Supplementary Information

Fourier transform infrared spectroscopy:

The FTIR spectra of GaO(OH) nanoparticles is shown in Fig.S1. The bands at 3411 cm⁻¹ and 1630 cm⁻¹ reveal broad H-O-H stretching and an OH bending vibration of water respectively.^{1,2} The presence of infrared bands at 2426 cm⁻¹, 2397 cm⁻¹ and 1384 cm⁻¹ are due to the atmospheric CO₂ absorption.^{1,3} The two bands observed at 3138 cm⁻¹ and 2780 cm⁻¹ were assigned to water molecules bonded with the GaO(OH) hydroxyl units and OH stretching vibration of the GaO(OH) units³ respectively. The bands corresponding to the Ga-O stretching modes were observed at 825 cm⁻¹, 660 cm⁻¹ and 484 cm⁻¹.¹⁻³ In addition to Ga-O mode, the bands at 1763 cm⁻¹ and 2065-2092 cm⁻¹, together with bands at 1018 cm⁻¹ and 920 cm⁻¹ are attributed to the Ga-OH bending modes.¹⁻³ However, the peak position of Ga-OH deformation modes and Ga-O stretching modes were sensitive to pH and thermal treatment of the materials.³



Fig.S1. FTIR spectra of GaO(OH) nanoparticles in the range (a) 400-4000 cm⁻¹, (b) and (c) shows the focused view of peak position in the range 2450-1730 cm⁻¹ and 1035-835 cm⁻¹ respectively.

Raman line shape analysis:

The Raman longitudinal optical (LO) phonon-plasmon coupled mode (LOPC) can be analyzed by fitting the line shape analysis based on a semi-classical model, which assuming the deformation potential (DP) and electro-optical (EO) mechanisms.^{4,7} The Raman intensity of the LOPC can be expressed as,⁵

$$I_A = S.A(\omega).Im[-\epsilon(\omega)^{-1}]$$
 ------(1)

where ω represents the Raman shift, S is proportionality constant and $\varepsilon(\omega)$ is the dielectric function. The parameter A(ω) that corresponds to the deformation potential and electro-optical mechanism is given by the following expression,⁴

$$\begin{aligned} \mathbf{A}(\omega) &= 1 + 2C\omega_{T}^{2} \left[\omega_{p}^{2} \gamma(\omega_{T}^{2} - \omega_{L+}^{2}) - \omega_{L+}^{2} \Gamma(\omega_{L+}^{2} + \gamma^{2} - \omega_{p}^{2}) \right] / \Delta \\ &+ (C^{2} \omega_{T}^{4} / \Delta) \left\{ \omega_{p}^{2} \left[\gamma(\omega_{L}^{2} - \omega_{T}^{2}) + \Gamma(\omega_{p}^{2} - 2 \omega_{L+}^{2}) \right] \right. \\ &+ \left. \omega_{L+}^{2} \Gamma(\omega_{L+}^{2} + \gamma^{2}) / \left(\omega_{L}^{2} - \omega_{T}^{2} \right) \right. \end{aligned}$$

where Δ is given by,

$$\Delta = \omega_{p}^{2} \gamma [(\omega_{T}^{2} - \omega_{L+}^{2})^{2} + (\omega_{L+} \Gamma)^{2}] + \omega_{L+}^{2} \Gamma (\omega_{L}^{2} - \omega_{T}^{2}) (\omega_{L+}^{2} \gamma^{2})] \qquad -----(3)$$

where, ω_T and ω_L are the frequencies of TO and LO phonons respectively, ω_p is the plasmon frequency, γ_p is the plasmon damping constant, Γ is the phonon damping constant, C is the Faust-Henry coefficient, ω_p is the Plasmon frequency and and ω_{L^+} is the frequency of coupled mode. The Faust-Henry coefficient can be deduced from the ratio of the intensity of the LO and TO phonon modes in undoped GaN⁵ using the following equation:

$$I_{\rm LO} / I_{\rm TO} = (\omega_{\rm l} + \omega_{\rm LO} / \omega_{\rm l} + \omega_{\rm TO})^4 + \omega_{\rm TO} / \omega_{\rm LO} \{1 + \omega_{\rm TO}^2 - \omega_{\rm LO}^2 / C \omega_{\rm TO}^2\} \quad ------(4)$$

where ω_l is the frequency of the laser. The dielectric function $\epsilon(\omega)$ is given by a sum of the contribution from both phonon and plasmons as follows,

$$\varepsilon(\omega) = \varepsilon_{\infty} \left[1 + \omega^2_L - \omega^2_T \right) / \left(\omega^2_T - \omega^2_{L^+} - i\omega\Gamma \right) - \omega^2_p / \left(\omega^2_{L^+} + i\omega_L \gamma \right) \right] \quad ------(5)$$

where ε_{∞} is the high frequency optical dielectric constant. The line shape of the coupled mode L+ can be fitted with the function given in Eqn.(1) which can be fully expressed and simplified by substituting Eqns. (2), (3) and (5) into it. The line shape of the coupled A₁(LO) mode can be fitted with the values of A₁(TO) = 533cm⁻¹, A₁(LO) = 735cm⁻¹, dielectric constant ε_{∞} = 5.35 and the Faust – Henry coefficient C = 0.48 as a constants. The value of plasmon frequency (ω_p), plasmon damping constant (γ) and phonon damping constant (Γ) can be obtained by fitting the experimental data with Eqn. (1). The free carrier concentration can be determined from the Plasmon frequency (ω_p) by using the following relation: ^{6,7}

$$(\omega_{\rm p})^2 = \pi n e^2 / \epsilon_{\infty} m^*$$
 ------(6)

where m* is the effective mass of the electron. The mobility of the free carriers can be obtained from the plasmon damping constant (γ) as follows: ^{6,7}

Hall measurement – Ohmic behaviour of the contact:

In Hall measurement, the output voltage is linearly increased with the current applied to the four indium contacts made on the periphery of the sample. This relationship gives the perfect linear I-V curve in a graphical representation, so called Ohmic behavior of the contacts. Figure S2 illustrates linear I-V curve for the contacts.



Fig.S2.Ohmic behavior of metal contacts

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