

## Supplementary Information

### Interface control by homoepitaxial growth in pulsed laser deposited iron chalcogenide thin films

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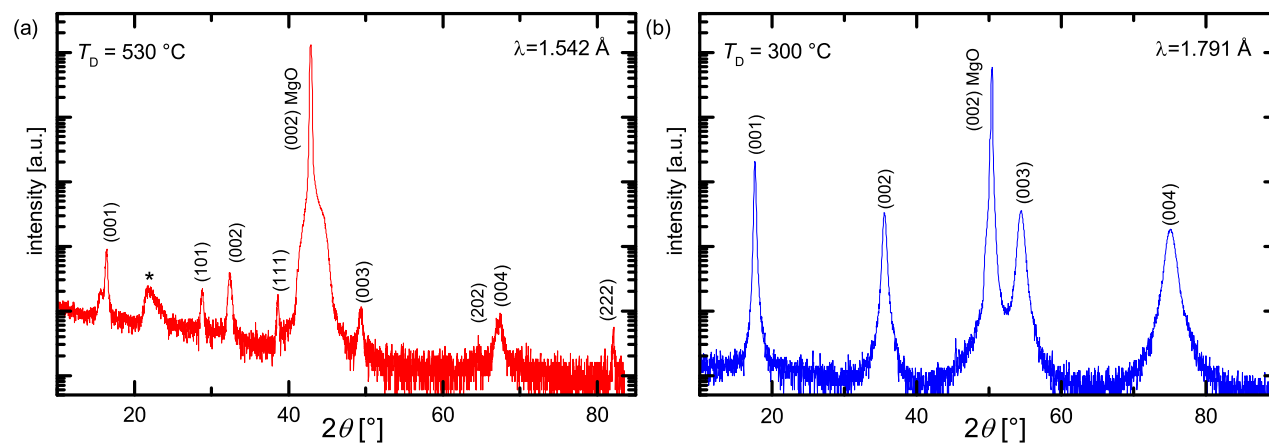
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### SI XRD of unseeded film at high deposition temperature

In Fig. S1 the XRD  $\theta$ - $2\theta$ -scans of films grown directly on MgO at  $T_D$ 's of 530 °C and 300 °C are shown. Besides  $c$ -axis grown components of the film with  $T_D = 530$  °C ((00 $l$ )-reflections) there are additional *out-of-plane* orientations observable. Peaks of the ( $h0l$ ) as well as the ( $hkl$ ) orientation are found [Fig. S1(a)]. With decreasing  $T_D$  the intensity of the peaks for orientations different from (00 $l$ ) decreases rapidly. The XRD  $\theta$ - $2\theta$ -scan of the film with  $T_D = 300$  °C shows exclusively  $c$ -axis orientation *out-of-plane* [Fig. S1(b)].



**Figure S1.** XRD  $\theta$ - $2\theta$ -scan of FeSe<sub>1-x</sub>Te<sub>x</sub> thin films grown on MgO at (a)  $T_D = 530$  °C and (b)  $T_D = 300$  °C. The  $\star$  indicates the  $\lambda/2$ -peak of MgO.

## S II In-plane texture: Comparison between unseeded and seeded films on MgO

Evaluating the FWHM of the peaks of the *in-plane* orientation of the temperature series [Fig. S 2] shows a strong increase of  $\Delta\phi$  for both texture components for the thin films without seed layer for decreasing  $T_D$ . In contrast to that, the  $\Delta\phi$  values of thin films with seed layer stay more or less constant with decreasing deposition temperature.

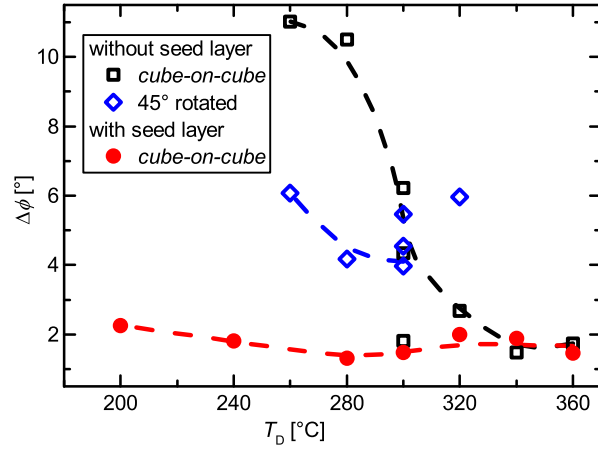


Figure S2.  $\Delta\phi$  of the texture components of the samples without and with seed layer in dependence of  $T_D$ .

