Monolithic Micro Light-Emitting Diode/Metal Oxide Nanowire Gas Sensor with Microwatt-Level Power Consumption

Incheol Cho, Young Chul Sim, Minkyu Cho, Yong-Hoon Cho,* and Inkyu Park*

ABSTRACT: High-performance, monolithic photoactivated gas sensors based on the integration of gas-sensitive semiconductor metal oxide nanowires on micro light-emitting diodes (μLEDs) are introduced. The μLEDs showed improved irradiance and energy conversion efficiency (i.e., external quantum efficiency, EQE), as the size of LEDs was reduced from 200 × 200 μm² (irradiance of 46.5 W/cm² and EQE of 4%) to 30 × 30 μm² (irradiance of 822.4 W/cm² and EQE of 9%). Gas-sensitive zinc oxide (ZnO) nanowires were directly synthesized on top of the μLED through a hydrothermal reaction. The direct contact between the sensing component and μLED sensor platform leads to high light coupling efficiency, minimizing power consumption of the sensor. Furthermore, the sensing performance (i.e., sensitivity) at optimal operating power was improved as the LED size was reduced. The smallest fabricated gas sensor (active area = 30 × 30 μm²) showed excellent NO₂ sensitivity (ΔR/R₀ = 605% to 1 ppm NO₂) at the optimal operating power (∼184 μW). In addition, the sensor showed a low limit of detection (∼14.9 ppb) and robustness to high humidity conditions, which demonstrate its potential for practical applications in mobile internet of things (IoT) devices.

KEYWORDS: gas sensor, micro LED, photoactivation, metal oxide nanowire, low power sensor
NO2 gas sensor directly mounted onto a blue LED. They suggested that monolithic integration can dramatically increase the transfer efficiency of the photon through analytic calculations and experiments. Despite this progress, there still remain many challenges, such as further miniaturization, in-depth investigation of the size dependence of sensing performance, and modulation of the activation wavelength.

Micro LED (μLED), which is commonly defined as the LED with the size less than \(100 \times 100 \mu m^2\), is an emerging technology for future display, light communications, and biomedical applications because of its high luminous efficiency, portability, excellent stability, and fast response. Herein, we report monolithically integrated photoactivated gas sensors combined with μLED platforms (μLP) for the first time. Gallium nitride (GaN)-based μLPs were successfully fabricated via standard microfabrication processes, which promises an industrial-grade throughput. The peak wavelength of μLPs is the near-UV region (\(\lambda_{\text{peak}} = 390 \text{ nm}\)) to match with the absorption band of ZnO nanowires as a sensing material for efficient energy transfer. To investigate the size-dependent sensor performances, LPs with various sizes \([30 \times 30 \text{ (30-μLP)}, 50 \times 50 \text{ (50-μLP)}, 100 \times 100 \text{ (100-μLP)}, \text{and } 200 \times 200 \text{ (200-μLP)}) \mu m^2\) were fabricated. Here, it should be noted that the LPs smaller than \(100 \times 100 \mu m^2\) were particularly named μLPs. An electroluminescence measurement was conducted to compare the irradiance and the energy conversion efficiency of fabricated LPs with different sizes. Then, gas-sensitive ZnO nanowires were directly synthesized on top of the fabricated μLPs through hydrothermal synthesis. Nitrogen dioxide (NO2) gas, which is a representative oxidizing gas, was used to test the sensing performances of fabricated sensors. We could conclude that the size reduction of LP greatly improves the sensor performance and the power efficiency. The μLP-based photoactivated gas sensor achieved ultralow power consumption (~184 μW) with a good sensing performance. In addition, lower limit of detection (LOD) (~15 ppb) and robustness to humidity interference were experimentally demonstrated for practical applications.

