Wurtzite vs rocksalt MnSe epitaxy: electronic and altermagnetic properties - supplementary information

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1 XRD

Table 1: Specification of MBE-grown structures analyzed in the supplementary information (main text of the article contains a separate table). Each one was grown on GaAs (111) oriented substrate.

Sample number	**Layers grown (thickness in nm)**
RS1 (UW1932)	**MnSe (200)**
WZ2 (UW1961)	ZnSe (70) | ZnTe (50) | (Cd,Mg)Se (50) | CdSe (10) | **MnSe (20)**
WZ4 (UW2045)	ZnSe (70) | ZnTe (50) | (Cd,Mg)Se (50) | CdSe (10) | **MnSe (2600)**
WZ5 (UW2166)	ZnSe (70) | ZnTe (50) | CdSe (40) | **MnSe (1000)**

1.1 Reciprocal space maps

The position of the 2θ angle read from reciprocal space maps (RSM) of the RS1 sample indicates the rocksalt crystal structure (RS) of the MnSe layer with values of $2\theta = 58.625°$ and $2\theta = 87.56°$ respectively for (2 2 2) symmetrical and (-2 2 4) asymmetric reflections. RSMs are shown in Figures 1 and 2, respectively.

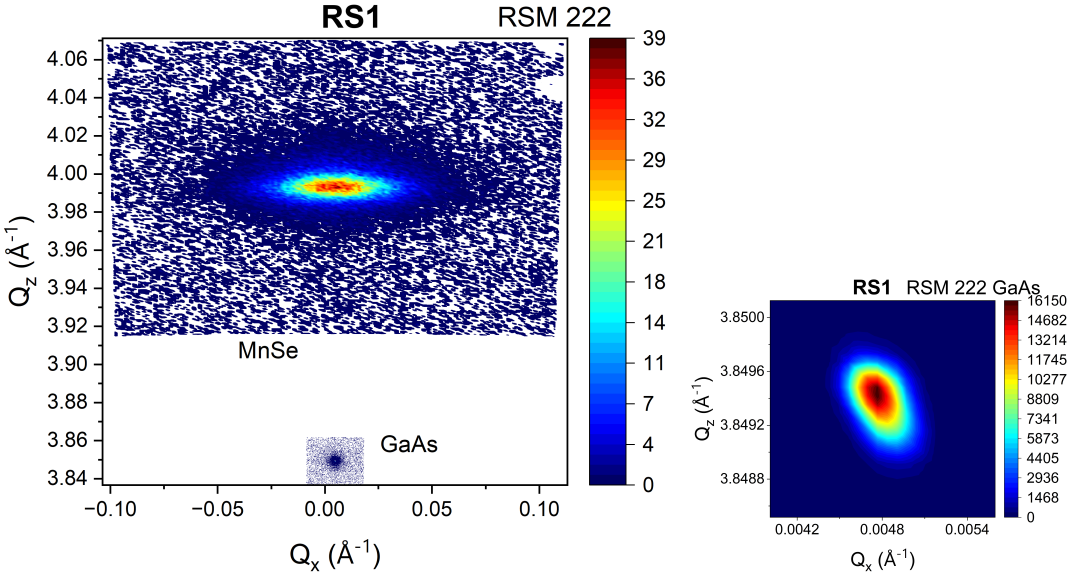

Figure 1: Reciprocal space map of symmetrical (2 2 2) reflection for 200 nm rocksalt MnSe layer (sample RS1). On the right hand side there is enlarged GaAs RSM.

Using Braggs' law $n\lambda = 2d \sin\theta$ (where $\lambda = 1.5406$ Å) and formula for d in a cubic system $\frac{1}{d} = \frac{\sqrt{h^2+k^2+l^2}}{a}$ we obtained lattice parameter $a = 5.4505$ Å from (2 2 2) reflection and $a = 5.454$ Å from (-2 2 4) reflection. This difference comes from the use of a model for a nondeformed cubic system, and therefore indicates probably slightly deformed MnSe crystall unit cell. Also, by looking at the triangle of relaxation in Figure 2, we can see that the MnSe layer is almost fully relaxed. The symmetrical (2 2 2) reflection from Figure 1 also shows that there is a mosaic in the MnSe layer. This is indicated by the horizontal oval shape of the peak.

