Observation of Asymmetric Nanoscale Optical Cavity in GaAs Nanosheets

Shermin Arab,*,‡ P. Duke Anderson,*,‡ Chun-Yung Chi,*,‡ P. Daniel Dapkus,*,‡,§∥,‡,§∥ Michelle L. Povinelli,†,‡ and Stephen B. Cronin*,‡,§∥

† Department of Electrical Engineering, ‡ Department of Physics, § Department of Chemical Engineering and Materials Science, and ¶ Center for Energy Nanoscience, University of Southern California, Los Angeles, California 90089, United States

Supporting Information

ABSTRACT: GaAs nanosheets with no twin defects, stacking faults, or dislocations are excellent candidates for optoelectrical applications. Their outstanding optical behavior and twin free structure make them superior to traditionally studied GaAs nanowires. While many research groups have reported optically resonant cavities (i.e., Fabry–Perot) in 1D nanowires, here, we report an optical cavity resonance in GaAs nanosheets consisting of complex 2D asymmetric modes, which are fundamentally different from one-dimensional cavities. These resonant modes are detected experimentally using photoluminescence (PL) spectroscopy, which exhibits a series of peaks or “fringes” superimposed on the bulk GaAs photoluminescence spectrum. Finite-difference time-domain (FDTD) simulations confirm these experimental findings and provide a detailed picture of these complex resonant modes. Here, the complex modes of this cavity are formed by the three nonparallel edges of the GaAs nanosheets. Due to the asymmetrical nature of the nanosheets, the mode profiles are largely unintuitive. We also find that by changing the substrate from Si/SiO₂ to Au, we enhance the resonance fringes as well as the overall optical emission by 5X at room temperature. Our FDTD simulation results confirm that this enhancement is caused by the local field enhancement of the Au substrate and indicate that the thickness of the nanosheets plays an important role in the formation and enhancement of fringes.

KEYWORDS: MOCVD, GaAs, nanosheet, nanowires, photoluminescence

GaAs nanostructures are among the best candidates for optoelectrical applications including solar cells, LEDs, and lasers.1−5 However, these structures, particularly nanowires, suffer from various defects, such as twin formation, dislocations, and stacking faults. While GaAs nanowires suffer from such defects, nanosheets grow easily without twin defects or stacking faults; hence, overall they show higher quality and optical properties in comparison to GaAs nanowires.7 Nanoscale lasers have been studied in a variety of nanostructure geometries. So far, both nanowire lasers and thin film semiconductors have demonstrated high intensity emission under optical or electrical pumping.1−6 Duan et al. reported electrically pumped lasing from n-type CdS nanowires.7 Fukui et al. reported observation of Fabry-Perot (FP) resonances and later lasing from GaAs nanowires.3,8,9 Most recently, Saxena et al.10 reported passivated GaAs nanowire lasing at room temperature by optimizing the size and surface passivation. In our previous work, we demonstrated enhanced FP resonance from GaAs nanowires at room temperature through surface passivation with wide band gap semiconductor AlGaAs and also ionic liquid electrolyte.11 Since the original paper reporting nanowire lasing,1 much of the effort in this area has been focused on reducing the effects of defects and impurities in order to improve the optoelectronic properties of these nanomaterials.

In the work presented here, we investigate GaAs nanosheets, which have demonstrated improved crystal structure over nanowires and present a new morphology for optically resonant devices and systems. This same general approach was previously applied by our group to GaAs nanowires.11 However, the results observed from 2D nanosheets are nonintuitive. It should be noted that a vast majority of previous papers reporting on optical cavity resonances in nanoscale systems are reported in 1D nanowires and 2D photonic crystals, although similar observations in non-nanoscale regime have been studied.12−14 Traditional photonic crystals require difficult nanofabrication techniques,15,16 and the 2D nanosheets provide a new platform for achieving 2D resonances without the need for complex nanofabrication. The Q-factor reported for artificially fabricated PCs are usually in a higher range (10⁵ and higher) in comparison to the optical cavity formed here; however, we believe after AlGaAs passivation, these GaAs nanosheets can provide higher Q-factors.17 The GaAs nanosheet optical cavities may also be compared to triangular-shaped lasers reported previously.18,19 However, in most cases, the triangular lasers are formed from heterostructures or...
superlattices. In general, the nonradiative recombination processes are well-controlled in triangular lasers, which lead to higher quality factors and longer carrier lifetimes; both of which result in higher emissions and lower lasing thresholds.