## RESULTS AND DISCUSSION

Figure 1a schematically presents the proposed μLP-based photoactivated gas sensor. It has two p–n contact electrodes for operating the LED and two interdigitated or parallel electrodes for measuring the gas sensing signal. The peak emission wavelength of the LP was 390 nm. LPs were designed to have different emitting areas of \(30 \times 30 \text{ (30-μLP)}, 50 \times 50 \text{ (50-μLP)}, 100 \times 100 \text{ (100-μLP)}, \text{and } 200 \times 200 \text{ (200-μLP)} \mu m^2\) to investigate the LED size dependence of power efficiency and sensing performance. Figure 1b describes the cross-sectional layer structure of the monolithic photoactivated sensor based on gallium nitride (GaN) μLP. On top of the p-GaN, 200 nm-thick indium tin oxide (ITO) was used for the Ohmic contact with the p-GaN layer. Silicon dioxide (SiO2) (500 nm thick) was deposited by chemical vapor deposition (CVD) in between the LED and the sensor electrodes for electrical insulation. Hydrothermally grown zinc oxide (ZnO) nanowires were chosen as the sensing material because of their excellent gas diffusivity, photocatalytic property, low electronic band gap, and simple synthesis process. The band gap of ZnO is 3.2 eV at room temperature. Therefore, the emission wavelength appropriate for gas sensing is around 387 nm, which is well matched to the designed LP’s wavelength. LPS were fabricated through well-defined microfabrication processes.

Figure 1c–e shows optical microscopic images of fabricated LPs with widths of 200, 100, and 30 μm, respectively, and Figure 1f shows the illumination from 30-μLP by applying forward bias of 5 V. Figure 1g shows the scanning electron microscopy (SEM) images of ZnO nanowires directly grown on top of the sensing electrodes through hydrothermal reaction. At first, as-synthesized ZnO nanowires covered the whole sensor chip. To define the sensing area, ZnO nanowires on the sensing electrodes were protected by the photoresist mask patterned via the standard photolithography method, and the other exposed nanowires were etched out with 3% nitric acid diluted in deionized (DI) water. As shown in Figure 1g, the average (standard deviation) of diameters and lengths of synthesized ZnO nanowires were 40.5 nm (±6.7 nm) and 349 nm (±114 nm), respectively. The detailed fabrication process is described in the overlap “Experimental Section” and in Figure S1 in the Supporting Information. Also, various SEM images of fabricated devices are provided in Figure S2 in the Supporting Information. As explained above, the combination of sensing nanomaterials and LP was carefully chosen to maximize the coupling efficiency. Figure S3 shows emitting spectrum of the fabricated LP and absorption spectrum of synthesized ZnO nanowires. The absorption band of ZnO nanowires well overlapped the emission spectrum of LP. Here, native defects in ZnO nanowires might contribute to broadening the absorbance spectrum beyond the intrinsic band gap of ZnO. LEDs with much shorter emission wavelength (\(\lambda_{\text{peak}} \ll 390 \text{ nm}\)) may guarantee more definite photoactivation of ZnO nanowires, increasing the coupling efficiency between the LP and the sensing component.
Figure 2. Performance of LPs with different sizes (30-μLP, 50-μLP, 100-LP, and 200-LP): (a) electrical power density depending on the applied forward bias from 3.5 to 6.7 V. (inset: $I−V$ curves) (b) irradiance, and (c) EQE depending on the input electrical power.

However, because the external quantum efficiency (EQE) of UV LEDs drastically decreases with increasing photon energy of UV LEDs, the actual energy efficiency with shorter wavelength of LED ($\lambda_{\text{peak}} \ll 390$ nm) can be lowered. In addition, electrical insulation between the LED and the sensing component can be difficult under irradiation of high energy photons. These technical issues should be solved to utilize UV B or UV C LEDs in the integrated photoactivated gas sensor in the future.