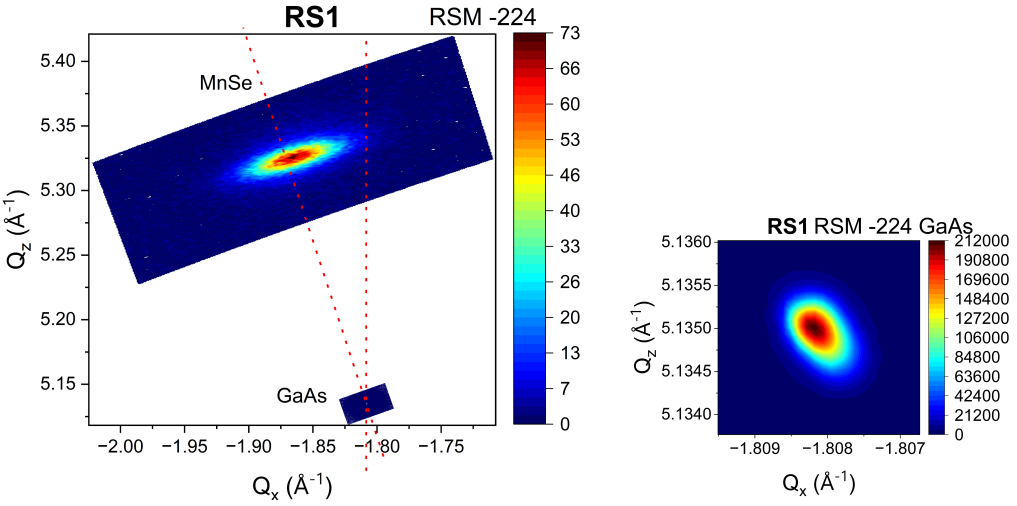

Figure 2: Reciprocal space map of asymmetrical (-2 2 4) reflection for 200 nm rock-salt MnSe layer (sample RS1). On the right hand side figure we present enlarged map of GaAs substrate.

In Figure 3, which shows WZ2 sample, the RSM of the (0 0 4) reflection of the ZnSe and ZnTe layers is presented. We observe that ZnTe has a zinc blende crystal structure with lattice parameter $a = 6.097$ Å computed from the peak position obtained from the shown map, and we do not observe a peak from the CdSe cubic crystal structure that should be visible close to the ZnTe peak. This means that CdSe must have another crystal structure, which is a hexagonal wurtzite structure. When we look at the relaxation triangle, we see that the ZnTe layer is fully relaxed.

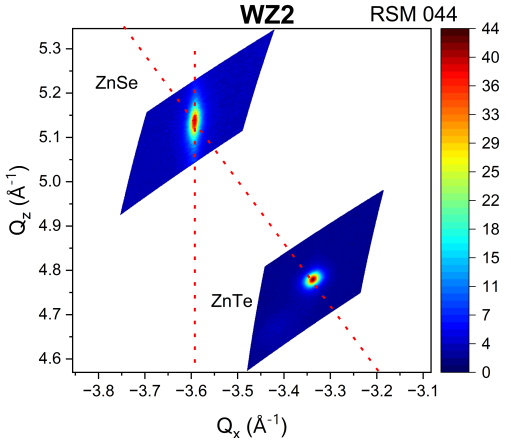

Figure 3: Reciprocal space map of asymmetrical (0 4 4) reflection for buffers in a sample containing wurtzite MnSe (sample WZ2) while being focused on the ZnSe and ZnTe layers.

1.2 ϕ-scans

We made a set of ϕ-scans to visualize the presence of crystal twinnings in the MBE-grown layers. ϕ-scans are made by rotating the sample in the ϕ angle with constant ω and 2θ angles. Their main role is to show the symmetry of the investigated materials. In Figure 4 we can see ϕ-scan of RS1 sample substrate GaAs for the (0 4 4) reflection in the skew geometry. Furthermore, we show three ω-scans with an optimized setup to show evidence of peaks only in intervals of 120° not 60°.

