Supplementary Information (SI)

Evolution of optical properties and electronic structures: band gaps and critical points in $Mg_xZn_{1-x}O$ ($0 \le x \le 0.2$) thin films

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1. XPS measurements

Here the needed information for XPS results is provided. Peak-differentation-imitating analysis for Zn 2*p* and Mg 2*p* is provided in Figure 1b-c, and their detailed parameters (including peak location, full-width-at-half-maximum (FWHM), energy separation and atomic ratio) are listed in Table S 1. The fitting formula is the combination of Lorentz function and Gaussian function. The global fitting accuracy is quantified by the standard deviation (*STD*), which is described as:

$$STD = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(X_{Mi} - X_{Ci}\right)^2}$$

where N is the number of measured data, X_{Mi} is the measured data, and X_{Ci} is the calculated data. Since the as-obtained *STD* is global wide, the much smaller *STD* compared with the intensity of data is able to indicate that the calculated atomic ratio is reasonable and accurate (Red Circle in Figure S 1).

Table S 1. Peak positions and FWHM of the XPS Zn 2*p* and Mg 2*p* peaks obtained from sample 4:1(x=0.2), 7:1(x=0.12) and 10:1(x=0.09), and the calculated [Zn]/[Mg] atomic ratio derived from XPS (Corresponding to the fitting results of Fig. 1 in the manuscript, no error bars).

Sample		4:1	7:1	10:1
Zn $2p_{3/2}$	Position/eV	1021.32	1021.24	1021.19
	FWHM/eV	1.60	1.64	1.74
Zn $2p_{1/2}$	Position/eV	1044.40	1044.32	1044.25
	FWHM/eV	1.75	1.77	1.87
Mg $2p_{3/2}$	Position/eV	49.58	49.54	49.54
	FWHM/eV	1.12	1.28	1.57
Mg $2p_{1/2}$	Position/eV	50.54	50.64	50.75
	FWHM/eV	1.35	1.28	1.28
Atomic ratio [Zn]/[Mg]	Experimental	3.80:1	6.92:1	10.01:1
	Theoretical	4:1	7:1	10:1

