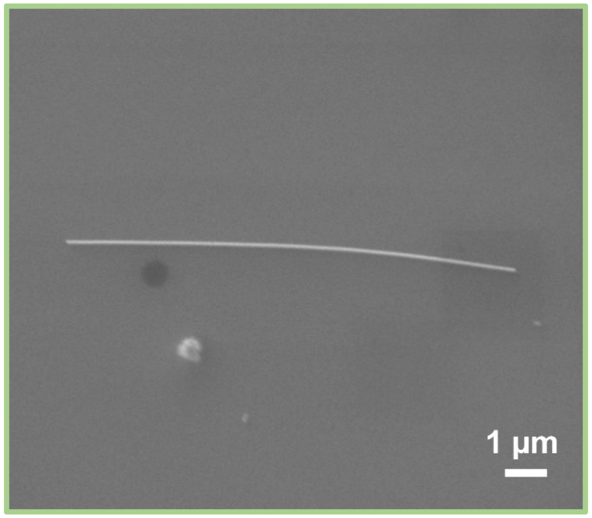
$T_1$ indicates the temperature of source zone. $T_2$ indicates temperature of sink zone (substrate). $\Delta T$ is the temperature gradient. $D$ is the distance between source and sink zones. $t$ is the growth duration.



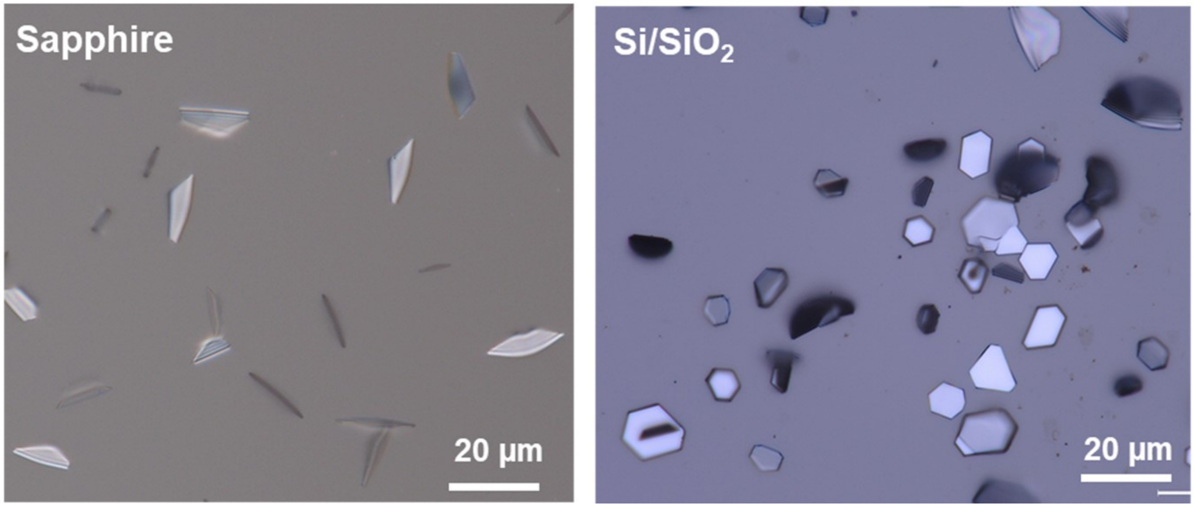

**SI. 6.** SEM images of $BiSb(Te_{1-y}Se_y)_3$ flakes show thickness – dependence of the temperature gradient. the temperature gradient. S1 and S2 were grown at $T_1$ = 500 °C, $T_2$ = 420 °C, $\Delta T$ = 80 and $T_1$ = 500 °C, $T_2$ = 330 °C, $\Delta T$ = 170 respectively.



