Ultraviolet photoconductive detector based on epitaxial Mg$_{0.34}$Zn$_{0.66}$O thin films

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We report on the fabrication and characterization of visible-blind ultraviolet photodetectors based on Mg$_2$Zn$_{1-x}$O. Using pulsed laser deposition technique, Mg$_{0.34}$Zn$_{0.66}$O thin films with a bandgap of 4.05 eV were epitaxially grown on c-plane sapphire substrates. The structural, electrical, and optical properties of epilayers were characterized using various techniques. Based on the Mg$_{0.34}$Zn$_{0.66}$O films, planar geometry photoconductive type metal–semiconductor–metal photodetectors were fabricated. At a 5 V bias, a high responsivity of 1200 A/W was achieved at 308 nm, and the visible rejection (R308 nm/R400 nm) was more than four orders of magnitude. The 10%–90% rise and fall time were 8 ns and 1.4 μs, respectively. © 2001 American Institute of Physics.

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ZnO is a wide band gap ($E_g = 3.3$ eV) semiconductor that can be used for ultraviolet (UV) photon detection. Its high radiation hardness enables it to be used in harsh environments. The availability of lattice-matched single-crystal ZnO substrates and the relatively low deposition temperatures (100–750 °C) ease the device processing. Owing to its large exciton binding energy (60 meV), ZnO has attracted increasing attention in recent years for potential low-threshold blue/UV lasers that can be integrated with photodetectors. However, the lack of reliable p-type ZnO hinders any p–n junction based optoelectronic devices. Only metal–semiconductor–metal (MSM) structured UV detectors with either Schottky or ohmic contacts were reported.

Mg$_2$Zn$_{1-x}$O, which is realized by alloying MgO with ZnO, exhibits the same material advantages as pure ZnO. By varying the Mg composition, the band gap can be tuned from 3.3 to 7.8 eV for wurtzite and cubic-structured Mg$_2$Zn$_{1-x}$O, extending the cutoff wavelength from UV-A (320–400 nm) to UV-B (280–320 nm) and UV-C (200–280 nm) regions. Such wide range of sensing spectra are expected to enable Mg$_2$Zn$_{1-x}$O UV detectors to be used in many applications such as solar UV radiation monitoring, ultra-high temperature flame detection, and airborne missile warning systems. Nevertheless, UV detectors based on Mg$_2$Zn$_{1-x}$O have not yet been reported, to the best of our knowledge. In this letter, we report on the epitaxial growth of high quality Mg$_2$Zn$_{1-x}$O films on c-plane sapphire by pulsed laser deposition, as well as the fabrication and characterization of photoconductive Mg$_2$Zn$_{1-x}$O UV detectors with MSM structure.

Details of our pulsed laser deposition system can be found elsewhere. In brief, the KrF excimer laser pulses (248 nm, 10 Hz, ~1.7 J/cm$^2$) were focused on a Mg$_{0.34}$Zn$_{0.66}$O target which was mounted in a high vacuum chamber with a base pressure of ~1×10$^{-8}$ Torr. The substrate temperature (750 °C), oxygen pressure (1×10$^{-4}$ Torr) and growth rate (~0.3 Å/pulse) were optimized in the early work. The Mg composition in the film was measured by the energy dispersive spectroscopy, and was found to be Mg$_{0.34}$Zn$_{0.66}$O. The difference between the Mg percentage in the target and in the film was attributed to the high vapor pressure of Zn. Without any intentional doping, the film was n-type with a resistivity of ~5×10$^3$ Ω cm, which was measured by the Seebeck effect and the transmission line method, respectively. The crystalline quality of the films was examined using four-circle x-ray diffraction (XRD) and Rutherford backscattering spectrometry (RBS) with ion channeling technique. Optical properties were evaluated using photoluminescence and ultraviolet–visible (UV–VIS) transmission spectroscopy. To characterize the Mg$_{0.34}$Zn$_{0.66}$O UV detectors, a monochromator (150 W xenon lamp, 1200 lines/mm grating) and a nitrogen gas laser (337.1 nm, ≤4 ns) were used as the excitation sources. The spectral response was measured with a low noise current preamplifier and a lock-in amplifier. A semiconductor parameter analyzer was employed for current–voltage ($I–V$) characterization.

Figure 1 shows the XRD $θ–2θ$, $ω$, and $φ$ scans of Mg$_{0.34}$Zn$_{0.66}$O films grown on Al$_2$O$_3$(0001). The appearance of only (000l) peaks in Fig. 1(a) indicates that the film is highly c-axis oriented normal to the sapphire (0001) plane. The sharp $ω$-rocking curve (FWHM=0.14°) and $φ$ scan confirm the high epitaxial quality. The appearance of the MgZnO(111) peak at $2θ=36.95$°, though orders of magnitude weaker in intensity than that of MgZnO(0002), indicates the onset of phase separation and also the solubility limit of Mg in the wurtzite ZnO lattice. The surface roughness of Mg$_{0.34}$Zn$_{0.66}$O films is rms~1.4 nm, as measured by the atomic force microscopy. The RBS ion channeling studies (1.5 MeV He$^+$) show a minimum yield ($X_{min}$) of ~5%.

Shown in Fig. 2 is the UV–VIS transmission spectrum
of Mg$_{0.34}$Zn$_{0.66}$O thin films. The spectra of Mg$_{0.18}$Zn$_{0.82}$O and ZnO are also presented for comparison. Within the visible region, the average transmittance for all three films is over 90%. ZnO and Mg$_{0.18}$Zn$_{0.82}$O exhibit sharp absorption edges at 377 and 341 nm, respectively, while Mg$_{0.66}$Zn$_{0.34}$O at about 308 nm, which is consistent with the peak wavelength of room temperature photoluminescence. The band gap energy was derived from the plot shown in the inset of Fig. 2. Clearly, the band gap increases with the Mg percent.

