Supplementary Information

Enhanced sub-band gap photosensitivity by an asymmetric source-drain electrode low operating voltage oxide transistor

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Structural Properties

Fig. S1 (a) and S1 (b) present the Grazing Incidence X-ray Diffraction (GIXRD) patterns for the thin films of LiInSnO₄ and ZnO, respectively. These patterns were analyzed to determine the crystalline or amorphous phases of the films. The thin films were prepared on cleaned heavily doped p^+ -Si substrates using the same method and conditions employed for Thin-Film Transistor (TFT) fabrication. The GIXRD pattern of the annealed LiInSnO₄ thin film at 550 °C shows an absence of sharp peaks, indicating that it is in an amorphous phase. On the other hand, the GIXRD patterns of the ZnO thin film display intense peaks corresponding to crystal planes (100), (002), (101), (110), (103), and (112) at their respective 2 θ values. These peaks have been matched with the JCPDS number (JCPDS-89-0510), confirming the hexagonal wurtzite structure of the ZnO thin film of LiInSnO₄ in the TFT. Based on these findings, it can be concluded that the dielectric thin film of LiInSnO₄ in the TFT is in an amorphous phase. In contrast, the ZnO thin film exhibits a polycrystalline hexagonal wurtzite structure, indicating its potential for use as an effective active layer in the TFT[2].



Fig.S 1 XRD pattern of a) LiInSnO₄ and b) ZnO thin film

Surface Morphology

Atomic force microscopy (AFM) measurements were utilized to investigate the surface morphology of a gate dielectric thin film made of LiInSnO₄. Fig. S2 (a) and Fig. S2 (b) depict 2-D and 3-D micrographs of the thin film, respectively. The root mean square (RMS) surface roughness of the LiInSnO₄ layer film was determined to be 0.54 nm. This analysis reveals that the dielectric surface exhibits extremely low roughness, indicating the suitability of LiInSnO₄ for producing high-performance TFTs [3]. This is also attributed to its amorphous nature and improved adhesion with the p⁺-Si substrate.



Fig.S 2 AFM image of LiInSnO₄ dielectric thin film (p⁺-Si/LiInSnO₄) (a) 2-D b) 3-D with $R_{rms} \sim 0.54$ nm

Optical Properties

The optical properties of a thin film of LiInSnO₄ gate dielectric were analyzed using a UV-Vis spectrophotometer, and the corresponding results are presented in Fig. S3. The fabrication process for depositing the LiInSnO₄ thin film on a quartz substrate followed the same procedures as those used for manufacturing MIM and TFT devices. Fig. S3 (a) illustrates that the average transmittance of the LiInSnO₄ dielectric thin film is above 85% in the visible range of the electromagnetic spectra (400-800 nm), which is expected for materials with a wide band gap. The high transparency observed in this range suggests minimal scattering from the thin film, indicating a low level of impurities, defects, voids, or roughness on the film surface. This low scattering leads to a reduced leakage current, which is crucial for achieving high-performance gate dielectrics in TFTs. To determine the band gap of the LiInSnO₄ material, a Tauc's plot was generated by plotting $(\alpha h\nu)^{1/2}$ energy versus (hv), as depicted in Fig. S3(b). The absorption coefficient (α) was calculated from the absorbance data, and the band gap was obtained by tangential extrapolation of the curve toward the energy axis (hv). The extracted band gap value for LiInSnO₄ from this Tauc's plot is 5.2 eV, which is reasonably high and makes it suitable for applications as a gate dielectric in TFTs. Additionally, UV-Vis spectroscopic measurements were performed for a ZnO thin film prepared

on a quartz substrate (Fig.S11(a)). The absorbance peak of the ZnO thin film was observed at 360 nm (Fig. S11 (a)). The band gap of ZnO, extracted from Tauc's plot (inset of Fig. S1 (a)), was found to be 3.2 eV. This result indicates that the ZnO thin film can serve as both a semiconductor and an optically sensitive material in TFTs.



