

Ultrawide strain-tuning of light emission from InGaAs nanomembranes

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Abstract: Single-crystal semiconductor nanomembranes provide unique opportunities for basic studies and device applications of strain engineering, by virtue of mechanical properties analogous to those of flexible polymeric materials. Here we investigate the radiative properties of nanomembranes based on InGaAs (one of the standard active materials for infrared diode lasers) under external mechanical stress. Photoluminescence measurements show that, by varying the applied stress, the InGaAs bandgap energy can be red-shifted by over 250 nm, leading to efficient strain-tunable light emission across the same spectral range. These mechanically stressed nanomembranes could therefore form the basis for actively tunable semiconductor lasers featuring ultrawide tunability of the output wavelength.

Several key application areas of semiconductor diode lasers, including optical communications, spectroscopy, and sensing, require the ability to tune the laser output wavelength dynamically over a broad spectral range. At present, this functionality is typically implemented by shifting the reflectivity peak of a distributed grating mirror or external cavity across a fixed laser gain spectrum.¹ As a result, the maximum achievable tuning range is necessarily limited to a fraction of the gain bandwidth, normally on the order of several ten nanometers for standard near-infrared devices. To expand on this limit, significant efforts have focused on the development of laser materials with intentionally broadened gain spectra, including compositionally or geometrically graded multiple-quantum-well structures²⁻⁴ and self-assembled quantum-dot ensembles featuring large size variations.⁵⁻⁸ In the present work we consider an alternative materials platform, consisting of mechanically stressed semiconductor nanomembranes (NMs), which could enable greatly enhanced tunability based on an entirely different approach, i.e., the controlled introduction of strain to shift the gain spectrum.

Nanomembranes are single-crystal sheets with macroscale lateral dimensions and highly sub-micron thicknesses, typically released from their native substrates through the selective wet etch of an underlying sacrificial layer. Because of these extreme aspect ratios, they can feature a high degree of mechanical flexibility (similar to that of soft polymeric materials), while at the same time retaining the superior electronic and optical properties of inorganic semiconductors. As a result, they are promising for a wide range of basic studies and device applications in flexible (opto)electronics.⁹⁻¹⁵ In the context of light-emitting materials and devices, NM strain engineering has been applied recently to mechanically stressed Ge, where sufficiently large tensile strain ($\sim 2\%$) could be introduced to obtain direct-bandgap behavior with highly enhanced radiative efficiency.^{16,}

¹⁷ Tensile strain in typical semiconductors (including Ge as well as standard III-V optoelectronic

materials) also has the effect of decreasing the bandgap energy, by simultaneously lowering the conduction-band edge and raising the valence-band maximum.¹⁸ Therefore, the external application of mechanical stress can also be used as a means to vary dynamically the interband emission spectrum of the underlying materials. Related prior work along these lines includes the report of wideband tunability of III-V nanowire luminescence¹⁹⁻²¹ and piezoelectric-controlled fine tuning of the exciton binding energies in NM-embedded quantum dots for quantum-optics applications.²²⁻²⁴ Anisotropic tensile strain introduced with piezoelectric actuators has also been utilized to control the low-temperature excitonic luminescence from GaAs NMs²⁵. Here we investigate room-temperature interband light emission from tensilely strained NMs based on InGaAs (one of the standard active materials for near-infrared diode lasers), and demonstrate an ultrawide tuning range of about 250 nm.

The experimental samples are based on a 100-nm-thick (001) $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ film grown by molecular beam epitaxy (MBE) lattice-matched on an $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$ layer on InP (wafer VA0927). A thin layer of photoresist is spin-coated on the InGaAs film, which is then released from the underlying sacrificial materials with a selective wet etch in HCl.²⁶ The resulting free-standing NM is then transferred from deionized water onto a 125- μm -thick flexible film of polyimide (PI) (Kapton[®] HN from DuPont[™]) using a wire loop. The NM initially settles and fully conforms on the PI film via hydrogen and van der Waals bonding, and is then more firmly bonded with a two-step annealing process (first at 60 °C for 20 minutes, and then, after removal of the photoresist in acetone, at 100 °C for 2 hours). The specific NM used in the strain-dependent measurements described below has lateral dimensions of about $5\times 5\text{ mm}^2$. In these measurements, the InGaAs NM and supporting PI film are mounted in a rigid cell that is then pressurized with a controlled gas inflow (Fig. 1). With this arrangement, the NM effectively lies on the surface of an expanding