The interdigital metal electrodes, which were defined on 250 μm long, 5 μm wide, and have an interelectrode spacing of 5 μm. Under 5 V bias, the measured average dark current is ~40 nA, which is close to the calculated dark current based on the resistivity of Mg$_{0.34}$Zn$_{0.66}$O. The low dark current ($I_{\text{dark}}$) is helpful to enhance the detector’s signal to noise (S/N) ratio since the shot noise, which exceeds the Johnson and 1/f noise if the operating frequency is not too low, is proportional to $I_{\text{dark}}$:

$$\langle i^2 \rangle = 2q [1 + 2(G - 1)^2] (I_{\text{ph}} + I_{\text{dark}}) B,$$

where $q$ is the electron charge, $G$ is the internal gain, $I_{\text{ph}}$ is the photocurrent, and $B$ is the bandwidth. Detailed noise analysis of Mg$_x$Zn$_{1-x}$O UV detectors will be published elsewhere. Upon UV illumination (308 nm 0.1 μW), the photocurrent jumped to 124 μA at 5 V bias, indicating a responsivity of ~1200 A/W. This responsivity value is comparable to that of ZnO (400 A/W at 5 V bias, 2–16 μm interelectrode spacing) and GaN (2000 A/W at 5 V bias, 10 μm interelectrode spacing) photoconductive detectors.

The spectral response of a Mg$_{0.34}$Zn$_{0.66}$O UV detector under front illumination is plotted in Fig. 4. The peak response is found at 308 nm, which is in agreement with the absorption edge shown in Fig. 2. The ~3 dB cutoff wavelength is 317 nm, and the visible rejection (R308 nm/R400 nm) is more than four orders of magnitude, indicating a high degree of visible blindness. For wavelengths beyond the cutoff, the responsivity drops at a rate of ~1.4 dB/nm, followed
by a slower decrease, which may be caused by the Mg$_{0.34}$Zn$_{0.66}$O alloy fluctuation. For wavelengths shorter than 308 nm, the responsivity drops initially followed by an increase at 210 nm due to the surface recombination that may be caused by ion milling. The inset of Fig. 4 is the responsivity as a function of bias voltage. A linear relationship was observed between 0.5 and 5 V, indicating no carrier mobility saturation or sweep-out effect up to the applied bias.

Figure 5 shows the temporal response of a Mg$_{0.34}$Zn$_{0.66}$O UV detector with 3 V bias and 50 Ω load. The 10%–90% rise and fall time are 8 ns and 1.4 μs, respectively. The signal drops to zero at about 30 μs as projected by the dashed line. No persistent photoconductivity was observed. The 8 ns rise time is limited by the excitation laser, which has a nominal pulse duration FWHM ~4 ns. In order to understand the cause of the much longer fall time, it is necessary to consider various physical effects. We fit the decreasing portion of the temporal response with an exponential decay curve, and found a fast decay component with a characteristic time \( \tau_1 = 4.1 \) ns and a slow decay component of \( \tau_2 = 3.9 \) μs. The RC constant may be responsible for the slow decay component. However, the detector capacitance was estimated to be ~1 pF and was confirmed by the actual capacitance–voltage \((C–V)\) measurement. Thus the RC limit is ruled out. The transit time limit can also be ruled out. At a bias of \( V_b = 3 \) V, the transit time \( T_t = s^2/(\mu_p V_b) \) is on the order of ~1 ns, where \( s \) is the interelectrode spacing and \( \mu_p \) is the electron mobility (estimated to be 50 ~100 cm$^2$/Vs).\(^{11}\) The \( T_t \) may contribute to the 8 ns rise time, but has a negligible effect on the fall time. Another factor that affected the temporal response is the excess life-time of trapped carriers, especially the trapped holes in n-type semiconductors. If this is the case, one can expect a large internal gain and a high current responsivity. The latter can be expressed as:\(^{14}\)

\[
R = q\lambda/hc \eta_{in}(1-r) \tau_p (\mu_n + \mu_p) V_b/s^2,
\]

where \( q \) is the electron charge, \( \lambda \) is the light wavelength, \( h \) is the Planck constant, \( \eta_{in} \) is the internal quantum efficiency with a value close to unity, \( \mu_n \) and \( \mu_p \) are electron and hole mobility, respectively, and \( r \) is the reflectivity which can be derived from the refractive index of Mg$_{x}$Zn$_{1-x}$O reported by Teng et al.\(^6\) Assuming the mean lifetime of holes (\( \tau_p \)) is on the same order of magnitude as the 10%–90% fall time, the responsivity was estimated to be \( R \approx 10^3 \) A/W, which is consistent with the measured responsivity shown in Figs. 3 and 4. Further evidence comes from the distribution of hole lifetime. Assuming the trapped holes have a mean lifetime of \( \tau_p = 1.4 \) μs, the plot of hole lifetime distribution \( P(t) = 1/\tau_p \exp(-t/\tau_p) \) fits well with the temporal decay curve. We believe the origin of trap states may relate to the surface damage during the device processing, or the interface states associated with the wurtzite-cubic phase separation and alloy fluctuation.

In summary, we have demonstrated visible blind UV detector based on Mg$_{0.34}$Zn$_{0.66}$O thin films. A high responsivity of 1200 A/W and a fast response of 8 ns rise time, 1.4 μs fall time have been achieved. The detector shows peak responsivity at 308 nm and ~3 dB cutoff at 317 nm. Visible rejection is more than four orders of magnitude.

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