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Supplementary Materials: Novel Terahertz Properties of Nanostructured $Mn_{3+0.53}Sn$ Films with Different Crystalline Orientations Driven by Ostwald Ripening on (0001) *c*-Al_2O_3

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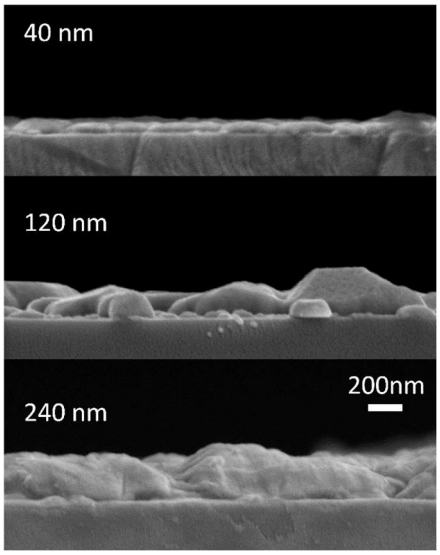

Figure S1. Cross-sectional SEM images of $Mn_{3+0.53}Sn$ films. From top to bottom, the images are corresponding to S4 (40 nm), S6 (120 nm) and S1 (240 nm), respectively.

Note:

The growth of $Mn_{3.53}Sn$ film on Al_2O_3 (0001) surface is very similar to a well-known Stranski - Krastanow (SK) mode. (The surface morphology of S1 and S4 can be found in the main text). The SK mechanism describes the emerge of islands under elastic stress due to lattice mismatch between the two materials at the initial stage of growth. The most important role in describing the growth of epitaxial nanostructures by the SK mode is played by the concepts of critical thickness of the transition from two-dimensional to three-dimensional growth [1].

However, there is a small lattice mismatch between (0001) *c*-Al_2O_3 and (0001) Mn_3Sn. And a 120 nm thick Mn_3Sn film (S6) still exist in an island morphology. The critical thickness is far beyond the one described in SK mechanism. Discontinuous surface morphology was observed even in samples with a thickness of 300 nm. Such being the case, the role of Ostwald Ripening during the film growth cannot be overlooked. The situation is more prominent in the case of a large difference between the melting points of alloy components while the temperature of phase formation of alloy is much higher than one of them, e.g., Mn and Sn.

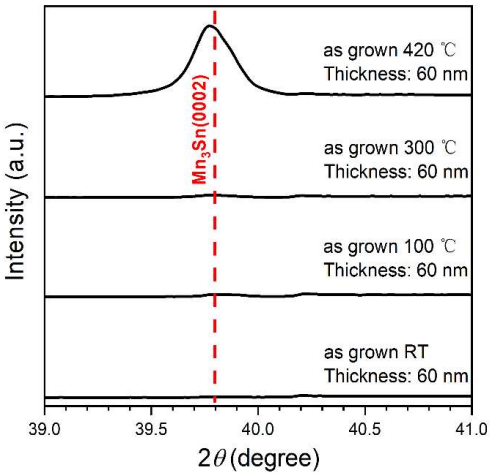

Figure S2. Structural characterization of 60 nm $Mn_{3+0.53}Sn$ films deposited at different substrate temperature. From the top to the bottom, they are S7, S8, S9 and S10.

Note:

A strong (0002) Mn_3Sn peak is observed in S7 without any noticeable impurity peaks. However, there is no diffraction peak belonging to P63/MMC Mn_3Sn in S8, S9 and S10 (Figure S2). It suggests that the phase formation temperature of Mn_3Sn nanofilms is 420 °C, which is consistent with literatures (~300–500 °C).

