Electronic Supplementary Information (ESI)

Ionic Liquid/ZnO$(000\bar{1})$ Single Crystal and Epitaxial Films Interfaces Studied through a Combination of Electrochemical Measurements and Pulsed Laser Deposition Process in Vacuum

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The supporting Information includes 8 Figures.

Annealing treatment of ZnO single crystal substrates

AFM images were measured on Seiko Instruments SPA-400.

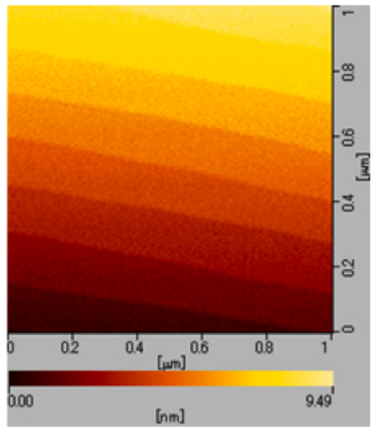

Figure S1. Typical AFM image of an O-polar ZnO(000$\bar{1}$) single crystal surface obtained by annealing in an air furnace at 1200°C for 2h.

Charge transfer resistance vs. electrode potential *U*

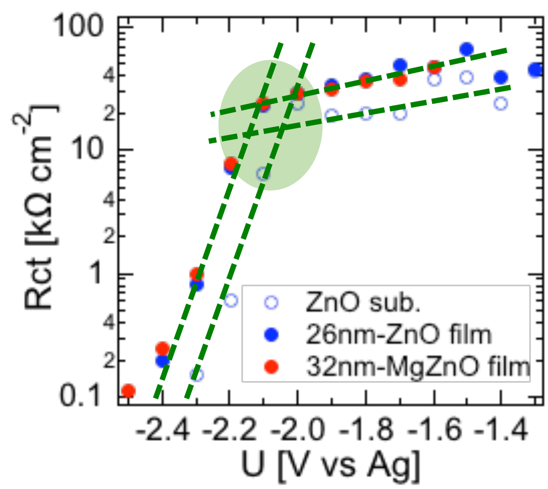

Figure S2. R_{ct} values in the equivalent circuit model are plotted against the electrode potential of around –2V vs. Ag, in which R_{ct} starts to drastically decrease at potentials around –2.1 V vs. Ag.

Surface states vs. electrode potential *U*

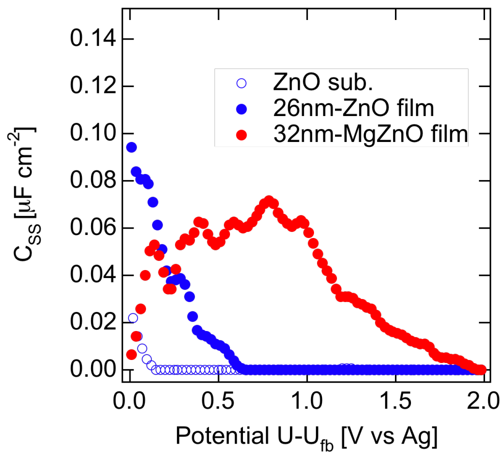

Figure S3. C_{SS} values for the ZnO substrate, ZnO and MgZnO films plotted against U relative to each flat band potential, respectively.

Thickness-dependences of the donor density for ZnO and MgZnO films

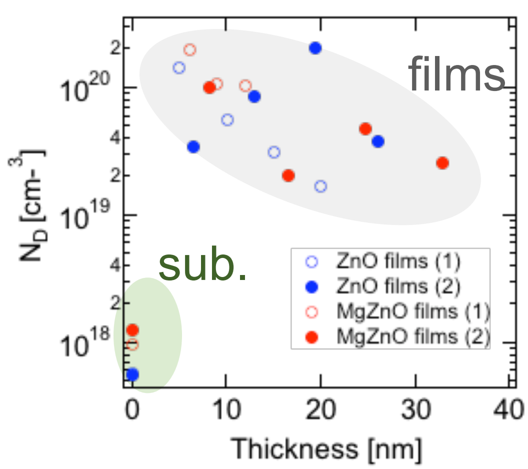

Figure S4. The donor densities plotted against the thicknesses for ZnO and MgZnO films, respectively.

Mg content in the MgZnO film

The Mg content x_{Mg} in MgZnO was estimated to be 0.250 from the band gap and *c*-axis lattice parameter of a MgZnO film grown on an α-Al_2O_3(0001) substrate under exactly the same conditions as on the single crystal ZnO substrate: the band gap *E*g was 3.76 eV (Fig. S5) and *c*-axis lattice parameter c_{axis} was 0.5175 nm (Fig. S6), respectively.

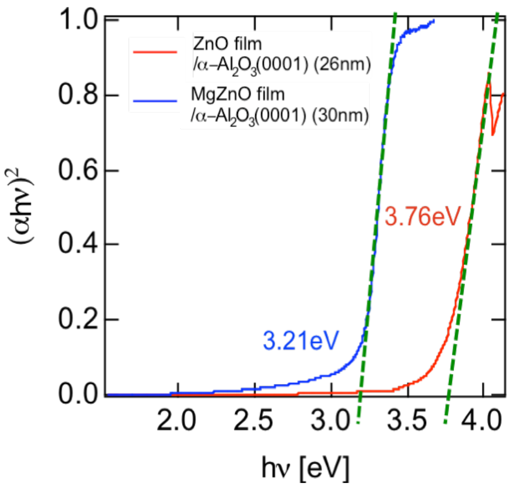

Figure S5. Absorption spectra of ZnO and MgZnO films fabricated on an α-Al_2O_3(0001) substrate, respectively.

