Polar Semiconducting Scandium Nitride as an Infrared Plasmon and Phonon–Polaritonic Material

Krishna Chand Maurya, Dheemahi Rao, Shashidhara Acharya, Pavithra Rao, Ashalatha Indiradevi Kamalasanan Pillai, Shankar Kumar Selvaraja, Magnus Garbrecht, and Bivas Saha*

ABSTRACT: The interaction of light with collective charge oscillations, called plasmon–polariton, and with polar lattice vibrations, called phonon–polariton, are essential for confining light at deep subwavelength dimensions and achieving strong resonances. Traditionally, doped-semiconductors and conducting metal oxides (CMO) are used to achieve plasmon–polaritons in the near-to-mid infrared (IR), while polar dielectrics are utilized for realizing phonon–polaritons in the long-wavelength IR (LWIR) spectral regions. However, demonstrating low-loss plasmon– and phonon–polaritons in one host material will make it attractive for practical applications. Here, we demonstrate high-quality tunable short-wavelength IR (SWIR) plasmon–polariton and LWIR phonon–polariton in complementary metal-oxide-semiconductor compatible group III–V polar semiconducting scandium nitride (ScN) thin films. We achieve both resonances by utilizing n-type (oxygen) and p-type (magnesium) doping in ScN that allows modulation of carrier concentration from $5 \times 10^{18}$ to $1.6 \times 10^{21}$ cm$^{-3}$. Our work enables infrared nanophotonics with an epitaxial group III semiconductor nitride, opening the possibility for practical applications.

KEYWORDS: plasmon polariton, phonon polariton, Reststrahlen band, alternative plasmonic materials, infrared plasmonics
could be quite challenging for many well-established dentistry, are proposed. 24,27 So far, the excitation and applications, such as passive radiative cooling, biomolecular coupling mechanism in the LWIR range and several other polariton (SPhP) has emerged as a promising light-matter conduction band edges, which could drastically change carrier Additionally, the dopant states should not alter the valence or moderately high mobility at the same time. To achieve this, dielectric permittivity (εω) and if the carrier concentration can be tuned from as low as 1018 to 1021 cm−3, while retaining a moderately high mobility that gives rise to tunable high-quality low-loss SWIR plasmon— and LWIR high-quality phonon—polariton resonance.

Achieving phonon—polaritons, on the other hand, requires polar dielectric materials, where macroscopic electric field stiffens the force constant of the longitudinal-optical (LO) phonons and splits the LO and transverse-optical (TO) phonon modes at the zone center. 25 Within the frequency range bounded by the LO and TO, termed as the Reststrahlen band, 26 the real component of the dielectric permittivity (ε1) becomes negative, and electromagnetic modes couple to the TO phonon giving rise to its evanescent character and confinement at the surface. Such confined surface-phonon—polariton (SPhP) has emerged as a promising light-matter coupling mechanism in the LWIR range and several other applications, such as passive radiative cooling, biomolecular fingerprinting, and diagnosis tools for cancer detection and dentistry, are proposed. 24,27 So far, the excitation and engineering of SPhP are performed with well-established dielectric materials, such as 4H-SiC, 28 h-BN, 29,30 or AlN, 31 with large bandgaps and low carrier densities. Active tuning of the SPhP mode frequency is also demonstrated with carrier injection in polar dielectrics, such as in InP, 4H-SiC, etc. 32,33

The demonstration of low-loss and high-quality plasmon— and phonon—polaritons in one host material will make it attractive for practical device implementations. Conceptually, achieving plasmon— and phonon—polariton in one host material is possible if the electronic and phonon resonances are spectrally separated from each other (see eq 1 for the total dielectric permittivity) and if the carrier concentration can be tuned from as low as 1018 to 1021 cm−3, while retaining a moderately high mobility at the same time. To achieve this, dopants should not introduce defect states inside the bandgap of semiconductors that otherwise could pin the Fermi level. Additionally, the dopant states should not alter the valence or conduction band edges, which could drastically change carrier effective mass and mobility. Satisfying all of the conditions could be quite challenging for many well-established semiconductors.

e_{\text{total}}(ω) = e_1 + ie_2

= e_\infty \left(1 - \frac{\omega_p^2}{\omega^2 - i\gamma \omega} + \frac{\omega_{\text{LO}}^2 - \omega_{\text{TO}}^2}{\omega_{\text{TO}}^2 - \omega^2 - i\omega\Gamma}\right)

(1)

ωp = \sqrt{\frac{ne^2}{m^*\varepsilon_0}}

(2)

The wavelength (λp) corresponding to the plasmon— and phonon—polariton resonance frequency are listed.