sphere of PI, and as a result highly isotropic in-plane biaxial tensile strain is introduced in the InGaAs lattice. It should also be noted that, at all pressures used in our measurements, the PI radius of curvature is always at least an order of magnitude larger than the NM lateral dimensions, so that any bending of the NM plane is negligible.

The NM strain as a function of gas pressure is determined by X-ray diffraction (XRD) with a Bruker D8 Discover system, by measuring the diffraction signal along the (001) direction in the rocking curve mode. The out-of-plane strain ϵ_{zz} is computed from the Bragg condition based on the measured change in diffraction angle with pressure, and the corresponding in-plane biaxial strain ($\epsilon_{xx} = \epsilon_{yy}$) is then derived using the elastic stiffness constants of $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ (obtained from a linear interpolation of the InAs and GaAs values from ref. 18). The resulting strain-stress curve (measured from a large sample area of about $0.5 \times 0.9 \text{ mm}^2$) is plotted in Fig. 2 up to a maximum pressure of 620 kPa, for which a biaxial tensile strain of over 1.1 % is obtained. Representative X-ray diffractograms are shown in the figure inset, illustrating the large shift caused by the modified lattice constants under pressure. The diffraction peak is also found to broaden with applied stress, indicating an increasingly non-uniform strain distribution across the NM surface (with an additional, relatively strain-independent shoulder, possibly originating from a defective or partially delaminated region of the NM). This behavior is quantified by the error bars shown in the figure, obtained from a Lorentzian fit of the main diffraction peak followed by a deconvolution of the system response²⁷ (which largely determines the linewidth at zero strain). It should also be noted that the strain plotted in Fig. 2 increases steadily with stress across the entire measured range, without any pronounced saturation, consistent with predominantly elastic expansion of the NM lattice.

The NM strain-dependent light emission properties are investigated via room-temperature photoluminescence (PL) studies. The pump light is provided by a tunable optical parametric oscillator (OPO), and consists of a train of pulses with 5-ns width, 20-Hz repetition rate, 1100-nm wavelength, and 0.5-mW average power, focused onto the NM with a spot size of about 1 mm. Optimal alignment of the sample in the PL setup is carefully verified after each change in applied gas pressure. The emitted light is dispersed through a monochromator and finally measured using a room-temperature extended-range InGaAs photodetector with 1.2–2.6 μm spectral response. The measured PL spectra are plotted in Fig. 3 for different values of the NM biaxial tensile strain, determined from the applied gas pressure based on the XRD data of Fig. 2. As the strain increases, a large red shift in the wavelength of peak emission is observed, from the initial unstrained value of 1650 nm to 1900 nm at the highest measured pressure of 620 kPa. These data therefore fully support the notion that the use of mechanically stressed NMs can be applied to standard III-V semiconductor laser materials to provide ultrawide active tuning of the emission spectrum. The PL spectra of Fig. 3 also maintain the same regular shape as the strain is increased up to nearly 1.1 %, again with some broadening originating from strain inhomogeneities similar to that observed in the X-ray diffractograms described above. In contrast, at higher pressures the spectral lineshape breaks up into multiple peaks of comparable amplitude, as illustrated by the right-most trace in the figure (measured at a strain of 1.16 %). This behavior may indicate the onset of extensive plastic relaxation, or alternatively delamination of the NM from the supporting PI film, and thus (currently) prevents further strain tuning of the emission peak to even longer wavelengths.

