Room-Temperature Lasing Action in GaN Quantum Wells in the Infrared 1.5 μm Region

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Supporting Information

ABSTRACT: Large-scale optoelectronics integration is strongly limited by the lack of efficient light sources, which could be integrated with the silicon complementary metal-oxide-semiconductor (CMOS) technology. Persistent efforts continue to achieve efficient light emission from silicon in extending the silicon technology into fully integrated optoelectronic circuits. Here, we report the realization of room-temperature stimulated emission in the technologically crucial 1.5 μm wavelength range from Er-doped GaN multiple-quantum wells on silicon and sapphire. Employing the well-acknowledged variable stripe technique, we have demonstrated an optical gain up to 170 cm⁻¹ in the multiple-quantum well structures. The observation of the stimulated emission is accompanied by the characteristic threshold behavior of emission intensity as a function of pump fluence, spectral line width narrowing, and excitation length. The demonstration of room-temperature lasing at the minimum loss window of optical fibers and in the eye-safe wavelength region of 1.5 μm are highly sought after for use in many applications including defense, industrial processing, communication, medicine, spectroscopy, and imaging. As the synthesis of Er-doped GaN epitaxial layers on silicon and sapphire has been successfully demonstrated, the results laid the foundation for achieving hybrid GaN-Si lasers, providing a new pathway toward full photonic integration for silicon optoelectronics.

KEYWORDS: silicone, GaN, quantum wells, lasing, infrared laser, rare earth

Driven by the strong need for cheap and integrable Si-based optoelectronic devices for a wide range of applications, continuing endeavors have been made to develop structures for light emission, modulation, and detection in this material system. Recent breakthroughs, including the demonstration of a high-speed optical modulator in Si,1,2 photo-detectors,3 and waveguides,4 have brought the concept of transition from electrical to optical interconnects closer to realization. However, the base for silicon photonics, namely, a group IV laser source, still has to be developed. Due to the relatively small and indirect band gap, silicon is a poor light emitter. Nevertheless, lasing devices based on Si have been demonstrated including Si-based impurity lasers,5 a Raman laser,6 Si nanocrystals,7 nanopatterned crystalline Si,8 GeSn alloy on Si,9 Ge dots in Si,10,11 and InGaAs/GaAs nanolasers grown on Si.12 However, these prototype devices essentially lack the advantages associated with the silicon system by requiring an external pump laser source or function only at low temperatures. While room-temperature luminescence has been realized,7,13 population inversion and optical gain have been under discussion and fundamental problems remain.

The incorporation of rare earth elements into semiconductor hosts gives rise to sharp, atomic-like, and temperature-independent emission lines under either optical or electrical excitation.14–16 Er ions with intra-4f shell transitions from its first excited state (4I13/2) to the ground state (4I15/2) produce 1.5 μm emission which falls within the minimum loss window of optical fibers for optical communications and in the eye-safe wavelength region. Lasers operating around 1.5 μm are highly sought-after for use in defense, industrial processing, communication, medicine, spectroscopy, and imaging, and various other applications where the laser beam is expected to travel long distances in free space. With a tremendous effort, 1.5 μm emission from Er-doped narrow-bandgap semiconductors including Si and GaAs has a low efficiency at room-temperature due to the strong thermal quenching effect.15,19

RESULTS AND DISCUSSION

Recently, we have successfully synthesized GaN:Er epilayers on Si(001)20,21 and c-plane sapphire22 substrates by metal organic
chemical vapor deposition (MOCVD) with excellent material qualities. The above host bandgap and electroluminescence excitation of Er optical centers produced predominant light emission at 1.5 μm range. In order to overcome the challenges of growth of III-nitrides on Si(100) substrate due to the different crystalline structures between GaN and Si, we have employed selective area growth and epitaxial lateral overgrowth techniques to prepare GaN/AlN/Si(100) templates. The X-ray diffraction and photoluminescence (PL) measurements indicated that GaN:Er epilayers grown on Si and sapphire have high crystallinity, without second phase formation, and exhibit a strong room-temperature emission at 1.5 μm with a low degree of thermal quenching. In this work, a set of 200-period Er-doped GaN/AlN multiple quantum wells (MQWs:Er) produced a significant improvement of the quantum efficiency of the 1.5 μm emission via carrier quantum confinement and strain engineering (see schematic in Figure 1a). The growth process was started with AlN buffer and template layers and then followed by the growth of the MQWs:Er. The structure consists of alternating layers of Er doped GaN quantum wells and undoped AlN barriers (see Methods and Supporting Information). A detailed description of the growth process and epilayer structure has been reported previously.

