Supplementary Information

The impact of post-deposition annealing on the performance of solution-processed single layer In_2O_3 and isotype In_2O_3/ZnO heterojunction transistors

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Section SI1. Experimental results and theoretical analysis

Table S1. Thickness of solution-processed In_2O_3 and ZnO layers annealed at various temperatures as measured by optical ellipsometry.

Annealing temperature (°C)	Layer thickness (nm)	
	ZnO	In_2O_3
200 °C	7.0	6.5
300 °C	6.8	5.8
400 °C	7.2	5.9
600 °C	8.5	6.7

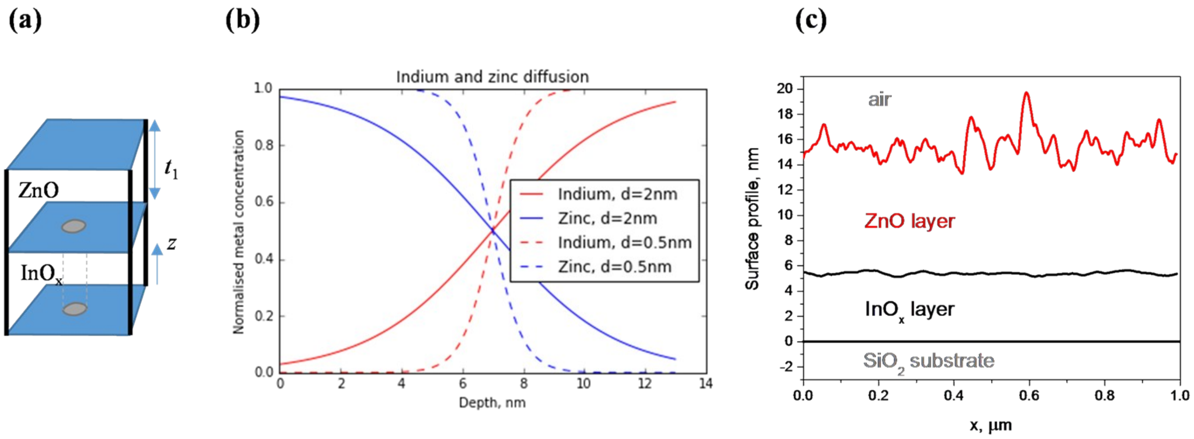

Figure S1. (a) Illustration of a simulation model with a bilayer film: a thin cylinder with flat bases is first considered for the XPS signal calculation. (b) Examples of atomic profile of the bilayer from a flat cylinder with different diffusion constants: 2 nm (solid line) and 0.5 nm (dashed line). (c) Example of a rough profile of a bilayer structure.

In order to calculate the XPS signal from zinc and indium atoms we assume a simple model illustrated in **Fig. S1(a)**. Vertical distribution of the atomic concentrations di depends on zinc oxide thickness t_1 and diffusion coefficient d:

$$Di_{Zn} = {}^{1}\!\big/\left[\exp\left((z - t_1)/d\right) + 1\right] \text{ for Zn and} \tag{SI-1}$$

$$di_{In} = 1 - {}^{1}\!\big/\left[\exp\left((z - t_1)/d\right) + 1\right] \text{ for In} \tag{SI-2}$$

The example of the atomic profile is shown in **Fig. S1(b)**. The approximate XPS signal intensity depends exponentially on the inelastic mean free path l which is equal 2 nm for the studied films. Thus in order to calculate the XPS signal intensity dI from the zinc atoms in a flat thin cylinder, we need to solve the following integral:

$$dI_{Zn} = \int_0^{\infty} \frac{\exp\left(-\frac{z}{l}\right)}{\exp\left(\frac{z - t_1}{d}\right) + 1} dz \tag{SI-3}$$

In general, the thickness is a variable which depends on roughness. Here we assume a similar roughness in both x and y direction (**Fig. S1(c)**). From histograms in **Fig.1 (c)** we can get the distribution of the deviations δt_1 from the average thickness t_{1avg}: $P_{\mathrm{Zn}}(\delta t_1)$, where deviation is defined as $t_1 = t_{1avg} + \delta t_1$. Therefore we obtain the following expression for the average signal from zinc atoms:

$$I_{Zn} = \int_{\delta t_1} \int_0^{\infty} \frac{P_{Zn}(\delta t_1) \cdot exp(- z/l)}{\exp\left(\frac{z - t_{1avg} + \delta t_1}{d}\right) + 1} dz\, d\delta t_1 \tag{SI-4}$$

In order to get a similar expression for indium atoms, it is necessary to integrate the flat cylinder intensity by both ZnO and InO$_x$ roughness:

$$I_{In} = \iint_{\delta t_1 \delta t_2} \int_0^{\infty} \mathrm{P}_{Zn}(\delta \mathrm{t}_1)\mathrm{P}_{In}(\delta \mathrm{t}_2)\exp\left(-\frac{z}{l}\right)\left[1 - \frac{1}{\exp\left(\frac{z - t_{1avg} + \delta t_1 + \delta t_2}{d}\right) + 1}\right] dz\, d\delta t_1 d\delta t_2. \tag{SI-5}$$

For comparison between XPS signal intensities from indium and zinc, we assume equal concentration of indium and zinc atoms in indium oxide and zinc oxide layers respectively. The Zn:In ratio is calculated simply as I_{Zn}/I_{In} . The diffusion coefficients derived from the fitting between the experimental ratio and calculated ratio are shown in the **Table S2** below:

Table S2. Summary of elemental composition measured via XPS, layer surface roughness and simulated diffusion coefficients as a function of annealing temperature.