To quantify these observations, we have used deformation-potential theory¹⁸ to calculate the band-edge energies of the conduction, heavy-hole (HH), and light-hole (LH) bands of $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ as a function of biaxial strain. The relevant materials parameters, including

unstrained bandgap energy E_g , spin-orbit split-off energy, deformation potentials, and elastic constants were again obtained from a linear interpolation (quadratic in the case of E_g) of the corresponding values for InAs and GaAs from ref. 18. As the strain is increased, the conduction-band edge is found to decrease while the degeneracy of the valence-band edges is lifted, with the LH and HH maxima increasing and decreasing, respectively. This behavior is illustrated schematically in the inset of Fig. 4(a), whereas the solid lines in the same figure show the calculated bandgap wavelengths between the conduction and both LH and HH valence bands versus biaxial strain.

For a comparison with the luminescence data, we note that in bulk semiconductors the bandgap energy generally determines the long-wavelength edge of the emission spectrum, because of the vanishing joint density of states at phonon energies approaching E_g . The strain dependence of such emission edge for the NM under study is illustrated by the symbols of Fig. 4(a), which show the spectral positions of the half-maximum points on the long-wavelength side of the emission peaks of Fig. 3. The comparison between these experimental data and calculation results clearly confirms the strain-related origin of the measured tunability of the PL emission. The same plot also indicates that, under strain, the measured luminescence mostly involves electronic transitions into the LH (rather than HH) valence band, consistent with its higher band-edge energy and therefore higher hole occupancy. Additionally, Fig. 4(a) suggests that, at the highest measured strain levels, the NM emission shifts to somewhat shorter wavelengths relative to the fundamental band gap. This behavior can be explained by noting that, as the applied gas pressure is increased, the PL pump light (incident at a fixed wavelength of 1100 nm) is absorbed more and more efficiently by the NM, because of its decreasing bandgap energies. As a result, more and more

photocarriers are introduced near the band edges, shifting the emission spectrum to shorter and shorter wavelengths through band filling effects.¹⁸

Finally, in Fig. 4(b) we show the measured strain variations in output PL intensity I_{PL} , obtained by integrating the spectra of Fig. 3 over their entire emission bandwidths. The initial increase in I_{PL} with strain can be explained in terms of the aforementioned enhancement in PL pump-light absorption with increasing gas pressure. The subsequent decrease can be at least partly ascribed to the polarization selection rules of interband optical transitions in cubic semiconductors. In the presence of increasing biaxial tensile strain, light emission from electron-LH recombination becomes increasingly TM polarized (i.e., with electric field perpendicular to the plane of the biaxial tension), and as a result it is collected less and less efficiently from the sample surface. In particular, basic theoretical considerations following ref. 28 indicate that, as the strain is increased from 0 to 1 %, the magnitude squared of the TE-polarized conduction-to-LH momentum matrix element of $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ (and therefore the corresponding spontaneous emission rate) decreases by a factor of $1.5\times$ [inset of Fig. 4(b)]. Additional effects that may contribute to the decrease in I_{PL} at high strain include increased heating caused by the enhanced pump-light absorption and the onset of defect formation providing additional channels for nonradiative recombination. In any case, the data of Fig. 4(b) suggest that a relatively high radiation efficiency (comparable to that of unstrained InGaAs) is maintained across the entire measured range of applied pressure.

These results therefore show that mechanical stress in InGaAs NMs can be effectively used to actively tune the emission wavelength over an ultrawide spectral range of at least 250 nm. The maximum achievable strain (about 1.1 % in the present samples) can be further increased by using thinner NMs, as a way to produce even wider tunability of the emission wavelength. This expectation is supported by prior work with Ge NMs,¹⁶ as well as basic considerations from thin-

film mechanics, i.e., the thinner the NM, the smaller the accumulated strain energy for fixed applied stress, and therefore the higher the strain threshold for plastic relaxation.²⁹ These NMs could therefore form the basis for semiconductor lasers providing record wide tunability, by combining the application of mechanical stress for coarse tuning of the gain spectrum with the use of an adjustable external cavity for fine tuning of the lasing wavelength. High-performance laser operation can be expected from the resulting devices, by virtue of the exceptional optical-gain properties of InGaAs combined with the planar extended geometry of NMs, allowing for the development of large-area high-gain active regions. In fact, it should be noted that the strain-induced splitting of the valence bands under mechanical stress is also advantageous for the purpose of reducing the laser threshold, because of the resulting decrease in the hole density of states near the valence-band edge.²⁸ Practical device configurations may be implemented by replacing the pressure cell used in the present work with piezoelectric^{23-25,30} or MEMS actuators³¹.

