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Flexible nanomembrane photonic-crystal cavities for tensilely strained-germanium light emission

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Flexible photonic-crystal cavities in the form of Si-column arrays embedded in polymeric films are developed on Ge nanomembranes using direct membrane assembly. The resulting devices can sustain large biaxial tensile strain under mechanical stress, as a way to enhance the Ge radiative efficiency. Pronounced emission peaks associated with photonic-crystal cavity resonances are observed in photoluminescence measurements. These results show that ultrathin nanomembrane active layers can be effectively coupled to an optical cavity, while still preserving their mechanical flexibility. Thus, they are promising for the development of strain-enabled Ge lasers, and more generally uniquely flexible optoelectronic devices. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4954188]
important step towards the demonstration of Ge lasers where optical gain is enabled entirely through the introduction of tensile strain. The same device geometry developed in this work could also be applied to other NM laser active materials, including traditional direct-bandgap III-V compounds. In particular, compared to prior work involving membrane assembly for semiconductor laser development, the samples produced with this approach can feature extreme mechanical flexibility, including the ability to sustain complex modes of deformation (e.g., bending, folding, and twisting) without damage. The resulting devices could therefore be employed in environments that are not suitable to traditional rigid semiconductor lasers, for applications in flexible optoelectronic systems. Stretchable photonic-crystal slabs are also interesting for use as mechanically tunable optical filters.

The Ge NMs used in this work are fabricated from a commercial Ge-on-insulator (GOI) wafer (from IQE Silicon Ltd.), by removing the buried oxide layer with a wet etch in a hydrofluoric-acid solution. To facilitate the NM release, etchant access holes are first patterned in the GOI Ge layer using photolithography and reactive ion etching (RIE). The released Ge NM is then bonded onto a flexible polyimide (PI) film using a wet transfer procedure. The cavity fabrication and assembly process is illustrated in Fig. 1. First, a periodic array of single-crystal Si pillars is patterned in the Si device layer of a Si-on-insulator (SOI) substrate using electron beam lithography and RIE, with the SOI buried oxide layer used as the etch stop. Next, the array is spin-coated with a film of photoresist PI, and the underlying oxide is completely dissolved with a selective wet etch. Once again, etchant access holes (patterned by photolithography through the PI) are used to facilitate this step. After the wet etch, the PI film containing the pillar array is released from the Si substrate, dipped in methanol for about 1 min (to release any built-in strain that could otherwise cause it to roll up), and then transferred onto a previously prepared Ge NM on PI. A two-step baking process (at 60° for 10 min and then 110° for 15 min) is finally performed to remove first any methanol residue and then water between the Si pillar array and the Ge NM. The latter process is found to be critical to obtain a strong adhesion between the two membranes. The adhesion can be further enhanced by spinning an additional layer of PI over the entire sample.

By virtue of their disconnected nature within the embedding polymer film, the resulting column arrays can be stretched to extremely high levels of tensile strain while still preserving their structural integrity and periodic arrangement. Therefore, when combined with an ultrathin NM active layer, these arrays can provide strong vertical confinement and in-plane waveguiding of the emitted light (if their thickness and spatially averaged refractive index are sufficiently large), without at the same time limiting the maximum strain that can be introduced in the NM. Furthermore, in-plane optical feedback and vertical outcoupling of the NM emission can be obtained through diffraction by the pillars, so that the sample geometry of Fig. 1 is promising for the development of band-edge photonic-crystal lasers.

It should also be noted that the pillars in these samples consist of high-quality crystalline Si (the SOI template material), and therefore provide negligible absorption losses for the Ge luminescence. This feature represents an important advantage over other possible methods for the fabrication of similar arrays on ultrathin NMs, such as the deposition and nanopatterning of high-index amorphous dielectric materials. For instance, in recent work we have developed periodic arrays of amorphous-Ge pillars on Ge NMs using electron-beam lithography, electron-beam evaporation, and liftoff. These arrays were used to demonstrate efficient vertical outcoupling of the long-wavelength strained-Ge luminescence, which is otherwise primarily emitted along the plane of the NM due to its predominant TM polarization. At the same time, however, they could not serve as suitable optical cavities for laser development, because of the large optical losses (≈10⁴ cm⁻¹) of amorphous Ge at the strained-Ge emission wavelengths, caused by band-tail absorption. In fact, only weak signatures of cavity modes could be observed in those samples.