Fig.S 3 (a) Optical transmittance spectra of solution proceeds LiInSnO₄dielectric thin film annealed at 550°C for Quartz/LiInSnO₄ (inset) (b) Tauc's plot of LiInSnO₄ and band gap is of 5.2 eV

Electrical Characterization of Dielectric Thin Film

To resolute, the electrical properties of spin coated (solution-processed) LiInSnO₄ thin films, device has been fabricated with a device architecture of metal-insulator-metal (MIM) as shown in Figure 1a). The current-voltage (I-V) characteristics of the device is shown in Fig. S4 a), which shows that the current density of LiInSnO₄ is quite low which resulted from the high compactness and large optical band gap (5.2 eV) of the LiInSnO₄ thin film. As shown in Fig. S4 a), the current density of LiInSnO₄ thin film is ~ 10^{-9} A/cm² under the 2 V, applied voltage, that is two orders lower than the off current of the device as shown in electrical characterization of TFT. Moreover, it is observed that the device remains stable up to applies external bias ~11 V, which aspire that this thin dielectric film can be used securely up to 11 V of operating voltage. The data is replotted in

terms of the leakage current (A/cm²) vs. the electric field (MV/cm), shown in Fig. S4 b), which is indicating that the breakdown field is greater than 1.75 MV/cm, which is reasonably high in comparison with the operating voltage of the TFT. As it is well known, structural flaws like thin film pinholes and non-uniformity rather than crystalline grain boundaries or ionic leakage current (Li⁺) are the most likely causes of leakage current [4]. Therefore, it can be concluded that LiInSnO₄ thin film has the least number of pinholes with a good uniformity that can be an excellent choice for fabricating low operating voltage TFT.

Inclusive of leakage current measurement, the frequency-dependent capacitance (C-f) of LiInSnO4 dielectric thin film has been examined with the same metal-insulator-metal (MIM) device in frequency, ranging from 20 Hz to 1 MHz and is depicted in Fig. S4 c). This data indicates that they areal capacitance of the LiInSnO₄ thin film is 310 nF/cm², at the frequency of 50 Hz, which linearly reduces up to 100 kHz. Here, when the frequency is raised to 10⁵ Hz, a reduction of 14 times the areal capacitance has been seen, which is because the mobile Li⁺ in dielectric is ineffective at promoting ionic polarisation at a higher frequency. It is worth noting that the areal capacitance of LiInSnO₄ dielectric film is extremely high (>310 nF/cm²) at lower frequency suitable ranges. due which it is for low operating voltage TFT 6]. to [5.



Fig.S 4 a) Leakage current vs. applied voltage, b) leakage current vs. applied electric field, and c) capacitance vs. frequency (C-f) curve of LiInSnO₄ dielectric in MIM (inset) structure



Fig.S 5 Cross-sectional SEM of the $p^+\mbox{-}Si/\mbox{LiInSnO}_4/\mbox{ZnO}$



Fig.S 6 a) Output and b) Transfer characteristics of TFT with the symmetric electrode (Interchanging the S and D), c) Output d) Transfer characteristics of TFT with an asymmetric electrode in opposite polarity (D - LiF/Al, S - MoO₃/Ag); Transfer characteristics of e) Device 1 and f) Device 2 with gate leakage (I_G).



Fig.S 7 Variation in the threshold voltage with the intensity of red and blue illumination a) Device 1 b) Device 2



Fig. S 8 The sensitivity vs gate voltage plot of the Device 1 (D1) and Device 2 (D2) under a) red and b) blue illumination at different intensities



Fig.S 9 Variation in the sensitivity of devices 1 and 2 with light intensity under a) red and b) blue at zero gate biasing ($V_G = 0$ V) and under c) red and d) blue illumination at $V_G = 2$ V



Fig.S 10 Variation in the Detectivity of devices 1 and 2 with light intensity under a) red and b) blue at zero gate biasing ($V_G = 0 V$) and under c) red and d) blue illumination at $V_G = 2 V$



Fig.S 11 Absorbance spectra of the a) ZnO, and b) LiInSnO₄/ ZnO thin film fabricated on quartz substrate ; photoluminescence spectra of a) ZnO, b) LiInSnO₄/ZnO and e) LiInSnO₄ thin film



Fig.S 12 Transient response of TFT for single cycle under a) red and b) blue for symmetric S-D electrode and c) red and d) blue for asymmetric S-D electrode devices for the calculation of response time (Rise time (T_{Rise}) and fall time (T_{Fall}))



Fig.S 13 Photoresponse of the photoconductor with a) symmetric (glass/ZnO/Al) and b) asymmetric (glass/ZnO/LiF-Al/ MoO₃-Ag) electrode under red illumination



Fig.S 14 Photo luminance spectra of the a) Red, and b) Blue light source

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