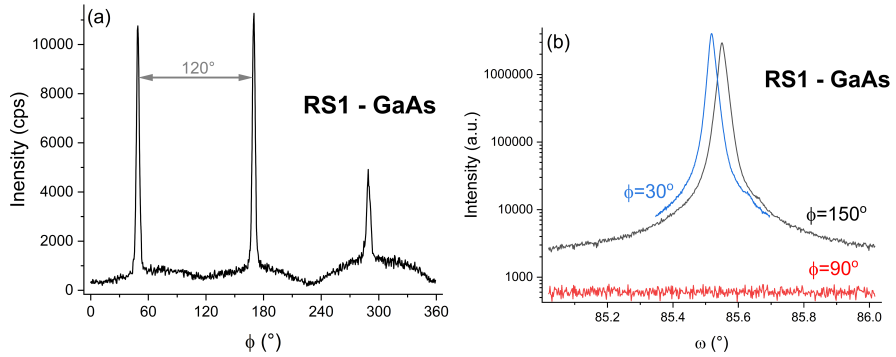

Figure 4: (a) ϕ-scan of 200 nm rock-salt MnSe layer (sample RS1) substrate GaAs reflection (0 4 4) in skew geometry. (b) ω-scan of GaAs substrate reflection (0 4 4) in skew geometry for three different ϕ angles.

In Figure 5 we show ϕ-scan of MnSe layer in RS1 sample. The clearly observed six-fold symmetry indicates crystal twinnings in the MnSe layer, which has a cubic crystal structure. The peaks shown in Figure 6 show that the difference in signal intensity is due to precession of the crystal axis and the intensity appears to be the same when we optimize the setup for exact peaks.

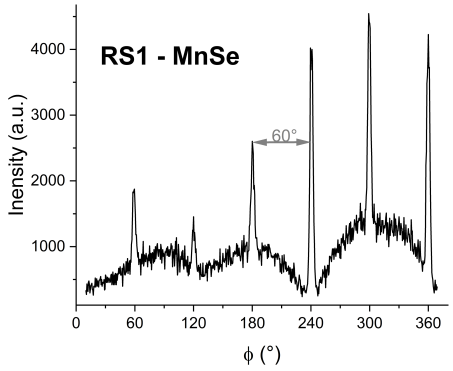

Figure 5: ϕ-scan of 200 nm rock-salt MnSe layer (sample RS1) reflection (0 4 4). 6-fold symmetry clearly shows presence of crystal twinnings in MnSe layer.

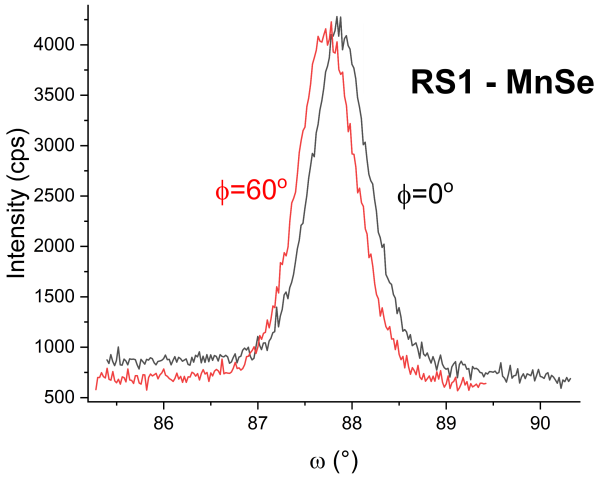

Figure 6: ω-scans of 200 nm rock-salt MnSe layer (sample RS1) reflection (0 4 4) in skew geometry for different values of ϕ angles.

We also show six ω-scans, each for different ϕ angle with an interval of 60° that we show in Figure 7. The visibility of six peaks is evidence of crystal twinning in the ZnTe layer in WZ2 sample.