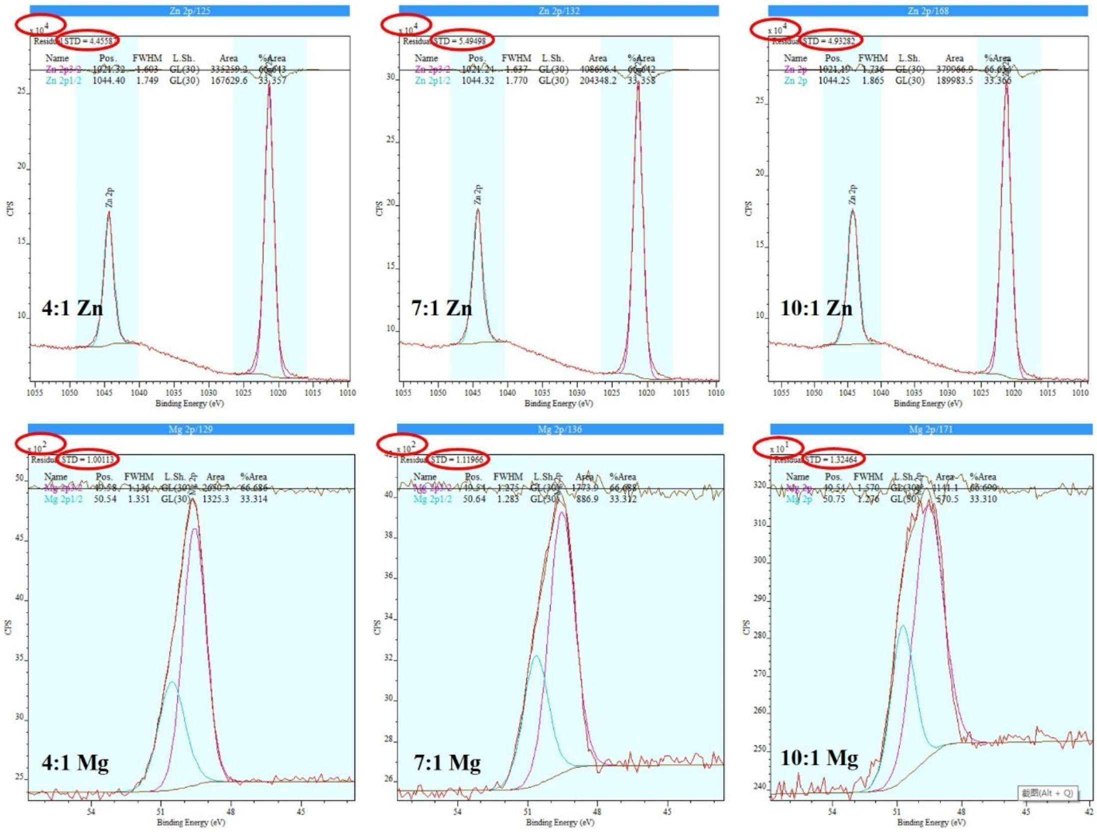

Figure S 1. The as-obtained standard deviation (*STD*) from XPS measurements of sample 4:1(x=0.2), 7:1(x=0.12) and 10:1(x=0.09).

2. Critical Point (CP) parameters

All of the fitting parameters of ZMO samples are summarized in Table S 2, in which the acquired eight CP energies (E_{th}) are in bold.

Table S 2. Main parameters of the SCP model for ZMO films extracted from the best fitting second derivatives of the dielectric functions.

	Sample	4:1	5:1	6:1	7:1	8:1	10:1	12:1	14:1	ZnO
A	A_m/no unit	0.265	0.352	0.323	0.214	0.198	0.213	0.241	0.222	0.178
	φ /rad	1.075	1.017	0.779	0.882	0.926	0.913	1.130	1.251	1.190
	E_{th}/eV	**3.650**	**3.581**	**3.523**	**3.497**	**3.482**	**3.430**	**3.424**	**3.414**	**3.322**
	γ /eV	0.233	0.239	0.230	0.183	0.174	0.175	0.178	0.169	0.149
B	A_m/no unit	0.148	0.188	0.097	0.119	0.110	0.081	0.055	0.063	0.071
	φ /rad	2.683	3.112	2.756	3.335	2.706	3.046	2.955	2.661	2.453
	E_{th}/eV	**3.973**	**3.946**	**3.859**	**3.885**	**3.814**	**3.724**	**3.692**	**3.654**	**3.544**
	γ /eV	0.265	0.220	0.126	0.163	0.159	0.107	0.137	0.171	0.178