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Table 1. Carrier Concentration, Mobility, and Resistivity of ScN Thin Films

<table>
<thead>
<tr>
<th>ScN films</th>
<th>carrier concentration (cm−3)</th>
<th>mobility (cm² V−1 s−1)</th>
<th>resistivity (Ωcm)</th>
<th>crossover wavelength λp (µm)</th>
<th>carrier and doping type</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>1.6 × 1018</td>
<td>29</td>
<td>1.4 × 10−4</td>
<td>1.83</td>
<td>n-type (oxygen-doped)</td>
</tr>
<tr>
<td>(b)</td>
<td>1.4 × 1018</td>
<td>6</td>
<td>7.6 × 10−4</td>
<td>2.08</td>
<td>n-type (unintentionally oxygen-doped)</td>
</tr>
<tr>
<td>(c)</td>
<td>7.7 × 1018</td>
<td>22</td>
<td>3.8 × 10−4</td>
<td>2.25</td>
<td>p-type (magnesium-doped)</td>
</tr>
<tr>
<td>(d)</td>
<td>3.3 × 1018</td>
<td>43</td>
<td>4.4 × 10−4</td>
<td>2.35</td>
<td>n-type (oxygen-doped)</td>
</tr>
<tr>
<td>(e)</td>
<td>4.3 × 1019</td>
<td>13</td>
<td>1.0 × 10−2</td>
<td>14.58 (LO)</td>
<td>m-type (oxygen-doped)</td>
</tr>
<tr>
<td>(f)</td>
<td>5.0 × 1018</td>
<td>10</td>
<td>1.3 × 10−1</td>
<td>14.58 (LO)</td>
<td>m-type (oxygen-doped)</td>
</tr>
</tbody>
</table>

The wavelength (λp) corresponding to the plasmon— and phonon—polariton resonance frequency are listed.

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ε_{\text{plasmon}}(ω) = -\frac{\omega_p^2}{\omega^2 - i\gamma \omega}

ε_{\text{total}}(ω) is the total dielectric permittivity 24 with contributions from the plasmon or Drude component (second term) and phonon (third term) resonances. ε_{\text{total}}(ω) = ε_{\infty} + \omega_p^2/\omega^2 - i\gamma \omega + \omega_{\text{LO}}^2 - \omega_{\text{TO}}^2/\omega_{\text{TO}}^2 - \omega^2 - i\omega\Gamma

(3)
from the phonon–polariton resonance. Therefore, high-mobility and tunable carrier concentration in ScN provide a perfect testbed to achieve high-quality plasmon–phonon–polariton in one host medium.

ScN thin films with carrier concentration ranging from $1.6 \times 10^{21}$ cm$^{-3}$ to $5 \times 10^{18}$ cm$^{-3}$ (see Table 1) are deposited inside an ultrahigh vacuum (UHV) chamber at a base pressure of $(2-4) \times 10^{-9}$ Torr (see Supporting Information, section 1). Without any intentional doping, n-type ScN films exhibit a carrier concentration of $3.6 \times 10^{20}$ cm$^{-3}$ with a mobility of 43 cm$^2$/V s. Intentional oxygen-doping increase its n-type carrier concentration up to $1.6 \times 10^{21}$ cm$^{-3}$, while Mg-hole doping results in p-type films with carrier concentration as low as $5 \times 10^{18}$ cm$^{-3}$. Though the intentional doping reduces the mobility slightly, it remains sufficiently high (see Table 1) for achieving low-loss resonances. With increasing n-type doping concentration, the Fermi level in ScN moves higher up inside the conduction band ($\sim 0.4$ eV from conduction band minima).
for the n-type doping concentration of $1.6 \times 10^{21}$ cm$^{-3}$.$^{41,48}$ Similarly, hole-doping shifts the Fermi level close to the valence band. A detailed discussion about the band structure, electron and hole effective mass, average carrier scattering time, and position of the Fermi level as a function of doping type and concentration are presented in the Supporting Information section 2.