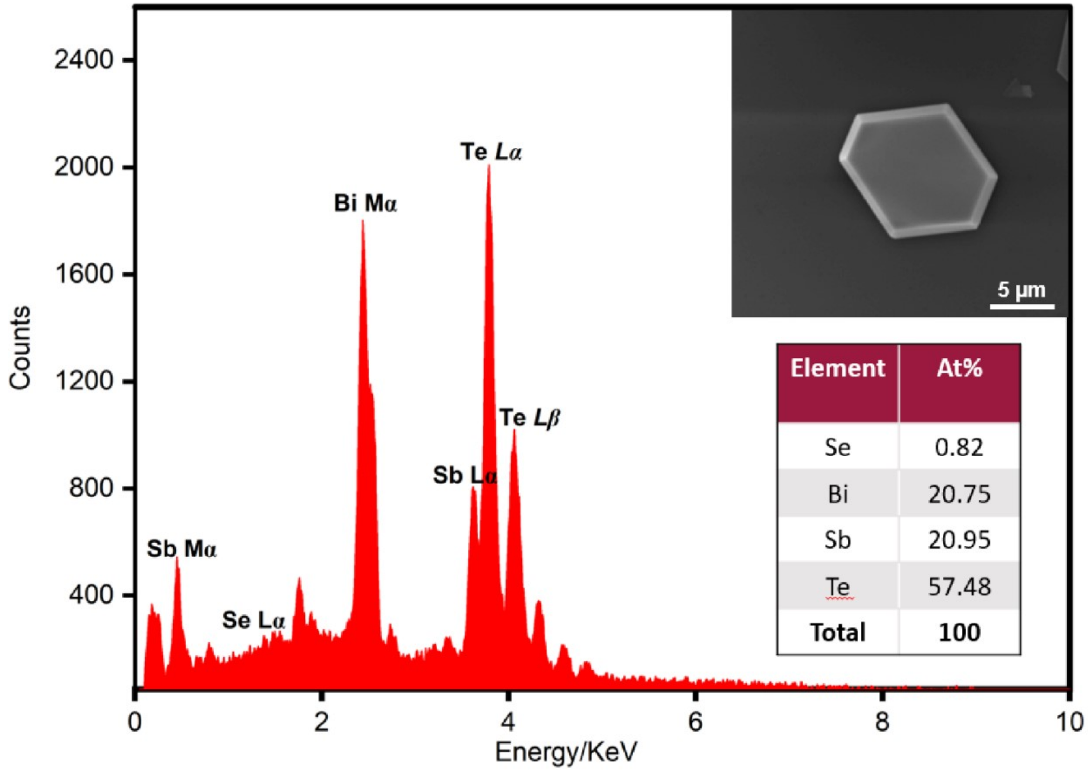

**SI. 7**. Optical microscope image of large size $BiSb(Te_{1-y}Se_y)_3$ nanocrystals prepared at $T_1$ = 630 °C, $T_2$ = 460°C.




**SI. 8**. SEM image of $BiSb(Te_{1-y}Se_y)_3$ nanowire prepared at short growth duration $t$ = 4 h.

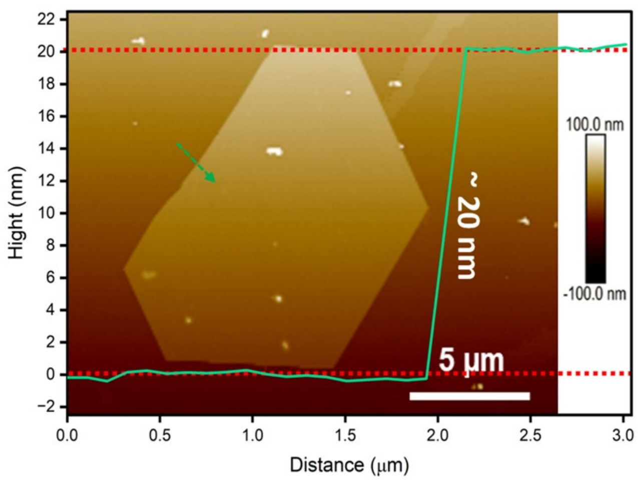



**SI. 9**. Optical microscope images of $BiSb(Te_{1-y}Se_y)_3$ nanocrystals show different growth direction on different substrate materials.




| Element | At% |
|---|---|
| Se | 0.82 |
| Bi | 20.75 |
| Sb | 20.95 |
| Te | 57.48 |
| **Total** | **100** |

**SI. 10**. EDX spectrum (15 kV) of $BiSb(Te_{0.99}Se_{0.01})_3$ crystal.




**SI. 11.** AFM image of a thin $BiSb(Te_{1-y}Se_{y})$ nanocrystal demonstrating the planer hexagonal morphology.