Typical room-temperature PL spectra from the MQWs:Er samples and a single GaN:Er epilayer show a broad spectral feature around 1.5 μm under the band-to-band excitation using an Ar laser at 351 nm (Figure 1b, inset). The PL spectrum was collected from the surface of the sample, which is the spontaneous emission of light from Er optical centers in MQWs. As previously reported, the full-width at half-maximum (fwhm) of the 1.5 μm emission at room-temperature is 50 nm for a single GaN:Er epilayer and 60 nm for MQWs:Er materials. The broadening of the emission in MQWs is due to the fluctuation of the GaN quantum well width and the local atomic structures around Er optical centers. This is especially a factor for Er optical centers located close to the quantum well/barrier interfaces.

The emission intensity from the MQWs:Er structure is an order of magnitude higher than that of a single GaN:Er epilayer with the same Er doping active layer thickness and concentration under the over bandgap excitation, λexc = 351 nm. The strong quantum confinement effects on the Er emission have been studied for different values of quantum
Schematic of experimental configuration for edge-emission and variable excitation length measurements. (b) PL spectrum at room-temperature with a pump fluence of 116 mJ cm$^{-2}$ and an excitation length of 0.2 mm. The spectrum was deconvolved into six Gaussian peaks. The most prominent modes show a fwhm of 1.60 ± 0.25 meV. Inset: The fwhm of the Gaussian peaks decreases with increasing the pump fluence. (c) L-L data on a log–log scale showing the dependence of the edge-emission intensity for PL peak at 805.3 meV on the pump fluence. The excitation length was 0.2 mm and the data were fitted using the S-curve model. Inset: The edge-emission intensity showing linear behavior below threshold and superlinear increase at higher pump fluence.

Figure 2. Amplified spontaneous emission, at room-temperature, from the MQWs:Er sample with $L_{QW} = 2$ nm and $L_B = 10$ nm using an Ar laser, $\lambda_{exc} = 351$ nm, for the band-to-band excitation. (a) Pump-fluence dependent PL spectra obtained with a 8 $\mu$m wide and 0.2 mm long pump excitation. At low excitation pump fluence, the emission is broad with the fwhm of 30 meV. When the pump fluence is high enough for the optical layer to have net gain, the spontaneously emitted photons are exponentially amplified by stimulated emission and the spectral peaks become narrower. Inset: Schematic of experimental configuration for edge-emission and variable excitation length measurements. (b) PL spectrum at room-temperature with a pump fluence of 116 mJ cm$^{-2}$ and an excitation length of 0.2 mm. The spectrum was deconvolved into six Gaussian peaks. The most prominent modes show a fwhm of 1.60 ± 0.25 meV. Inset: The fwhm of the Gaussian peaks decreases with increasing the pump fluence. (c) L-L data on a log–log scale showing the dependence of the edge-emission intensity for PL peak at 805.3 meV on the pump fluence. The excitation length was 0.2 mm and the data were fitted using the S-curve model. Inset: The edge-emission intensity showing linear behavior below threshold and superlinear increase at higher pump fluence.
Er ions, resulting in the stronger PL emission intensity. The contamination of Al in MQW structures can also provide a factor that affects the thermal property of the PL intensity of Er ions. We have further conducted the temperature dependence of the PL emission intensity for different MQW structures (see Supporting Information). In this work, we compare the PL intensity from MQWs:Er samples with the optimal structure, that is, $L_{QW} = 2$ nm and $L_{B} = 10$ nm, and a GaN:Er single layer.