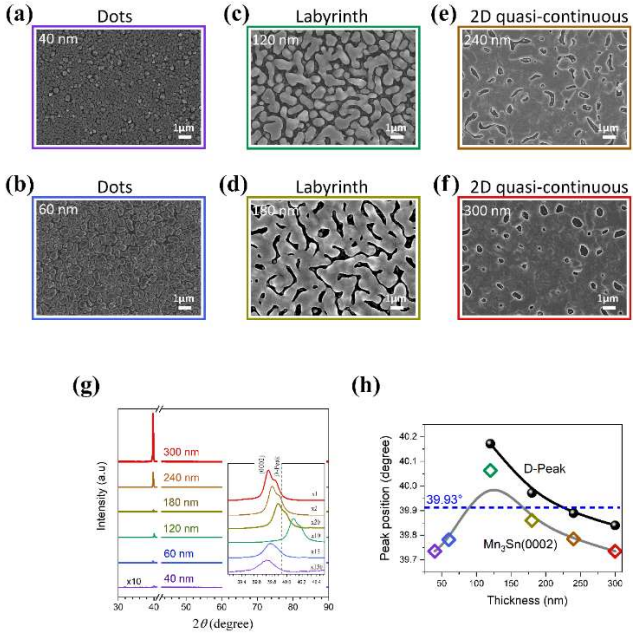

Figure S3. Structural evolution of ‖-type $Mn_{3+0.53}Sn$ films with different thickness. SEM images of $Mn_{3+0.53}Sn$ films, (**a**) S11 (40 nm), (**b**) S7 (60 nm), (**c**) S12(120 nm), (**d**) S13 (180 nm), (**e**) S14 (240 nm) and (**f**) S15 (300 nm). (**g**) XRD pattern of the samples in (**a**–**f**). The insert one is the details around the (0002) peak of Mn_3Sn (39.20°–40.45°), from which a distortion peak (D peak) can be observed at the right side of (0002) peak while the thickness is larger than 120 nm. (**h**) The position of the two peaks versus film thickness. The dash line for standard value of Mn_3Sn (0002) is also drawn together.

Note:

The nanomorphology of polycrystalline ‖-type Mn_3Sn films evolve from nanodot and labyrinth to 2D quasi-continuous film with the increase of film thickness, which

suggests strongly the role of OR. The abnormal shift of both (0002) and D peak indicates a controllable lattice distortion in nanostructured Mn_3Sn films.

Table S1. List of samples on (0001) *c*-Al_2O_3 without seed layer.

Sample Pool	Thickness	Status	Grown Temperature	Post-annealing Conditions	Relevant Figures
S1	240 nm	∥-type	420 °C	NA	Figure 2a,b Figure S1
S2	240 nm	N	100 °C	NA	Figure 2c,d
S3	240 nm	⊥-type	100 °C	5h@450 °C	Figure 2d–f
S4	40 nm	⊥-type	420 °C	NA	Figure 3a,b,d Figure 4b–d Figure 5a–d Figure S1
S5	40 nm	∥-type	420 °C	10h@450 °C	Figure 3c,d Figure 4a,c,d Figure 5a–d
S6	120 nm	∥-type	420 °C	NA	Figure S1
S7	60 nm	∥-type	420 °C	5h@450 °C	Figure S2 Figure S3b,g,h
S8	60 nm	N	300 °C	5h@450 °C	Figure S2
S9	60 nm	N	100 °C	5h@450 °C	Figure S2
S10	60 nm	N	RT	5h@450 °C	Figure S2
S11	40 nm	∥-type	420 °C	5h@450 °C	Figure S3a,g,h
S12	120 nm	∥-type	420 °C	5h@450 °C	Figure S3c,g,h
S13	180 nm	∥-type	420 °C	5h@450 °C	Figure S3d,g,h
S14	240 nm	∥-type	420 °C	5h@450 °C	Figure S3e,g,h
S15	300 nm	∥-type	420 °C	5h@450 °C	Figure S3f,g,h

References

1 Lozovoy, K. Comparative analysis of germanium-silicon quantum dots formation on Si(100), Si(111) and Sn/Si(100) surfaces. *Nanotechnology*. **2018**, *29*, 054002.