Most previous studies report one-dimensional cavities formed either between two parallel mirrors or between the two end facets of a nanowire. In the work presented here, we demonstrate an in-plane (2D) cavity formed by the three nonparallel edges of the GaAs nanosheets. The nature of this resonance is studied using photoluminescence (PL) spectroscopy as a function of the GaAs nanosheet dimensions, laser power, temperature, and underlying substrate. In order to obtain a fundamental understanding of the resonances and underlying PL enhancement mechanism, finite-difference time-domain (FDTD) simulations are carried out on an individual GaAs nanosheet. The simulations are configured to separate various enhancement mechanisms of light absorption and emission. Optical cavity modes are visualized by plotting the electric field intensity through a cross-sectional slice of the nanosheet. Both the strength and quality factors of the resonances are extracted from measurements and calculations. Formation of such cavities provides a path toward the development of room-temperature 2D nanolasers. An optically pumped, cost-effective nanolaser with reduced footprint could be used in optical communications.

GaAs nanosheets are synthesized on (111)B GaAs substrates by metal organic chemical vapor deposition (MOCVD) with selective area growth (SAG). The detailed synthesis method has been described in previous publications. Briefly, trimethylgallium (TMGa) and arsine are used as precursors for Ga and As deposition, and nanosheets are n-type doped using disilane (Si₂H₆). A 28 nm layer of Si₃N₄ is deposited on the GaAs substrate using plasma enhanced chemical vapor deposition (PECVD), which serves as the growth mask. Electron beam lithography is used to create nanoscale strips in the Si₃N₄, which is selectively etched, leading to the formation of windows exposing the underlying GaAs substrate. The nanosheets are grown in a hydrogen environment at 0.1 atm using a V/III ratio of 1.5 and Ga/Si ratio of about 16000 for 30 min at 790 °C. The partial pressure of TMGa, AsH₃, and Si₂H₆ are 1.124 × 10⁻⁶, 1.63 × 10⁻⁶, and 2.857 × 10⁻⁹ atm, respectively. The as-grown nanosheets are sonicated for 5 s in isopropanol and subsequently transferred onto Si/SiO₂ (300 nm SiO₂) or Si/SiO₂/Au (300 nm- SiO₂/200 nm Au) substrates. These GaAs nanosheets grow on (111)B surfaces through a self-terminating growth mechanism, which results in their triangular shape with a base length of 6–10 μm and height of 2–4 μm, as shown in Figure 1a. The thickness of these nanostructures is 200 ± 25 nm. Micro-Raman and photoluminescence spectroscopy are collected using a 40X objective lens, an 1800 L/mm grating, and a silicon CCD camera to detect the photoluminescence (PL) in the range of 750 to 950 nm. Low to moderate power excitation (10²–10⁵ mW/cm²) using a 532 nm laser is used to avoid optical heating.