The percentage of Er ions that emit photons at 1.5 $\mu$m is a crucially important parameter for potential applications of Er-doped GaN since it determines the PL intensity as well as population inversion. An estimate of the number of emitting Er optical centers can be made by comparing the PL intensity of the MQWs:Er sample with that of a SiO$_2$:Er reference sample with the same shape and under the same experimental conditions. In order to avoid the quantum confinement effect for excitons and carriers, the experiment has been performed at room-temperature under the resonant excitation $^{4}I_{15/2} \rightarrow ^{4}I_{9/2}$ transition using the Ti:sapphire laser at 809 nm. Note that under the band-to-band excitation several Er centers are excited, including dark, bright centers. We have observed two kinds of optical centers in the single GaN:Er epilayer, including the defect-related and isolated Er optical centers. The PL from defect-related Er optical centers can be observed under the band-to-band excitation and at the sample temperature below 150 K. The isolated Er optical centers can be excited by both resonant and band-to-band excitation. Thus, it is the correct way to employ the resonant excitation for this estimation. The time-integrated PL intensity of these samples are collected as a function of applied photon fluxes (Figure 1d).

By comparing the PL from Er ions in the MQWs:Er sample with the reference sample of Er-doped SiO$_2$, we have estimated that the fraction of Er ions that emits photon at 1.5 $\mu$m is approximately 65% (Supporting Information). Using the same method, we found that a fraction of ~68% of Er ions in the single GaN:Er layer is optically active. This achievement of the high percentage of Er ions that emit photons at 1.5 $\mu$m represents a significant step in realization of GaN:Er as an optical gain medium.

The edge-emission from optically pumped MQWs:Er sample under the band-to-band excitation provides evidence of room-temperature lasing from Er optical centers in the MQW structure (Figure 2a, inset). In order to achieve lasing from the MQWs:Er sample, both edges of the sample were polished to obtain a cavity. The edge-emission spectra from Er optical centers are different at different pump fluence near threshold (Figure 2a). In the measurements we have employed a long excitation area with 8.0 $\pm$ 0.3 $\mu$m width and 200 $\pm$ 0.5 $\mu$m length (Figure 2a, inset and Methods). The photon fluence of the Ar laser was varied from 0.05 to 120 mJ cm$^{-2}$. At low excitation pump fluence ($P < 15$ mJ cm$^{-2}$), the emission at 1.5 $\mu$m shows a broad spectrum with the fwhm of 60 nm ($\sim$30 meV) that corresponds to the spontaneous emission. The broad PL spectrum is similar to those of Er ions incorporated in insulators including SiO$_2$ at room-temperature. When the pump fluence is high enough ($P > 15$ mJ cm$^{-2}$) for the MQWs:Er sample to obtain a net optical gain, the spontaneously emitted photons are exponentially amplified by stimulated emission as they travel through the waveguide in the active medium, leading to a superlinear increase in emission. Since the gain is maximum near the peak of the spontaneous emission spectrum, the spectrum exhibits “gain narrowing”. Consequently, an intense beam with spectral narrowing is emitted from the edge of the sample. The amplified spontaneous emission occurs at the wavelength where the spontaneous emission spectrum is strongest. The PL spectra at high excitation pump fluence indicate a number of strong and narrow PL lines. This is the signature of optical amplification of the spontaneous emission from Er optical centers around 1.5 $\mu$m in the MQW structure (Figure 2b). We focus to analyze the strongest PL lines at 797.31, 805.30, and 807.55 meV. A deconvolution of the PL spectrum into different Gaussian components is also shown. When the pump fluences were above 15 mJ cm$^{-2}$, the fwhm of the spectrum dropped to 1.60 $\pm$ 0.25 meV (Figure 2b, inset). The PL intensity evolution of narrow peaks indicates a clear threshold transition from subthreshold to linear evolution and ultimately reaches saturation at high pump-fluence (Figure 2b).