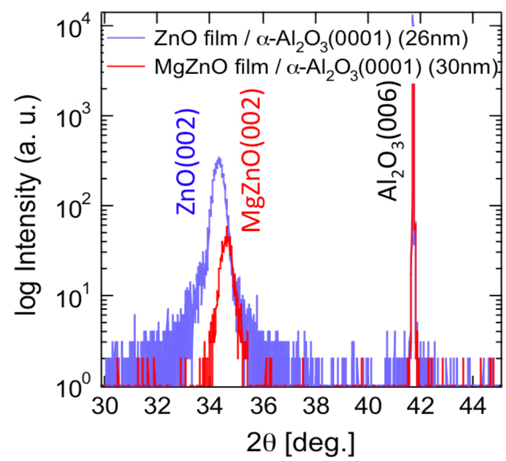

Figure S6. XRD patterns of ZnO and MgZnO films fabricated on an α-Al_2O_3(0001) substrate, respectively.

From the correlation(s) between the Mg content and the band gap, and/or the Mg content and the lattice parameters as expressed by the following equations [S1],

$$E_g = (3.26 \pm 0.02) + (2.01 \pm 0.0014)x_{Mg}$$

$$C_{axis} = (0.52052 \pm 7.36 \times 10^5) - (0.011945 \pm 0.00133)x_{Mg}$$

the Mg contents were estimated to be x_{Mg}=0.248 and 0.253, respectively (Fig. S7), averaging out to be 0.250.

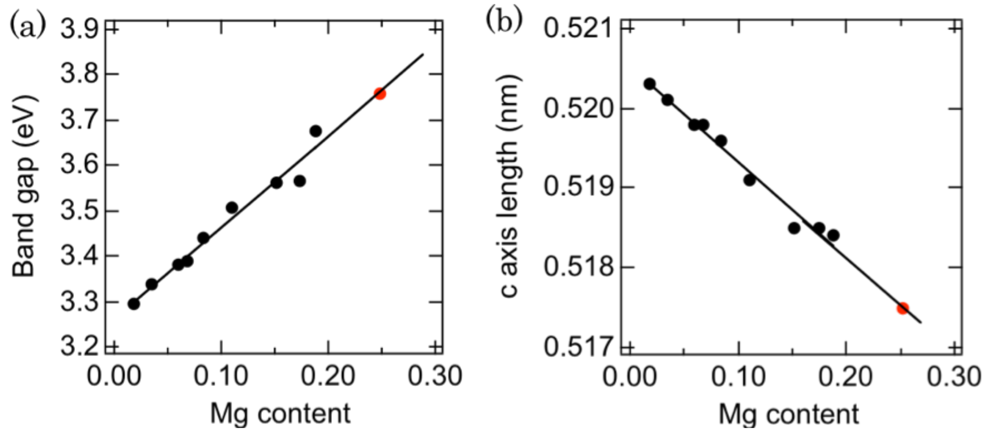

Figure S7. Correlation(s) between the Mg content and the band gap, and/or the Mg content and the lattice parameters. The data points marked with a red filled circle are for the MgZnO film in the present experiment.

RHEED patterns for ZnO and MgZnO films

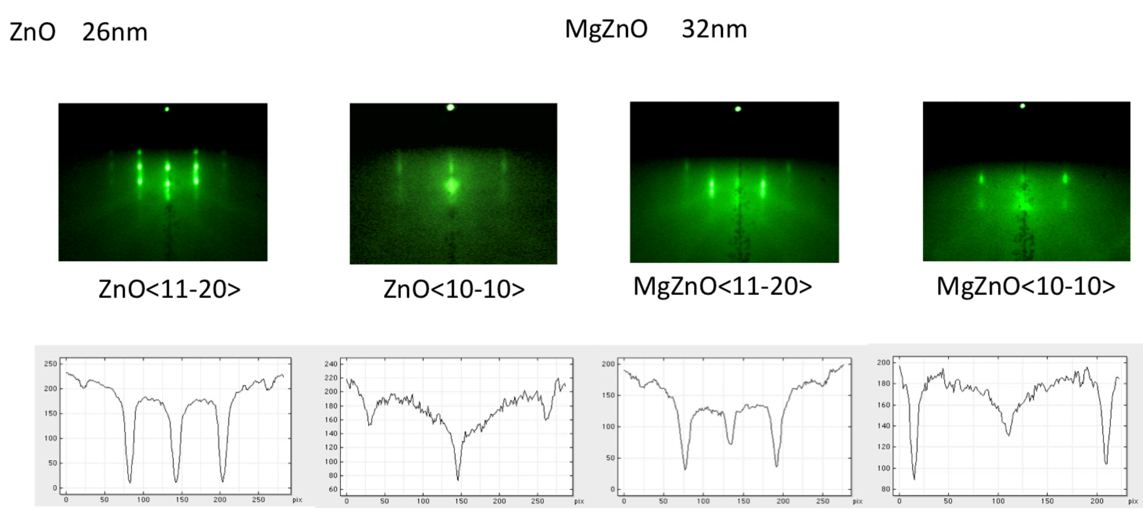

Figure S8. Set of RHEED patterns and their intensity profiles of 26nm-thick ZnO and 32nm-thick MgZnO films, observed with the incident electron beam along the directions of <11-20> and <10-10>, respectively. The primary electron beam energy is 15-20 keV.

REFERENCES

(S1) Y. Matsumoto, M. Murakami, Z.-W. Jin, A. Ohtomo, M. Lippmaa, M. Kawasaki and H. Koinuma, Combinatorial Laser Molecular Beam Epitaxy (MBE) Growth of Mg-Zn-O Alloy for Bnad Gap Engineering, *Jpn. J. Appl. Phys.*, 1999, **38**, L603-605.