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References:

1. L. A. Coldren, G. A. Fish, Y. Akulova, J. S. Barton, L. Johansson, and C. W. Coldren, IEEE J. Lightwave Technol. **22**, 193 (2004).
2. H. S. Gingrich, D. R. Chumney, S.-Z. Sun, S. D. Hersee, L. F. Lester, and S. R. J. Brueck, IEEE Photon. Technol. Lett. **9**, 155 (1997).
3. C.-F. Lin, Y.-S. Su, and B.-R. Wu, IEEE Photon. Technol. Lett. **14**, 3 (2002).
4. S. C. Woodworth, D. T. Cassidy, and M. J. Hamp, IEEE J. Quantum Electron. **39**, 426 (2003).
5. P. M. Varangis, H. Li, G. T. Liu, T. C. Newell, A. Stintz, B. Fuchs, K. J. Malloy and L. F. Lester, Electron. Lett. **36**, 1544 (2000).
6. G. Ortner, C. Ni. Allen, C. Dion, P. Barrios, D. Poitras, D. Dalacu, G. Pakulski, J. Lapointe, P. J. Poole, W. Render, and S. Raymond, Appl. Phys. Lett. **88**, 121119 (2006).
7. K. A. Fedorova, M. A. Cataluna, I. Krestnikov, D. Livshits, and E. U. Rafailov, Opt. Express **18**, 19438 (2010).
8. H.-H. Yuan, F. Gao, and T. Yang, Opt. Lett. **43**, 3025 (2018).
9. M. M. Roberts, L. J. Klein, D. E. Savage, K. A. Slinker, M. Friesen, G. Celler, M. A. Eriksson, and M. G. Lagally, Nature Mater. **5**, 388 (2006).
10. D. H. Kim, J. H. Ahn, W. M. Choi, H. S. Kim, T. H. Kim, J. Z. Song, Y. G. Huang, Z. J. Liu, C. Lu, and J. A. Rogers, Science **320**, 507 (2008).
11. H. Yang, D. Zhao, S. Chuwongin, J.-H. Seo, W. Yang, Y. Shuai, J. Berggren, M. Hammar, Z. Ma, and W. Zhou, Nature Photon. **6**, 615 (2012).
12. P. Sookchoo, F. F. Sudradjat, A. M. Kiefer, H. Durmaz, R. Paiella, and M. G. Lagally, ACS Nano **7**, 2326 (2013).

13. X. Xu, H. Subbaraman, S. Chakravarty, A. Hosseini, J. Covey, Y. Yu, D. Kwong, Y. Zhang, W.-C. Lai, Y. Zou, N. Lu, and R. T. Chen, *ACS Nano* **8**, 12265 (2014).
14. J.-H. Seo, K. Zhang, M. Kim, D. Zhao, H. Yang, W. Zhou, and Z. Ma, *Adv. Opt. Mater.* **4**, 120 (2016).
15. Q. Guo, Z. Di, M. G. Lagally, Y. Mei, *Mater. Sci. Eng. R* **128**, 1 (2018).
16. J. R. Sánchez Pérez, C. Boztug, F. Chen, F. F. Sudradjat, D. M. Paskiewicz, R. B. Jacobson, M. G. Lagally, and R. Paiella, *Proc. Natl. Acad. Sci. USA* **108**, 18893 (2011).
17. C. Boztug, J. R. Sánchez Pérez, F. Cavallo, M. G. Lagally, and R. Paiella, *ACS Nano* **8**, 3136 (2014).
18. S. L. Chuang, *Physics of Photonic Devices* (2nd ed.; Wiley: Hoboken, NJ, 2009).
19. I. Zardo, S. Yazji, C. Marini, E. Uccelli, A. Fontcuberta i Morral, G. Abstreiter, and P. Postorino, *ACS Nano* **6**, 3284 (2012).
20. G. Signorello, S. Karg, M. T. Björk, B. Gotsmann, and H. Riel, *Nano Lett.* **13**, 917 (2013).
21. J. Greil, S. Assali, Y. Isono, A. Belabbes, F. Bechstedt, F. O. Valega Mackenzie, A. Yu. Silov, E. P. A. M. Bakkers, and J. E. M. Haverkort, *Nano Lett.* **16**, 3703 (2016).
22. F. Ding, R. Singh, J. D. Plumhof, T. Zander, V. Křápek, Y. H. Chen, M. Benyoucef, V. Zwiller, K. Dörr, G. Bester, A. Rastelli, *Phys. Rev. Lett* **104**, 067405 (2010).
23. R. Trotta, J. Martín-Sánchez, J. S. Wildman, G. Piredda, R. Marcus, S. Christian, Z. Eugenio, S. Stroj, J. Edlinger, A. Rastelli, *Nature Comm.* **7**, 10375 (2016).
24. Y. Chen, J. Zhang, M. Zopf, K. Jung, Y. Zhang, R. Keil, F. Ding, O. G. Schmidt, *Nature Comm.* **7**, 10387 (2016).