The $\varepsilon_r$ of ScN (measured with a spectroscopic ellipsometer, see Figure 1a and Supporting Information for detail) with the highest carrier concentration of $1.6 \times 10^{21}$ cm$^{-3}$ exhibits epsilon near zero (ENZ) (a positive-to-negative crossover) at 1.83 $\mu$m (see Figure 1b). At longer wavelengths, $|\varepsilon_r|$ increases monotonically because of the increasing metallic response. The optical loss, characterized by the imaginary component of the permittivity $\varepsilon_r$, increases too, but it is still monotonically. The $\varepsilon_r$ of ENZ at 1.83 $\mu$m (see Figure S3) corresponds to the direct bandgap $\varepsilon_2$ of visible wavelength plasmonic materials, such as Au,$^{49}$ TiN,$^{50}$ etc., at their respective $\lambda_p$. Below 1.83 $\mu$m, ScN acts as a dielectric medium with positive $\varepsilon_r$ and a peak in $\varepsilon_3$ near $\sim\lambda_0$ nm (see Figure 1b) is representative of the plasmonic nature. Angle-dependent reflectivity measurements (see Figure 1c) clearly show the Brewster’s angle in the p-polarized/s-polarized reflection curve. The calculated reflectance spectrum (see Figure 1d) utilizing the permittivity of ScN in the Fresnel’s equation (see Supporting Information section 6) matches well with the measured reflectivity which highlights consistency between the experiment and ellipsometry data fitting.

Excitation of the surface plasmon–polaritons (SPP) is demonstrated (see Figure 1e) with polarization-dependent reflectivity measurement in the attenuated-total-reflection (ATR) configuration inside an FTIR-spectrometer in the Kretschmann configuration at 45° angle-of-incidence. Diamond is used as a high refractive index medium and to provide the additional momentum for the light coupling to the SPP modes. A clear dip in the p-/s-polarized reflection spectrum at $\sim1.95 \mu$m (see Figure 1f) with a full-width-at-the-half-maxima (fwhm) of $\sim0.27 \mu$m demonstrate the coupling of energy from the incident radiation to the SPP mode. The SPP dispersion is calculated (see Figure 1f) taking into account the measured dielectric permittivity of ScN that show close matching of the $\lambda_{SP}$ (wavelength corresponding to the SPP mode frequency) with the experimentally measured dip in reflectivity curve.

While the above analysis unambiguously demonstrates SWIR plasmon excitation in ScN, spectral position of the plasmon-resonance and the SPP mode frequencies are varied by altering the carrier concentration (see eq 2) through doping control. With a decrease in the carrier concentration from $1.6 \times 10^{21}$ cm$^{-3}$ (a) to $1.4 \times 10^{21}$ cm$^{-3}$ (b), $7.7 \times 10^{20}$ cm$^{-3}$ (c), and $3.3 \times 10^{20}$ cm$^{-3}$ (d), ellipsometry measurements show that the $\lambda_p$ shifts from 1.83 to 2.08, 2.25, and 2.35 $\mu$m, respectively (see Figure 2a). Such monotonic red-shift in the $\lambda_p$ is consistent with the predictions from the Drude model (see eq 3) of dielectric permittivity, and cover a wide SWIR range. In the Figure 2b, $\varepsilon_2$ of ScN with $3.3 \times 10^{20}$ cm$^{-3}$ carrier densities show the lowest value, primarily because of its higher mobility of 43 cm$^2/(V \cdot s)$. Since the highest mobility and the lowest optical loss is obtained in the as-deposited ScN without any intentional doping, $\lambda_p$ of 2.35 $\mu$m with the lowest optical loss of 1.0 could be regarded as the baseline plasmon response in ScN. Dielectric permittivity at the longer wavelength regions (1.5–5 $\mu$m) is measured further with an IR-ellipsometer (see Figure S2) which demonstrates ScN’s metallic response in the SWIR-to-mid-IR spectral region. Similar to the tunable bulk plasmon frequency, tunability of the SPP mode frequencies are further demonstrated by the polarization-dependent ATR measurements, which show a dip at 2.70 $\mu$m with a fwhm of 0.56 $\mu$m for ScN with $3.3 \times 10^{20}$ cm$^{-3}$ (d) carrier concentrations (see Figure 2c).

The plasmonic response is further characterized by temperature-dependent Hall measurements (see Figure 2d). The resistivity of all ScN films increases slightly with an increase in temperature (see Figure 2e), which is representative of their degenerate semiconducting or semimetallic nature due to high carrier concentrations. On the other hand, mobility decreases with an increase in temperature as found in Figure 2f. A combination of ionized impurity and dislocation scattering model is found to fit the temperature-dependence of mobility very well with a high dislocation density in the $10^3$–$10^4$ cm$^{-2}$ range (see Figure S5), which can be seen as well in transmission electron microscopy (TEM) images. The carrier concentration of the ScN films remain nearly unchanged within the measured temperature range (see Figure 2g).