The light-current–voltage ($L−I−V$) measurements were performed to characterize the electrical and the optical properties of fabricated LPs (Figure 2). In Figure 2a, the electrical power densities of various sized LPs (30-μLP, 50-μLP, 100-LP, and 200-LP) were obtained by measuring $L−V$ characteristics (inset graph in Figure 2a) with forward voltage ranging from 3.5 to 6.7 V. The on-voltage measurement was measured around 4 V for all sized LPs. The $L−V$ characteristics showed a strong size-dependent behavior. The overall range of forward current was much lower at the smaller size of μLPs. On the other hand, the electrical power density was substantially increased as decreasing the size of the LP because of improved current spreading in small μLPs. From an equivalent circuit model of a lateral LED, the current density at the n-contact edge can be written as

$$J_n(L) = J_p \exp \left( \frac{L}{L_s} \right)$$

(1)

where $J_n$ and $J_p$ are the current densities near n and p contact edges, respectively, and L is the lateral size of the LED. In case of lateral LEDs, the current is lateral spreading mainly through ITO and n-GaN layers. Therefore, the current spreading length ($L_s$), where the current density drops to 1/e (~0.37), is determined by the resistivity and thickness of each layer as

$$L_s = \sqrt{\frac{(\rho_n + \rho'_p)t_p}{\rho'_n/t_n - \rho_{\text{ITO}}/t_{\text{ITO}}}}$$

(2)

where $\rho_n$ is the contact resistivity, $\rho'_p$, $\rho'_n$, and $\rho_{\text{ITO}}$ are the resistivity, and $t_p$, $t_n$, and $t_{\text{ITO}}$ are the thickness of p-GaN, n-GaN, and ITO layers, respectively. Although achieving perfect current spreading is possible at the critical condition $\rho_n/t_n = \rho_{\text{ITO}}/t_{\text{ITO}}$ (which means that $L_s$ is infinite), the current spreading length is limited in the real situation. In general, carrier recombination mainly occurs near the p-contact, called as “current crowding” because the resistance of n-GaN is lower than that of ITO ($\rho_n/t_n \ll \rho_{\text{ITO}}/t_{\text{ITO}}$), and the carrier mobility of p-GaN is nearly 1 order of magnitude lower than that of n-GaN. As a result, smaller μLPs (30-μLP and 50-μLP) with small L/$L_s$ show more effective current spreading and uniform carrier distribution than larger LPs (100-LP and 200-LP).

The irradiance ($W/m^2$) of various sized LPs was obtained by the electroluminescence measurement. Theoretically, the irradiance is determined by the electrical power density and the energy conversion efficiency of LED as follows

$$E_e \approx M_e = \frac{\Phi_e}{A} = \eta_{\text{EQE}} \frac{P_{\text{electric}}}{A}$$

(3)

where $E_e$ and $M_e$ are the irradiance and the radiant exitance of the LED in the radiometry, respectively, $\Phi_e$ is the total radiant flux, $\eta_{\text{EQE}}$ is the EQE, $P_{\text{electric}}$ is the input electrical power, and A is the emission area of the LED. Because of the close proximity of the light source and the sensing material (~500 nm), the irradiance on the sensing material equals the radiant exitance from the LP. As shown in Figure 2b, the smallest LP (i.e., 30-μLP) showed the highest irradiance (~820 W/cm² at 90 mW), which is nearly 430 times larger than the irradiance of the biggest LP (i.e., 200-LP) (~2.1 W/cm² at the same input power). This large enhancement of irradiance was contributed by the increased EQE and the electrical power density of μLPs. Figure 2c represents the EQE of various sized LPs according to the input electrical power. Overall EQE were increased as decreasing the size of LPs, and the maximum EQE of 30-μLP (~9.1%) was 2.2 times higher than that of the 200-LP (~4.2%). Interestingly, this size-dependent tendency of our UV LED is opposite to those of general visible LEDs. Generally, nonradiative recombination at the sidewall defects induced by dry-etching leads the degraded EQE of visible μLEDs. However, EQE tendency of our UV LED can be explained by the effect of self-heating during LED operation. If the temperature of UV LED increases, carriers (i.e., electrons and holes) can thermally escape from the shallow energy level in the quantum wells to the surface state instead of radiative recombination (a schematic explanation is in Figure S4a in the Supporting Information). The effective current spreading of small μLPs alleviates the current crowding and localized heating. Furthermore, large sidewall area per volume of μLPs leads to the effective heat dissipation and lower junction temperature. Figure S4b in the Supporting Information shows peak wavelength shift under increasing current density. The negligible peak shift of small μLPs (30-μLP and 50-μLP) is the strong evidence of the low junction temperature, while the large red-shift is observed in bigger LPs (100-LP and 200-LP).
LP) because of the shrinkage of band gap energy by the localized heating. The performance of photoactivated gas sensor depends on the irradiance reaching the sensing material. Therefore, based on the improved energy efficiency, smaller μLP can produce similar effect on the gas sensing performance with much lower input power.