C	A_m/no unit	0.001	0.016	0.040	0.029	0.068	0.015	0.003	0.002	0.011
	φ /rad	2.527	1.778	1.372	0.740	0.952	1.198	1.846	2.035	3.198
	$\boldsymbol{E_{th}}$**/eV**	**3.330**	**3.304**	**3.282**	**3.273**	**3.280**	**3.275**	**3.297**	**3.285**	**3.364**
	γ /eV	0.080	0.171	0.216	0.194	0.246	0.138	0.102	0.098	0.146
D	A_m/no unit	0.007	0.001	/	/	/	0.012	0.003	0.007	0.002
	φ /rad	1.704	2.216	/	/	/	1.760	3.442	3.432	2.079
	$\boldsymbol{E_{th}}$**/eV**	**3.480**	**3.481**	**/**	**/**	**/**	**3.497**	**3.490**	**3.475**	**3.477**
	γ /eV	0.134	0.089	/	/	/	0.140	0.094	0.129	0.090
E	A_m/no unit	0.099	0.045	0.084	0.052	0.028	0.067	0.006	0.020	0.002
	φ /rad	1.656	1.635	2.253	2.257	1.490	1.255	0.550	1.471	1.575
	$\boldsymbol{E_{th}}$**/eV**	**4.104**	**4.112**	**4.054**	**4.078**	**4.061**	**4.053**	**4.061**	**4.059**	**4.106**
	γ /eV	0.387	0.227	0.313	0.241	0.225	0.257	0.146	0.275	-0.125
F	A_m/no unit	0.008	0.005	0.006	0.007	0.010	0.008	0.009	0.001	0.004
	φ /rad	0.665	1.097	1.026	0.837	0.711	0.404	0.441	0.538	0.508
	$\boldsymbol{E_{th}}$**/eV**	**4.345**	**4.342**	**4.351**	**4.336**	**4.349**	**4.345**	**4.345**	**4.345**	**4.348**
	γ /eV	0.168	0.143	0.164	0.185	0.200	0.186	0.213	0.107	0.147
G	A_m/no unit	0.008	0.009	0.039	0.002	0.025	0.006	0.002	0.002	0.018
	φ /rad	3.575	4.526	3.576	0.917	3.239	2.213	1.880	2.109	2.904
	$\boldsymbol{E_{th}}$**/eV**	**3.718**	**3.719**	**3.687**	**3.658**	**3.708**	**3.732**	**3.718**	**3.727**	**3.686**
	γ /eV	0.167	0.122	0.230	0.081	0.145	-0.070	-0.062	-0.070	0.122
H	A_m/no unit	0.050	0.019	0.011	0.018	0.006	0.106	0.008	0.010	0.008
	φ /rad	6.226	6.045	6.230	5.316	5.308	2.444	3.633	2.662	1.920
	$\boldsymbol{E_{th}}$**/eV**	**3.844**	**3.818**	**3.825**	**3.859**	**3.796**	**3.862**	**3.831**	**3.829**	**3.766**
	γ /eV	-0.353	-0.185	-0.121	-0.133	-0.118	0.252	0.108	0.144	0.113

3. SEM images

Here some cross-sectional SEM images are provided. After the sample preparation in the process of SEM test, in order to improve the conductivity and to acquire more clear SEM images, we deposited ~2.5 nm gold film on the section by sputtering (19 s).

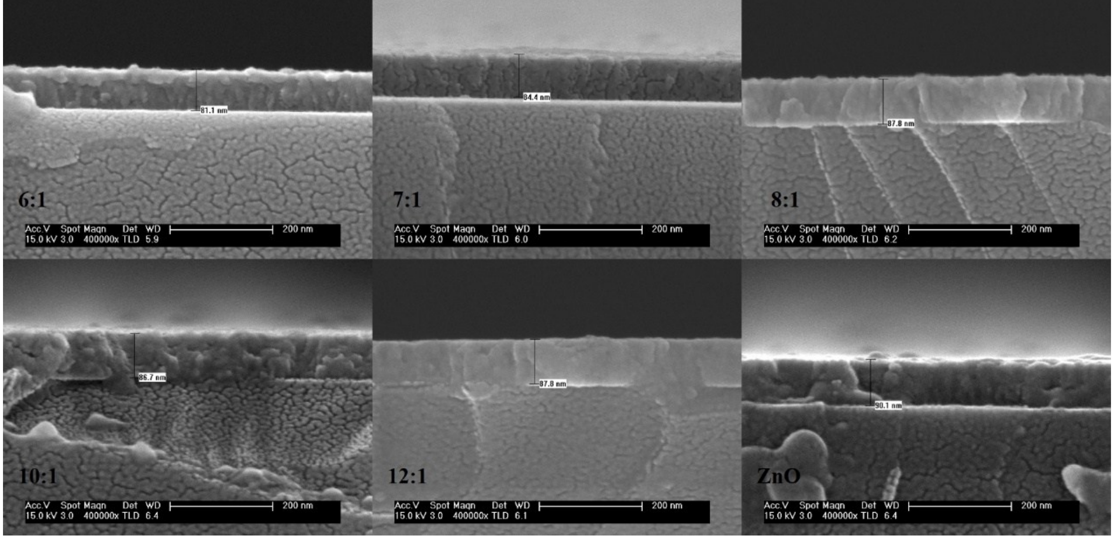

Figure S 2. The cross-sectional SEM images of sample 6:1(x=0.14), 7:1(x=0.12), 8:1(x=0.11), 10:1(x=0.09), 12:1(x=0.08) and ZnO(x=0).