Temperature-dependent dielectric permittivity is measured further to highlight the refractory plasmonic behavior of ScN (see Supporting Information section 7 for detail). Results show that, with an increase in temperature from 100 to 500 K, $\lambda_p$ exhibits a redshift from 1.76 to 1.83 $\mu$m (see Figure S4a), and $\varepsilon_2$ increases from 0.95 to 1.32 at the corresponding $\lambda_p$ for the ScN film with $1.6 \times 10^{21}$ cm$^{-3}$ carrier concentrations. Such an increase in $\lambda_p$ and optical losses, especially at longer wavelengths (see Figure S4b) can be directly attributed to the decrease in mobility as shown in the Figure S4c. However, near the $\lambda_p$ wavelength region, the increase in $\varepsilon_2$ is rather small that highlight the suitability of ScN for high-temperature applications. Note that, though the permittivity is measured until 500 K because of instrumental limitations, ScN exhibits a high melting temperature of $\sim2600$ °C and, hence, could be useful for many plasmonic applications at high-temperatures.

With the above refractory plasmonic properties in the SWIR spectral range, ScN’s suitability for various nanophotonic applications,$^{15,52}$ such as in SPP waveguides, localized surface-plasmon-resonance (LSPR), epsilon-near-zero (ENZ), hyperbolic metamaterials (HMM), and transformation optics, are determined. Each of these applications requires its own optimum operating conditions that are determined by the structure and geometry of devices, as well as material properties. Our analysis show that the plasmon-resonance and SPP in ScN should be useful for the nonresonant SWIR applications, such as waveguides, ENZ, and HMM$^{15,52}$ (see Figure S6). As an example, ScN exhibits a high SPP propagation length ($L$) and low electric-field confinement length ($D$) that is comparable to other doped-semiconductors and CMOs for high-performance waveguides. A ratio between the $L$ and $D$, referred as the figure-of-merit (FOM) of ScN is high in ScN and compares well to its alternatives in the near and mid-IR spectral range. Similarly, a low $\varepsilon_2$ of 1.00 at $\lambda_p$ should be suitable for ENZ device applications,$^{54}$ such as photon funnels or spatial filtering for beaming. Even for the localized SPP resonance, the figure-of-merit (FOM) of ScN is comparable to its counterparts. Nevertheless, with more advanced deposition methods, such as molecular-beam-epitaxy (MBE) and hybrid vapor phase epitaxy, mobility of ScN could be increased further which should improve its performance metrics.
While the carrier concentration control leads to the SWIR plasmonic response in ScN, demonstration of SPhP excitation requires that the electronic resonance do not contribute to the total dielectric permittivity in the LWIR spectral range. To achieve this condition, Mg (hole)-doped ScN films with low carrier concentrations are deposited (see Table 1). To separate the contributions of MgO (substrate) phonon modes, a 100 nm IR reflective TiN buffer layer is deposited on (001) MgO substrates before ScN depositions (see Figure S7). Infrared reflectivity measurement (see Figure 3a) of ScN with $5 \times 10^{18}$ cm$^{-3}$ carrier concentration shows well-defined Reststrahlen band (see Figure 3b), a highly reflective region between the TO (359 cm$^{-1}$) and LO (686 cm$^{-1}$) phonon modes, where light couples with the polar optical lattice vibrations. Calculated $\varepsilon_1$ is found to exhibit negative values within the Reststrahlen band, with an epsilon-near-pole (ENP) resonance at 359 cm$^{-1}$ since light couples directly to the TO phonon mode (see Figure 3c). Concomitantly, $\varepsilon_2$ exhibits a sharp peak at the TO phonon frequency. Both LO and TO phonon frequencies are consistent with recent inelastic X-ray scattering (IXS) phonon dispersion of ScN. IXS measurement has also shown a phonon lifetime of 0.21 ps for the LO phonon mode at the $\Gamma$-point that is dominated primarily by three-body phonon–phonon interactions. Since the crystalline quality of ScN is very high, optical losses in the SPhP resonance should primarily arise from the phonon–phonon interactions (see Supporting Information section 13).

Further, polarization-dependent ATR measurements are performed that show a dip at 626 cm$^{-1}$ in the $p$-polarized light, representing coupling of light with the SPhP mode (see Figure 3d). Calculated SPhP dispersion is consistent with the experimental observations and highlight the bulk phonon–polaritons at frequencies above and below the Reststrahlen band (see Figure 3e). Performance FOM of SPhP modes is calculated and compared with other well-established polar dielectric materials, such as SiC, h-BN, c-BN, AlN, and MgO.
Figure 5. STEM images and EDS maps of ScN deposited on (001) MgO substrate. (a) Low-magnification STEM image showing homogeneous and uniform ScN growth with a few dislocations. The electron diffraction pattern in the inset confirms the cubic epitaxial growth. (b) Atomic resolution STEM image of the ScN/MgO demonstrating a sharp interface. STEM–EDS elemental mapping of (c) Sc, (d) N, (e) Mg, and (f) O shows the homogeneous elemental distribution.