Annealing temperature (°C)	Experimental Zn:In ratio, from XPS	Roughness (nm)		Simulated diffusion coefficient (nm)
		ZnO	InO$_x$	

200	96:4	2.5	0.22	0.5
300	96:4	2.7	0.25	0.5
400	91:9	2.6	0.8	1.4
600	81:19	4.4	1.3	2.9

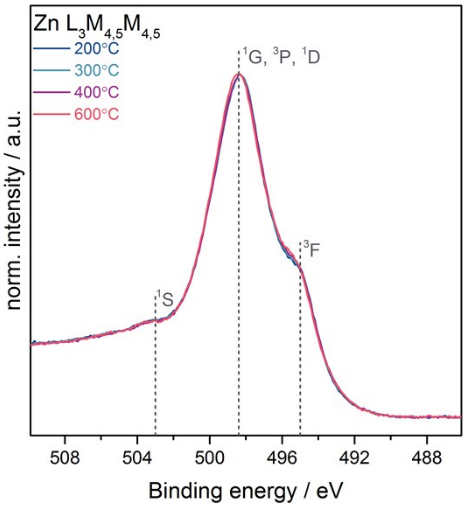

Figure S2. Zn $L_3M_{4,5}M_{4,5}$ Auger line of the In_2O_3/ZnO bilayer samples annealed at varying temperatures.

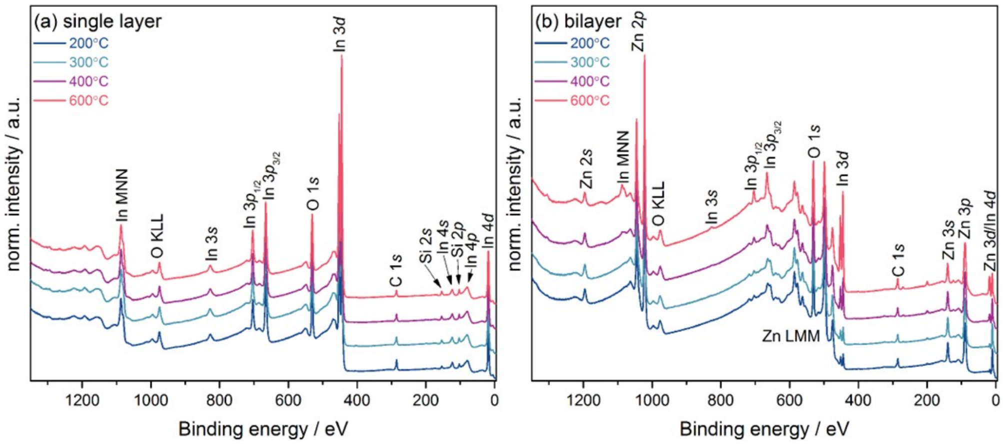

Figure S3. XPS survey spectra of the (a) In_2O_3 single layer and the (b) In_2O_3/ZnO bilayer samples annealed at varying temperatures.

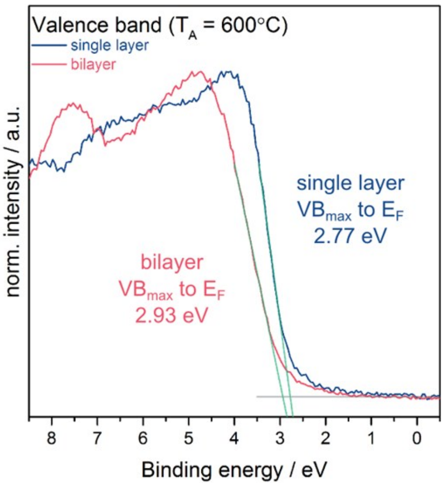

Figure S4. XPS valence band spectra of the In_2O_3 single layer and the In_2O_3/ZnO bilayer samples annealed at 600 ˚C including the linear fits used to determine the VB_{max} to E_F separation.

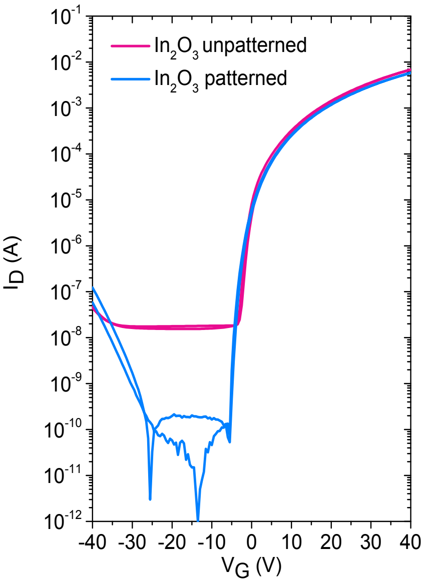

Figure S5. Transfer curves of patterned and un-patterned In_2O_3 single layer transistors annealed at 400 °C.