A closer examination of this threshold behavior of the PL intensity emitted from the edge of the sample shows the typical superlinear (exponential) to linear transition, indicating the classical spontaneous-to-stimulated emission transition widely observed in semiconductor lasers. Figure 2c shows the light-in-light-out (L-L) data that is the dependence of the amplified spontaneous emission on the pump fluence for the PL peak at 805.3 meV (1539.61 nm) with the excitation length of 0.2 mm. Below the threshold, the PL intensity is linear and a superlinear increase in emission intensity with pump fluence has been observed. Subsequently, stimulated emission dominates and the PL intensity evolution becomes linear (Figure 2c, inset). Fitting the L-L data in the log−log plot to the S-curve model, a lasing threshold of $P_{th} \sim 15$ mJ cm$^{-2}$ has been determined (Figure 2c), with the spontaneous emission coupling factor, $\beta$, of 0.1. The L-L curves for different $\beta$ values are also plotted for comparison, clearly showing the distinct position of the lasing threshold. Additional representative L-L plots of MQWs:Er lasing under optical pumping with different excitation lengths can be found in Supporting Information. Above the threshold for amplified spontaneous emission, most of the excitation was stimulated to emit light into waveguide modes, leading to a large fraction of the light emerged from the edge.

Together with the threshold behavior, we have observed the shortening of the lifetime of Er emission in the MQW structure. A typical lifetime of Er spontaneous emission in the MQW structures with the pump fluence below the threshold is around 2.5 ms (red curve, Figure S4, Supporting Information). Under a high pump fluence (above the threshold), the lifetime of Er emission shows a shortening with a dynamics $<10$ $\mu$s (blue curve, Figure S4, Supporting Information). The value is limited by the time-constant of our time-resolved photoluminescence setup. We have integrated the PL intensity for the fast and slow components. The PL spectrum for the fast component shows narrowing features as indicated in the Figure 2a at high pump fluence. The PL spectrum for the slow component is similar to the spectrum at low pump fluence. The PL intensity of the slow component can be originated from surface emission of the sample.

We have employed the well-known variable excitation length method to determine the gain coefficient from the evolution of the peak-emission intensity (Methods). The sample was optically excited by the argon laser providing the band-to-band excitation in a stripe geometry. The amplified spontaneous emission signal, $I_{ASE}$, was collected as a function of the illuminated length or the excitation length, $l$, from the edge of the sample. As a result of stimulated emission, the spontaneous...
emitted light is amplified as it passes through the excited volume to the edge of the sample. Assuming a one-dimensional amplification model, the modal gain, $g_{mod}$, can be extracted from the amplified spontaneous emission intensity and the excitation length:

$$I_{ASE}(l) = \frac{A \times I_{SPONT}}{g_{mod}} \left( e^{g_{mod}l} - 1 \right)$$  

where $I_{SPONT}$ is the spontaneous emission intensity per unit length and $A$ is the cross-section area of the excited volume, and the modal gain is the gain minus the loss of the material.

When the excitation length was varied, the emission spectral peaks became narrower as the excitation length was increased, and the output intensity grew exponentially. Figure 3a presents the PL spectra with an optical excitation of 109 mJ cm$^{-2}$ for different excitation lengths. The observation of an exponential increase in the intensity and a substantial decrease in spectral line width (Figure 3a, inset) of light emitted as the excitation length is varied is a direct indication of the optical gain. Figure 3b shows the output intensity at the observation peak at 805.3 meV as a function of excitation length. The modal gain of the MQWs:Er sample was determined by using eq 1 to the data.

In conclusion, our investigations provide conclusive evidence of light amplification and stimulated emission in MQWs:Er samples. In the past, this formidable goal was unsuccessfully attempted by Er doping of crystalline Si, and later of SiO$_2$ sensitized with Si nanocrystals. Here we have demonstrated the realization of this long-sought goal of Si photonics in state-of-the-art GaN MQW structures grown on-Si and GaN, demonstrating for the first time the added value provided by merging of these two most important semiconductor materials – Si for standard electronics and photovoltaics and GaN for power electronics, photonics and automotive applications.