Gas sensing tests of fabricated sensors were conducted with nitrogen dioxide (NO₂) gas. NO₂ is the representative outdoor hazardous gas released from chemical plants and automobiles. The short-term recommended exposure limit of NO₂ is 1 ppm by National Institution for Occupational Safety and Health (NIOSH) guidelines. The NO₂ sensing mechanism promoted by UV photoactivation can be explained by the following reactions

\[ \text{O}_2^{(\text{gas})} + e_{(h\nu)}^{-} \rightarrow \text{O}_2^{(ad)^{-}} \]

\[ \text{NO}_2^{(\text{gas})} + e_{(h\nu)}^{-} \rightarrow \text{NO}^{(ad)}^{2-} \]

\[ \text{NO}^{(ad)}^{2-} + \text{O}_2^{(ad)^{-}} + 2e_{(h\nu)}^{-} \rightarrow \text{NO}_2^{(\text{gas})} + 2\text{O}^{(ad)}^{2-} \]

where \( e_{(h\nu)}^{-} \) is the photoelectron generated by UV irradiation. These reaction mechanisms indicate that photoelectrons promote the adsorption of ionized oxygen species (\( \text{O}_2^{(ad)^{-}} \) or the form of \( \text{O}^-/\text{O}^{2-} \) by the spontaneous transition) and dissociation of NO₂ molecules at the surface of metal oxides. Because of surface depletion by electron transfer from metal oxides to surface oxygen, the resistance of n-type ZnO increases under NO₂ exposure. Figure 3a,b shows the responses of monolithic photoactivated gas sensors with different sizes (30-μLP and 200-μLP) to 1 ppm NO₂ gas under increasing LED operating power. The base resistance of both sensors in air decreased as increasing the operating power of LED because of the generation of photoelectrons. The overall resistance of the ZnO nanowires on 30-μLP was higher than that of ZnO nanowires on 200-μLP because the sensor on a smaller area has less electrical paths of ZnO nanowires between sensor electrodes (the gap between electrodes was consistent for all LP; 5 μm). The response of the sensor on 100-μLP in the same experimental condition is presented in Figure S5 in the Supporting Information. We summarized the normalized response \( [(R - R_{\text{air}})/R_{\text{air}} \times 100\%] \) and the recovery ratio \( |\Delta R_{\text{recovery}}/\Delta R_{\text{response}}| \times 100\% \) for 6 ks of recovery for the sensors on 30-μLP, 100-μLP, and 200-μLP, according to the applied LED power in Figure 3c,d. Normalized responses of each sensor basically showed bell-shaped tendencies. Prades et al. provided explanations of this bell-shaped tendency using a single SnO₂ nanowire and an UV LED. They suggested physical models of relationship between the sensor response and irradiated photon flux and found that the optimal photon flux to maximize the sensing response is determined by the balance of surface densities of adsorbates (\( \text{O}_2^- \) and NO₂) on the surface of metal oxides. In dark conditions (\( \Phi_\text{h} \rightarrow 0 \)), inflowing NO₂ molecules cannot replace pre-adsorbed oxygen ions because of the inactive surface (small \( R_{\text{NO}_2} < R_{\text{air}} \)), while adsorbates are totally desorbed in high-flux conditions (\( \Phi_\text{h} \rightarrow \infty \)). In the same manner, our sensor also had optimal LED operating power maximizing the sensor response. Data points were well fitted to the log-normal distribution function

\[ f(x) = \frac{1}{\sigma x \sqrt{2\pi}} \exp\left(\frac{-(\ln x - \mu)^2}{2\sigma^2}\right) \]