25. J. Martín-Sánchez, R. Trotta, G. Piredda, C. Schimpf, G. Trevisi, L. Seravalli, P. Frigeri, S. Stroj, T. Lettner, M. Reindl, J. S. Wildmann, J. Edlinger, A. Rastelli, *Adv. Opt. Mater.* **4**, 682 (2016).
26. D.-S. Um, S. Lim, Y. Lee, H. Lee, H. Kim, W.-C. Yen, Y.-L. Chueh, and H. Ko, *ACS Nano* **8**, 3080 (2014).
27. A. R. Stokes *Proc. Phys. Soc.* **61**, 382 (1948).
28. G. Jones and E. P. O'Reilly, *IEEE J. Quantum Electron.* **29**, 1344 (1993).
29. L. B. Freund and S. Suresh, *Thin Film Materials: Stress, Defect Formation and Surface Evolution* (Cambridge University Press, 2003).
30. J. Martín-Sánchez, R. Trotta, A. Mariscal, R. Serna, G. Piredda, S. Stroj, J. Edlinger, C. Schimpf, J. Aberl, T. Lettner, J. Wildmann, H. Huang, X. Yuan, D. Ziss, J. Stangl, and A. Rastelli, *Semicond. Sci. Technol.* **33**, 013001 (2018).
31. J. W. Christopher, M. Vutukuru, D. Lloyd, J. S. Bunch, B. B. Goldberg, D. J. Bishop, A. K. Swan, arXiv:1803.02787, (2018).

Figure Captions:

Fig. 1: Schematic illustration of the gas pressure cell used to introduce biaxial tensile strain, loaded with a NM bonded on a PI film. The curvature of the PI film is exaggerated to illustrate the effect of the applied gas pressure.

Fig. 2: In-plane biaxial tensile strain $\epsilon_{xx} = \epsilon_{yy}$ introduced in a 100-nm-thick InGaAs NM measured by XRD as a function of applied gas pressure. Inset: normalized X-ray diffractograms recorded at a pressure of 0 and 552 kPa.

Fig. 3: Room-temperature PL spectra of the InGaAs NM of Fig. 2 measured for different values of the applied biaxial tensile strain.

Fig. 4: (a) Solid lines: calculated bandgap wavelengths between the conduction band and the strain-split HH and LH valence bands of $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ plotted as a function of biaxial strain. Symbols: emission-edge wavelengths obtained from the PL spectra of Fig. 3 as described in the text. Inset: schematic illustration of the band structure of unstrained (left) and tensilely strained (right) InGaAs. (b) Integrated output PL intensity obtained from the spectra of Fig. 3 plotted as a function of strain. Inset: strain dependence of the magnitude squared of the TE component of the conduction-to-LH momentum matrix element of $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$, normalized to the unstrained bulk momentum matrix element squared M_b^2 .¹⁸