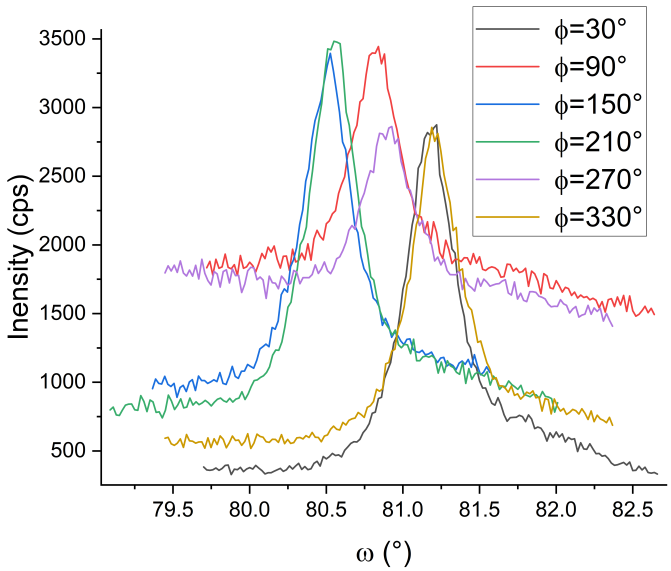

Figure 7: ϕ-scans of a buffer ZnTe layer (in a sample containing wurtzite MnSe - WZ2) of reflection (0 4 4) for different ϕ angles. There is clearly visible 6-fold symmetry in ZnTe layer which indicates to crystal twinnings.

hkl	ICDS 643584 2θ [deg]	Experimental 2θ [deg]
111	28.34	28.33
222	58.63	58.61
002	32.84	32.83
004	68.85	68.83

Table 2: The table presents comparison of the ICDS and experimental values of 2θ for rock-salt MnSe.

2 Reference crystal structure data

The reference rock-salt MnSe structural data can be obtained from XRD and Inorganic Crystal Structure Database (ICSD). The following information can be found. ICSD 643584: lattice parameter a=5.450 Å, angles: 90°, 90°, 90°, space group $Fm-3m$ (225). The parameters obtained in our layers are calculated in the previous section. In our case the unit cell is slightly deformed, $a = 5.452 \pm 0.002$ Å.

The reference structural data of the wurtzite MnSe can be obtained only from [9] and [10] references in the main text. ICSD code 643594 contains data obtained from publication [9], network parameters $a = 4.12$ Å, $c = 6.72$ Å, angles: 90°, 90°, 120°, space group: $P63mc$ (186). In our case, the c parameter is, depending on the sample, between 6.74 Å and 6.77 Å and is close to the parameter from work [10] 6.783 Å.

3 Thickness of wurtzite MnSe layers

In the experiments presented in the main text we focused on the thin wurtzite layers of several tens of nanometer thickness. They exhibit the best surface quality and RHEED patterns consisting of sharp and bright lines. With the increasing layer thickness, RHEED pattern dims. The lines also become broadened for the thicknesses larger than around 500 nm. At the growth termination around 1μm of the MnSe thickness the pattern is still present but strongly dimmed and lines are very broad (Fig. 8). (Similar qualitative picture has been obtained for the surface of another 2.6 μm thickness MnSe sample - UW2045.) It may be a suggestion that the surface is strongly developed and complex but within single grains of the crystal atoms are still highly-ordered.

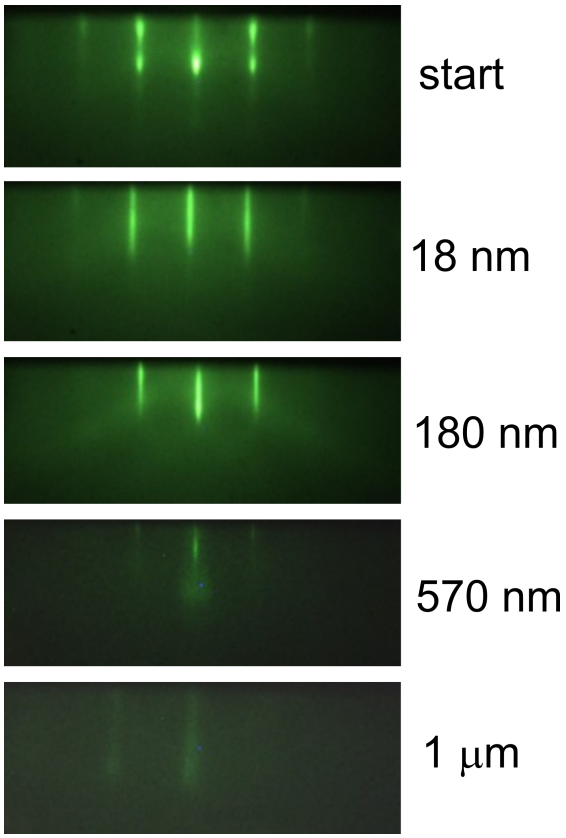

Figure 8: Evolution of the RHEED pattern of the crystal surface with increasing thickness of the wurtzite MnSe layer (number on the right) measured during the growth of UW2166 sample. The first image has been taken just after the termination of the CdSe buffer growth.