where  \( \exp(\mu - \sigma^2) \) of each curve decreased with reducing the size (peak mode = 6800, 1600, and 244 μW at 200-μLP, 100-μLP, and 30-μLP, respectively). Furthermore, they showed improvement of the peak response at the optimal power with the size reduction of LP (334, 436, and 605% for 200-μLP, 100-μLP, and 30-μLP, respectively). These improvements by the size reduction can be explained by the enhanced current spreading of μLPs. From eq 1, \( J_p/J_0 \approx 1 \) is ideally desired for uniform light power density (W/cm²) across the emitting area. However, \( J_p/J_0 \) drops as the lateral length (\( L \)) of the LED increases because of increasing lateral series resistance. It means that poor current spreading induces inhomogeneous light power density across the emitting area. The lower average irradiance of the 200-μLP shown in Figure 2b and optical microscopy images of illuminating LPs in Figure S6 in the Supporting Information support this fact. Therefore, the sensing materials on the less irradiating region (dark spot) can give less contribution to gas sensing, similar with the “cold zone” or “dead zone” of the SMO-based gas sensor on the
heating platforms. Thus, if the portion of these “dead zones” in the overall sensing resistance become larger as increasing the LED size, normalized sensor responses will become lower. Complete recovery of the sensor signal is also important for the repeatability of the gas sensor. Figure 3d shows the recovery ratio, defined as the ratio of resistance change during recovery to response |ΔR_{recovery for 6ks}|/|ΔR_{response for 6ks}| × 100 (%) of the sensors on 30-μLP, 100-LP, and 200-LP. For the tests, response and recovery times were set as 6000 s each. During the response and recovery, the recovery ratio increased as the LED operating power (which is proportional to irradiance) increased. It is because the rate of photo-adsorption and photo-desorption is promoted by sufficient photon flux, and the surface state reaches the equilibrium much faster. In the present state, slow response and recovery may cause limited practical applications. Surface modification of sensing materials with catalytic materials or the heterojunction structure of multiple SMOs can be adopted for improving the response and recovery speeds through future investigation.

We performed further NO₂ gas tests with our smallest sensor device, 30-μLP. Whole tests were conducted under its optimal operating power of ~190 μW: dynamic response to (a) 0.25–2 ppm and (b) 60–250 ppb NO₂ gas. The inset graph shows the noise level of the measurement. (c) Calibration graph of normalized sensor response [(R – R_{air})/R_{air} × 100 (%)] to NO₂ concentrations, and lower LOD is ~14.9 ppb. (d) Humidity interference test under 30 and 60% of relative humidity (RH) and 1 ppm NO₂ gas. (e) Gas selectivity test to 1 ppm NO₂, 160 ppm carbon monoxide (CO), 3.2% hydrogen (H₂), and 10 ppm hydrogen sulfide (H₂S).

Figure 4. Nitrogen dioxide (NO₂) sensing performance of the photoactivated gas sensor on 30-μLP. Whole tests were conducted under an operating power of ~190 μW: dynamic response to (a) 0.25–2 ppm and (b) 60–250 ppb NO₂ gas. The inset graph shows the noise level of the measurement. (c) Calibration graph of normalized sensor response [(R – R_{air})/R_{air} × 100 (%)] to NO₂ concentrations, and lower LOD is ~14.9 ppb. (d) Humidity interference test under 30 and 60% of relative humidity (RH) and 1 ppm NO₂ gas. (e) Gas selectivity test to 1 ppm NO₂, 160 ppm carbon monoxide (CO), 3.2% hydrogen (H₂), and 10 ppm hydrogen sulfide (H₂S).
reduction of responses by the humidity effect, the compensation of the sensor signal from humidity interference will be possible with real-time humidity monitoring, because the sensor signal to NO\textsubscript{2} gas is distinct even under the humid condition.