**METHODS**

**Sample Fabrication.** Er-doped GaN/AlN MQWs (MQWs:Er) were grown by MOCVD. The aluminum source was trimethylaluminum (TMA), the gallium source was trimethylgallium (TMGa), and the nitrogen source was ammonia (NH$_3$). Tris(isopropylcyclopentadienyl)erbium (TRIPEr) was used as a precursor for in situ Er doping. The growth started with a thin (30 nm) AlN buffer layer (buffer 1) grown at 950 °C and 30 mbar followed by a second 100 nm AlN buffer layer (buffer 2) grown at 1100 °C grown at 30 mbar, and a 1.0 μm AlN template grown at 1325 °C and 30 mbar. It was then followed by the growth of the MQWs:Er structure of Er-doped GaN quantum wells and undoped AlN barrier layers. The growth temperature and pressure were 1000 °C and 30 mbar.

**PL Intensity Measurement.** The PL spectra were collected with a high resolution Horiba iHR550 spectrometer equipped with a 900 grooves/mm grating blazed at 1.5 μm and detected by a high sensitivity liquid nitrogen InGaAs DSS-IGA detector. The resolution of PL spectrum is 0.05 nm. The PL...
experiments were carried out in a variable temperature closed-cycle optical cryostat (Janis) within the temperature range from 10 to 300 K. We have employed resonant and nonresonant excitation to investigate optical properties of the MQWs:Er structures. An Argon laser emitting light at 351 nm (3.531 eV) was employed for the nonresonant (band-to-band) excitation of MQWs:Er materials. The PL spectra under the resonant excitation from $^4I_{15/2} \rightarrow ^4I_{9/2}$ of Er$^{3+}$ in GaN were obtained using a tunable wavelength Ti:sapphire laser around 809 nm (1.533 eV).\(^{23}\)

**Edge-Emission Measurements.** The pump laser was magnified and then focused onto the sample’s top surface using a cylindrical lens with $f = 7.5$ cm (Figure 2b, inset). Only the central part of the laterally unfocused laser spot was used to excite the sample so that the pump fluence was uniform across the entire excited area. The excitation area was measured to be a long stripe of $8.0 \pm 0.3 \mu m$ wide exciting the entire sample length of $1000 \pm 0.5 \mu m$. We used a two-dimensional linear stage to scan our UV photodetector at the focal position of the laser to verify that within experimental conditions the laser pump fluence on the sample surface was constant and independent of the length. No influence of diffraction effects on the uniformity of the pump laser were detected. An aperture was used to ensure that none of the light, which has passed out of the excited volume before reaching the edge of the sample, was detected. The edge-emission was collected using a set of two lenses $f = 10$ and 24 cm, with a diameter of 5 cm, and focused onto the entrance slit of the high resolution spectrometer through a long-pass filter blocking all background light with a wavelength shorter than 950 nm. All the edge-emission measurements were done at room-temperature. The photon fluence on the sample was varied from 0.05 to 500 mJ cm$^{-2}$.

**Gain Measurements.** Net optical gain of the MQWs:Er sample with optimized structural parameters (i.e., GaN $L_{\text{QW}} = 2$ nm and AlN $L_{\text{Al}} = 10$ nm) was measured using variable excitation length method. In the same edge-emission measurement configuration, a mobile blade was mounted on an ultraprecise linear translation stage (relative accuracy of 80 μm). The edge-emission was collected and focused onto the entrance slit of the spectrometer as described. The measured gain coefficients show a fluctuation of within 5% for different distances between the sample’s top surface and the mobile blade, suggesting that diffraction effects can be safely disregarded.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsphotonics.7b01253.

Details of the fabrication procedure of MOVCVD growth of Er-doped multiple quantum wells, temperature dependence of MQWs:Er samples, optical activity determination, and rate equation analysis (PDF).

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**Author Contributions**

V.X.H. performed the optical and lasing experiments supervised by N.Q.V.; N.Q.V., H.X.J., J.Y.L., and J.M.Z. designed the research program; H.X.J., J.Y.L., and T.M.A. were in charge of MOVCVD growth of Er-doped GaN epilayers and MQW samples; and N.Q.V., V.X.H., J.Y.L., and J.M.Z. wrote the paper.

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

N.Q.V. acknowledges the support from NSF (ECCS-1358564). The materials growth effort at TTU was supported by JTO/ARO (W911NF-12-1-0330).

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