GaN,\textsuperscript{56} SPhP FOM of ScN is around two times higher than its peers due to its high $\varepsilon_\infty$ of 12.8 (see Figure 4b and Supporting Information section 11). Further improvement in materials quality is likely to reduce the phonon damping and increase FOM in ScN.

The comparison of plasmonic properties (crossover wavelength and optical loss) shows that the ENZ wavelength of ScN covers $\sim$1750–2500 nm spectral region that lies in between the ENZ wavelength typically exhibited by TCOs and doped-CdO. $\varepsilon_\infty$ of ScN is slightly higher compared to the NIR plasmonic materials such as ITO, AZO; as well as with the MIR plasmonic materials, such as CdO, primarily because of its lower carrier mobility (see Figure 4a). However, compared to the MIR plasmonic materials, such as $n$-Si, $p$-Si, InAs, etc., the optical loss of ScN is smaller. Since advanced deposition methods, such as hybrid vapor phase epitaxy (HVPE) and molecular beam epitaxy (MBE), results in higher carrier mobility (with a maximum reported mobility of $\sim$280 cm$^2$/V s in ScN),\textsuperscript{58} the optical losses could be reduced.

While the compelling optical properties of ScN makes it an attractive IR polaritonic material, SPP waveguides, HMM, ENZ, and other device implementations require low surface roughness, a lattice-matched interface with the substrate and compatibility with industrially relevant materials.

Scanning transmission electron microscopy (STEM), energy-dispersive X-ray spectroscopy (EDS) mapping and electron diffraction were applied to characterize the microstructure of the film (see Supporting Information section 15 for detail). Both oxygen and magnesium-doped ScN films deposited in this work on (001) MgO substrates at high-temperatures grow epitaxially with [001]/[001] ScNll/[001] (001) MgO (see Figure 5a and 5b). The ScN/MgO interface is sharp despite the presence of a few misfit-dislocations, resulting from the 7% lattice-mismatch between ScN and MgO. Importantly, both oxygen and magnesium dopants make homogeneous solid-solutions with ScN without any precipitations or secondary phase formations (see EDS maps in Figure 5c–f) resulting in a small rms surface roughness of $\sim$2 nm (see Figure S9). Electron energy loss spectroscopy (EELS) O K-edge and Mg L-edges are consistent with the bonding of O with Sc and Mg with N respectively and splitting of the peaks highlight hybridization between different orbitals (see Figure S10). While the present work utilizes (001) MgO as a substrate with the same crystal structure, it must be mentioned that ScN films are regularly deposited on industrially important $Al_2O_3$ and Si substrates that should provide seamless chip integration. In addition, because of its near-perfect lattice-matching, (111) ScN films have been deposited on (0001) GaN with very little defects\textsuperscript{43,44} that would also lead to its integration with GaN-based light-emission and power electronic applications.

In conclusion, we present epitaxial refractory group-III scandium nitride (ScN) as a dual plasmon– and phonon–polariton material, where tunable plasmon resonance in the short-wave-infrared (SWIR) spectral range and the phonon–polariton in the long-wave-infrared (LWIR) region are obtained by carrier concentration control. Oxygen-doped ScN films with carrier concentrations between $10^{20}$–$10^{21}$ cm$^{-3}$ exhibit high-quality and low-loss plasmon resonance in the 1.8–2.3 $\mu$m SWIR spectral region, where most plasmonic materials do not work satisfactorily. High figure-of-merit (FOM) surface phonon–polariton resonance is also achieved in the long-wavelength-infrared (LWIR) by reducing the carrier concentration with Mg-hole doping in ScN. Demonstration of the plasmon– and phonon–polariton in one host material, ScN with doping-control makes it an attractive material for applications in waveguides, hyperbolic, and epsilon-near-zero metamaterials, optical communication, solar-energy harvesting, and infrared photonic applications.

### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.2c00912.

Details of sample preparation, optical measurements (ellipsometry and Fourier-transform infrared spectrosc-
copy), structural characterization (SEM, AFM, and TEM), and detailed analysis of results.

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The authors declare no competing financial interest.

**Notes**

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