Figure 1a shows an SEM image of a GaAs nanosheet deposited on a Si/SiO₂ substrate. The thickness of the nanosheet is measured using atomic force microscopy (AFM), which gives a measured value of 200 ± 20 nm. The center of the sample is excited by a 532 nm CW laser focused to a spot size of 1.5 μm with an excitation power of 10⁵ mW/cm². The corresponding PL spectrum taken at 77 K is shown in Figure 1b. The spacing between the resonance peaks varies between 6.5 and 10 nm, with an average spacing of 8.5 nm. In this spectrum, two predominant peaks are observed at 1.52 and 1.44 eV. The peak at 1.52 eV corresponds to the nominal band gap of GaAs (1.507 eV at 77 K), blueshifted by 12 meV due to the n-type doping by Si dopant impurities, which results in Pauli blocking of band-edge to band-edge optical transitions. The 1.44 eV sub-band gap peak is attributed to an acceptor-bond transition arising from carbon impurities originating from the metalorganic precursors of the MOCVD growth. The quality factors of the resonance peaks range from 88 to 125, indicating that trapped light is totally internally reflected roughly 100 times. These resonance fringes are only observed in nanosheets with thickness of 200–220 nm, whereas no such resonance is observed in thicker GaAs (see Figure S1 in the Supporting Information). These values of the Q-factor and optimal thicknesses are quite similar to those of GaAs nanowires described in our previous work. Table 1 lists a comparison of resonance parameters between nanowires and nanosheets.

### Table 1. Comparison of Resonance Parameters for GaAs Nanowires and Nanosheets

<table>
<thead>
<tr>
<th>Parameter</th>
<th>GaAs nanowires</th>
<th>GaAs nanosheets</th>
</tr>
</thead>
<tbody>
<tr>
<td>peak-to-peak spacing (experimental)</td>
<td>8–10 nm</td>
<td>6.5–10 nm</td>
</tr>
<tr>
<td>peak-to-peak spacing (theoretical)</td>
<td>11 nm</td>
<td>12.3 nm</td>
</tr>
<tr>
<td>quality factor</td>
<td>181</td>
<td>125</td>
</tr>
<tr>
<td>optimal thickness</td>
<td>270 nm ±20 nm</td>
<td>210 nm ±10 nm</td>
</tr>
</tbody>
</table>

This work demonstrates the potential of GaAs nanosheets as a platform for the development of optically pumped, cost-effective nanolasers with reduced footprint. The optical cavity modes are analyzed through photoluminescence spectroscopy as a function of the GaAs nanosheet dimensions, laser power, temperature, and underlying substrate. The quality factors of the resonances range from 88 to 125, indicating that trapped light is totally internally reflected roughly 100 times. These resonance fringes are only observed in nanosheets with thickness of 200–220 nm, whereas no such resonance is observed in thicker GaAs. The values of the Q-factor and optimal thicknesses are quite similar to those of GaAs nanowires described in our previous work. This work demonstrates the potential of GaAs nanosheets as a platform for the development of optically pumped, cost-effective nanolasers with reduced footprint. The optical cavity modes are analyzed through photoluminescence spectroscopy as a function of the GaAs nanosheet dimensions, laser power, temperature, and underlying substrate. The quality factors of the resonances range from 88 to 125, indicating that trapped light is totally internally reflected roughly 100 times. These resonance fringes are only observed in nanosheets with thickness of 200–220 nm, whereas no such resonance is observed in thicker GaAs. The values of the Q-factor and optimal thicknesses are quite similar to those of GaAs nanowires described in our previous work.
the NS and the lowest for the case where the polarization is perpendicular to the base.

In Figure 2, the PL spectra of a similar GaAs nanosheet deposited on a Si/SiO₂/Au (200 nm of Au deposited on Si/SiO₂) substrate is compared to that of one taken on Si/SiO₂/Au (200 nm of Au deposited on Si/SiO₂). Spectra were collected at room temperature under 532 nm CW laser excitation.

![Figure 2](image)

**Figure 2.** Photoluminescence spectra from GaAs nanosheets on Si/SiO₂ and Si/SiO₂/Au. Spectra were collected at room temperature under 532 nm CW laser excitation.