## S III Resistance measurements of films without seed layer

From the resistance measurements [Fig. S 3]  $T_{c,90}$  and  $\Delta T_c$  for the thin films without seed layer are obtained.  $\Delta T_c$  is calculated from the temperature where the resistance reaches 90% of the normal state resistance minus the temperature where the resistance reaches 10% of the normal state resistance.

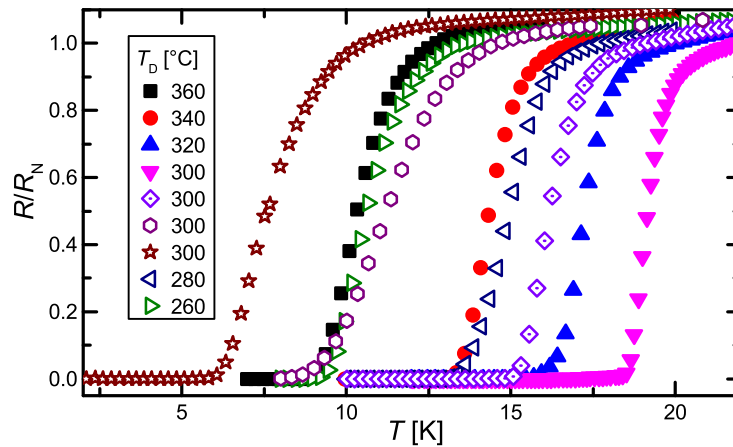
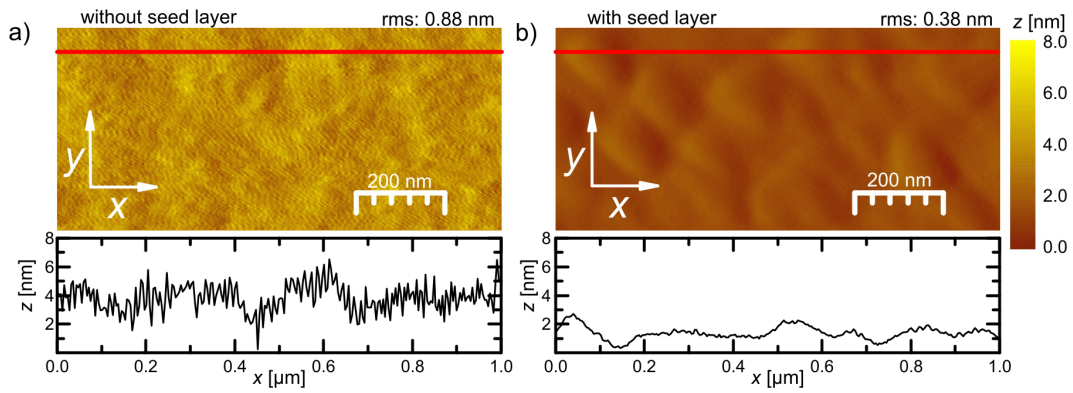


Figure S3. Temperature dependence of the normalized resistance ( $R_N = R(100\%)$ ) for thin films without seed layer.

## SIV Comparison of the surface morphology

The surface morphology of a sample without seed layer [Fig. S4(a)  $T_D = 300^\circ\text{C}$ ,  $\tau = 1$ ] and a sample with seed layer [Fig. S4(b)  $T_D = 240^\circ\text{C}$ ] was evaluated with AFM. The rms-values of the samples show an improvement of the surface roughness by a factor of 2.



**Figure S4.** AFM images of a thin film grown without seed layer [(a)  $T_D = 300^\circ\text{C}$ ,  $\tau = 1$ ] and a film on the seed layer [(b)  $T_D = 240^\circ\text{C}$ ]. The red lines mark the position of the profiles below.