

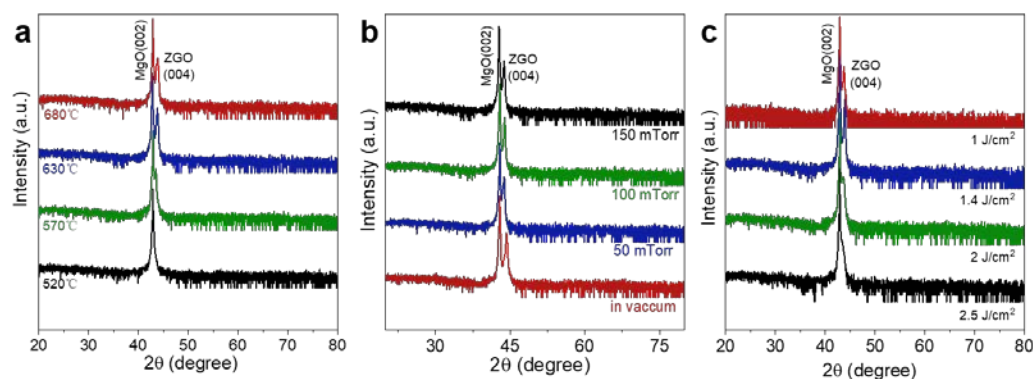
# Supplementary Materials: Epitaxial Growth and Stoichiometry Control of Ultrawide Bandgap $\text{ZnGa}_2\text{O}_4$ Films by Pulsed Laser Deposition

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**Figure S1.**  $\theta$ - $2\theta$  XRD full scans of ZGO films over MgO showing c-axis-oriented growth which deposited under different conditions of (a) substrate temperature, (b) oxygen pressure and (c) laser fluence.

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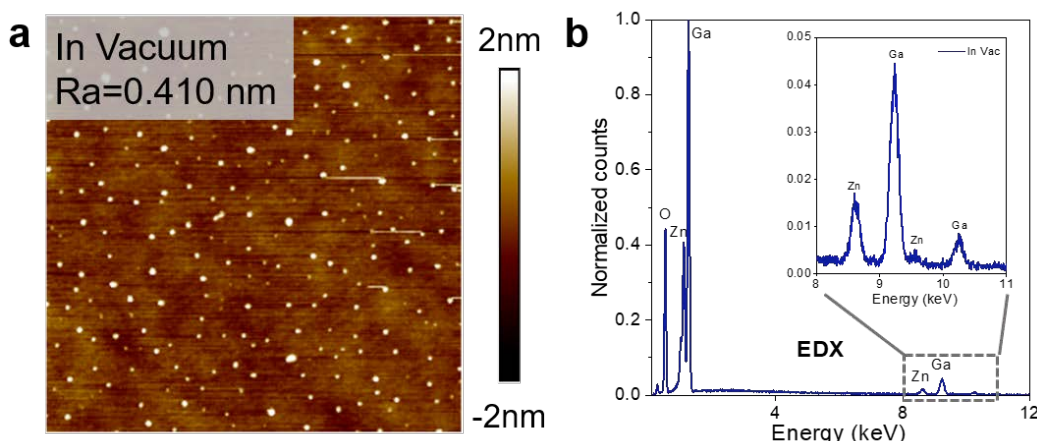
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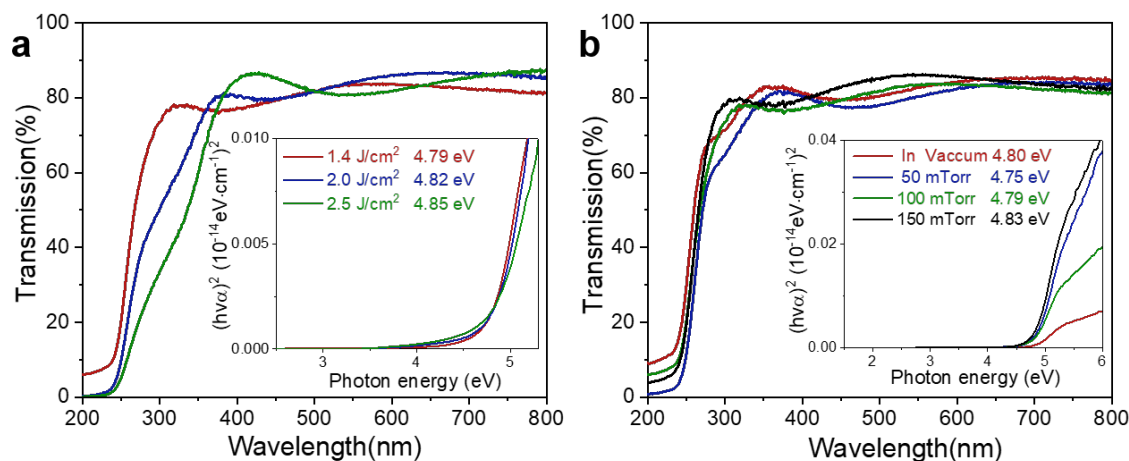
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**Figure S2.** (a) Topology AFM image (scan area of  $2 \times 2 \mu\text{m}^2$ ) of the ZGO films deposited in vacuum ( $\sim 1 \times 10^{-6}$  Pa). (b) EDX spectra image of a Zn-deficient film deposited in vacuum showing  $\text{Zn}/\text{Ga} < 1/2$  due to Zn volatilization during the film growth.



**Figure S3.** Thin film transmission as a function of excitation wavelength for ZGO films with (a) different laser fluence and (b) different oxygen pressure. Inset shows a linear extrapolation of  $E_g$  for ZGO films from their UV-Visible transmission spectra.