With the pump excitation at 532 nm, the absorptive enhancement factor (EF_ABS) for a single wavelength is simply the ratio of the integrated electric-field intensities:

\[
EF_{\text{ABS}} = \frac{\int |E_{\text{Au}}|^2 \, dA}{\int |E_{\text{SiO}_2}|^2 \, dA}
\]

With the pump excitation at 532 nm, the absorptive enhancement factor is nearly 11.5X, indicating that the Au substrate leads to 11.5X more optical absorption within the nanosheet. Here, the metallic film appears to form a vertical cavity with the top of the nanosheet that traps light and results in a large absorptive enhancement factor.

We model PL emission within the GaAs nanosheet using electric dipole sources. Each electric dipole emits between 750 and 1050 nm. A Gaussian distribution is superimposed upon the emission spectra so that the results more closely resemble the PL data collected experimentally. In order to examine the impact of the emitter position on the observed optical resonances, we consider four emitter positions: one position near each corner of the nanosheet and one position near its center. At each emitter position, we consider three orthogonal polarizations (i.e., x, y, and z). In order to avoid coherence effects, we include one dipole per simulation and later perform an incoherent summation of the emission results.

We introduce a quantity, the extracted power (EP), which is defined as the total power collected from the top of the GaAs nanosheet, measured using a flux-plane monitor. Figure 4a shows the incoherently summed EP results for the cross-sectional position located near the smallest corner of the nanosheet. Optical fringes are clearly visible in the simulated spectra for both the SiO₂ and Au underlying substrate. Emission characteristics relating to the remaining three emitter positions are described in Supporting Information. Figure 4b shows the electric-field intensity patterns for the wave-
enhancement is also seen in our FDTD simulations, which show a product of absorptive enhancement and extracted power underlining metallic contributions. The majority of the enhancement is owed to absorptive enhancement, whereas the green dot indicates the position of the dipole emitters.

The integrated extracted power enhancement \(\left(EP_{\text{enh}}\right)\) is defined as

\[
EP_{\text{enh}} = \frac{\int EP_{\text{Au}} \, d\lambda}{\int EP_{\text{SiO}_2} \, d\lambda}
\]

and represents the comparative enhancement in collected emission due to the Au film. We find that this enhancement is independent of the emitter’s position (see Supporting Information, Figure S2) with a value always near unity. The broadband EP enhancement for the emission results shown in Figure 4a is 1.2×. If we consider the overall PL enhancement to be a product of absorptive enhancement and extracted power (EP) enhancement, then we theoretically predict a PL enhancement of nearly 13.8×. The majority of the enhancement contribution is owed to absorptive enhancement, resulting from the formation of a vertical cavity due to the underlying metallic film. The observation of optical resonances, however, is a consequence of the in-plane cavity formed by the cross-sectional profile of the nanosheet. Both the number and quality-factor of observed resonances is strongly dependent on the emitter’s position (see Supporting Information).

In conclusion, we report the observation of an optical resonance in GaAs nanosheets resulting from the formation of an asymmetric cavity created by the nonparallel edges of these nanostructures. The optical resonance is seen experimentally in the photoluminescence spectra of the nanosheets, as a series of peaks of fringes observed on top of the normal photoluminescence spectrum of bulk GaAs. The resonance consists of complex in-plane modes resulting from total internal reflections within the nanosheet itself, as seen in the electric field intensity profiles calculated by finite-difference time-domain simulations. Q-factors of 125 are observed in these nanosheets, which is comparable to previous measurements of GaAs nanowires. By changing the underlying substrate from Si/SiO\(_2\) to Si/SiO\(_2\)/Au, a 5× enhancement in the PL intensity and resonance fringes is observed in these GaAs nanosheets. This substrate-induced PL enhancement is also seen in our FDTD simulations, which indicate that the enhancement is primarily due to absorptive enhancement rather than photoemission enhancement. The overall theoretical enhancement due to both absorptive and extracted power is predicted to be 13.8× and does not include any losses due to nonradiative recombination effects.