Figure 4e shows the response \(\frac{\Delta R}{R_{\text{air}}} \times 100\%\) of the sensor for different types of gases such as carbon monoxide (CO), hydrogen (H\textsubscript{2}), and hydrogen sulfide (H\textsubscript{2}S). These reducing gases provide electrons to SMOs when they adsorb on the surface of SMOs. Therefore, they make the resistance of n-type ZnO decrease. The concentrations of each tested gas were the maximum concentration that our experimental setup could generate (160, 3.2\%, and 10 ppm of CO, H\textsubscript{2}, and H\textsubscript{2}S respectively), and all are close to the permissible exposure limit or lower explosive limit by the safety guideline.\textsuperscript{40} As shown in Figure 4e, the sensor showed the greatest response to the NO\textsubscript{2} gas \(\Delta R/R_{\text{air}} \times 100\% = 388\%\). Other reducing gases showed reverse response \(\Delta R/R_{\text{air}} < 0\) and relatively smaller responses \(\Delta R/R_{\text{air}} = -7.2, -16.6,\) and -42\% for 160 ppm CO, 3.2\% H\textsubscript{2}, and 10 ppm H\textsubscript{2}S respectively. Therefore, the developed sensor can be utilized for selective detection of NO\textsubscript{2} gas.

\section*{CONCLUSIONS}

In conclusion, an ultra-low power photoactivated gas sensor, monolithically integrated onto a miniaturized \(\mu\)LP was introduced. The 30 \(\mu\)m-sized \(\mu\)LED platform (30-\(\mu\)LP) achieved the superior energy efficiency because of proximity (\(\sim 500\) nm) with sensing materials and the intrinsic high irradiance and the EQE (\(\sim 9\%\)) of \(\mu\)LPS. High irradiance and EQE of the \(\mu\)LP was mainly due to the improved current spreading that can suppress the undesired current crowding near the p-contact. Furthermore, ZnO nanowires integrated on 30-\(\mu\)LP showed superior sensing performance \(\Delta R/R_{\text{air}} = 605\%\) to 1 ppm NO\textsubscript{2} compared to the sensor on 200-\(\mu\)LP \(\Delta R/R_{\text{air}} = 334\%\) to 1 ppm NO\textsubscript{2} because effective current spreading of \(\mu\)LP allowed uniform emission across the emitting area. The sensor on 30-\(\mu\)LP showed ultralow LOD (\(\sim 15\) ppb NO\textsubscript{2}), humidity robustness, and high NO\textsubscript{2} selectivity to other interfering gases under low operating power (\(\sim 190\) \(\mu\)W). All sensing tests were conducted at room temperature so that the sensor can avoid undesired degradation because of thermal aging. Excellent gas sensing performance and ultralow operating power promise applications on the battery-driven IoT devices. Finally, this work provides a new route of emerging \(\mu\)LED technology as chemical sensors for future healthcare, personalized environmental monitoring, and smart factory applications.

\section*{EXPERIMENTAL SECTION}

\subsection*{Fabrication of LPs.}

The whole fabrication process is graphically described in Figure S1 in the Supporting Information. A stack of high-purity n-type (3 \(\mu\)m) GaN, InGaN/GaN multi-quantum well (MQW) layer (50 nm), and p-type GaN (200 nm) layer was epitaxially grown on a sapphire substrate through a metal–organic CVD (conducted by GenUV, South Korea). GaN layers were patterned with photoresist (AZnelof 2035, MicroChemicals, Germany) and vertically etched via inductively coupled plasma-reactive ion etching (ICP-RIE) for 2 min to formate mesa structures of LPs. ICP-RIE was conducted in the Cl \(200 \text{ sccm}\) and Ar (20 \text{sccm}) atmosphere, maintaining the 10 mtorr pressure with a radio frequency power of 250 \(\text{W}\) (plasma power = 700 \(\text{W}\)). Here, the widths of square LEDs were designed to 30, 50, 100, and 200 \(\mu\)m. On top of the mesa structure, 200 nm thick ITO and p–n contact metals (10 nm chromium (Cr) and 200 nm gold (Au)) were sequentially patterned by UV photolithography and electron-beam (e-beam) evaporation. Here, the ITO layer roles as a current spreading layer. As-deposited ITO was heat-treated at 750 \(\text{°C}\) through rapid thermal annealing process for the enhanced optical transparency and electrical conductivity. Silicon dioxide (SiO\textsubscript{2}) insulation layer (500 nm thick) was deposited using plasma-enhanced CVD process and vertically etched via RIE (\(\text{CF}_2/\text{O}_2 = 9:1, 150\) mtorr, 50 \text{W}) for opening contact pads. Finally, sensor electrodes (10 nm Cr/200 nm Au) were patterned on an insulation layer by UV photolithography and e-beam evaporation.

\subsection*{Growth of Zinc Oxide Nanowires on the Fabricated LPs.}

Zinc oxide (ZnO) nanowires were synthesized by the well-established hydrothermal reaction.\textsuperscript{49–52} Prior to the synthesis, a thin layer of ZnO nanoparticles was deposited using the sputtering system (5 mtorr argon atmosphere, 150 \text{ W}, 3 min) on fabricated LPs. A precursor solution was prepared by mixing 25 \(\text{mM}\) zinc nitrate hexahydrate, 25 \(\text{mM}\) hexamethylenetetramine, and 6 \(\text{mM}\) polyethyleneimine in DI water. Prefabricated LPs were immersed into the precursor solution and heated up to 95 \(\text{°C}\) for 2 h in a convection oven. Then, ZnO nanowires grew on the whole surface of the device. In order to define the sensing area, ZnO nanowires grown on the sensor electrodes were covered with a 3 \(\mu\)m thick photoresistor (AZ5214, MicroChemicals, Germany). Exposed ZnO nanowires out of the sensing area were removed in an acidic etchant (3% nitric acid diluted in DI water) for 5 s. Finally, photoresist masks were removed by acetone, and the device was washed with DI water.

\subsection*{Characterization of the Fabricated Devices.}

The fabricated sensor devices were visually observed by field emission scanning electron microscopy [FE-SEM (SU8230, Hitachi, Japan)]. In order to analyze the photo absorbance of ZnO nanowires, they were synthesized on a transparent quartz substrate, and an absorbance spectrum was measured using the ultraviolet–visible (UV–vis) spectrophotometer (Lambda 1050, PerkinElmer, USA) with an integrating sphere. Optical and electrical properties of LPs were obtained using the \(L–I–V\) measurement system (OPI 160, WITHLIGHT, Korea) with an integrating sphere and a source meter (Keithley 2400, USA).

\subsection*{Gas Sensing Test.}

The sensor devices were mounted on a custom-made testing chamber and connected to current sourcemeters (Keithley 2400 and 2635b, USA) for applying forward bias on a \(\mu\)LPs and measuring the sensing resistance, respectively. Forward bias applied on the LP was swept from 3.2 to 6 \(\text{V}\) for each gas test to find the optimal operating power. Gas was supplied to the sensor device, and the concentration of NO\textsubscript{2} gas was adjusted by controlling the flow rates of NO\textsubscript{2} gas and dry air (that is, \(\sim 0\%)\) with mass flow controllers. For the humidity interference test, RH was controlled to 30 and 60 \(\%\) by changing the proportion of the dry air and the humid air generated by a water-bubbling system. Carbon monoxide (CO), hydrogen (H\textsubscript{2}), and hydrogen sulfide (H\textsubscript{2}S), which are indoor/outdoor hazardous gases, were utilized for the gas selectivity test.

\section*{ASSOCIATED CONTENT}

\subsection*{Supporting Information}

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssensors.9b02487.

Detailed fabrication process, SEM images of fabricated devices, electroluminescence spectrum of fabricated LED, absorbance spectrum of hydrothermally grown zinc oxide nanowires, simplified band model of MQWs in the GaN-LED, peak wavelength shift of LEDs, response of the gas sensor on the 100-\(\mu\)LP, response of emission uniformity according to the size of LED, and summary of gas sensors based on various activation sources (PDF)
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**Author Contributions**
I.C., Y.C.S., and M.C. have contributed equally. I.C. and Y.C.S. planned and conducted the experiments and wrote the paper. M.C. conceived the original ideas. Y.-H.C. and I.P. supervised the project. All authors have given approval to the final version of the manuscript.

**Notes**
The authors declare no competing financial interest.

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