The structural quality of the Si-pillar arrays fabricated in the present work is illustrated in Fig. 2. The optical micrograph of Fig. 2(a) shows an entire array (the dark hexagonal shape) after transfer onto a Ge NM, and very good adhesion between the two membranes can be inferred from the lack of bubble-like features anywhere (the small circles seen in this image are the aforementioned etchant access holes). In the higher-magnification image of Fig. 2(b), the individual pillars can be resolved, showing that the array periodicity is perfectly preserved within the PI membrane. Finally, a scanning electron microscopy (SEM) image of a similar array after the RIE step is shown in Fig. 2(c), where highly uniform pillars with straight and smooth sidewalls are observed.

For the strain-dependent PL measurements, the PI films with the attached NMs are used to seal a metallic chamber which is then pressurized with a controlled gas inflow. This straining procedure is particularly convenient for the purpose of initial device development, while for practical applications similar results can in principle be obtained with integrated devices on chip, e.g., using MEMS technology, piezoelectrics, or microfluidics. The biaxial tensile strain introduced in the Ge NM is measured as a function of applied gas.
pressure using Raman microscopy. Representative results measured on an array-coated 50-nm-thick Ge NM are shown in Fig. 2(d), where the average strain and related error bar for each value of the applied stress were obtained from five random sites on the NM. These data are similar to previous Raman microscopy results from uncoated Ge NMs of similar thickness. The implication is that the overlying PI film with the embedded Si-pillar array does not appreciably limit the NM flexibility, as we also expect from a simple analysis of the relative moduli. Therefore, the array-coated NMs can be strained to a level where significant enhancements in the Ge radiative efficiency are expected.

Figures 3(a)–3(c) show the room-temperature PL spectra of three different photonic-crystal devices for a range of strain values. In each device, the Ge NM is 50-nm thick and nominally undoped. The Si-pillar arrays are embedded in a 4-μm-thick PI film and consist of a triangular lattice with different periods \( a \) and column diameters \( D \), while the pillar height is 500 nm in each sample. The specific values of \( a \) and \( D \), as determined from SEM images, are listed in the figure caption. For comparison, in Fig. 3(d) we show the strain-dependent emission spectra of an identical device where the PI film does not contain any pillar array. The PL measurement setup is described in the supplementary material. As shown in Fig. 3, a substantial enhancement in the PL intensity of each sample is obtained with increasing strain, together with a large red shift in the emission spectra. These observations are, of course, consistent with the decrease in direct-bandgap energy caused by tensile strain in Ge.

The strong coupling between the photonic-crystal cavities and the underlying NMs is clearly highlighted by the complex pattern of well resolved emission peaks observed in the PL spectra of Figs. 3(a)–3(c). In contrast, the output spectra of the unpatterned device of Fig. 3(d) simply consist of two overlapping broad features associated with the strain-split heavy-hole and light-hole valence bands, similar to previous measurements with uncoated NMs. The relatively narrow multiple emission peaks of Figs. 3(a)–3(c) can therefore be attributed to the \( \Gamma \)-point cavity resonances of the Si-pillar photonic-crystal slabs. This conclusion is substantiated by extensive numerical simulations of the photonic band structure of these cavities, described in detail in the supplementary material. First, simulations based on a slab-waveguide approximation indicate that each sample in Figs. 3(a)–3(c) supports three guided transverse modes (two with predominantly TE and one with predominantly TM polarization). These modes are primarily confined in the photonic-crystal slab, due to its relatively large average refractive index and thickness. At the same time, they have an appreciable spatial overlap with the underlying Ge, with calculated confinement factors in the NM in the range of 3% to 7%. Therefore, they can all be excited by the Ge luminescence. The dispersion curves of all three transverse modes were then computed using the two-dimensional plane-wave-expansion method (PWEM), and the results are presented in the supplementary material. A fairly good

Fig. 3. (a)–(c) Strain-resolved room-temperature PL spectra of three photonic-crystal samples with different column periods \( a \) and diameters \( D \). (a) \( a = 1060 \text{ nm}, D = 850 \text{ nm} \); (b) \( a = 1170 \text{ nm}, D = 900 \text{ nm} \); and (c) \( a = 1340 \text{ nm}, D = 950 \text{ nm} \). Inset: zoom-in of the features within the black ellipse. The arrow in each plot indicates the calculated photon energy of the main TM-polarized \( \Gamma \)-point resonance. (d) Strain-resolved room-temperature PL spectra of a Ge NM coated with a PI film without any Si-pillar array. For clarity, the spectra in each plot [but not in the inset of (c)] are shifted vertically relative to one another, in order of increasing strain.
agreement is obtained between the calculated $\Gamma$-point cavity resonance wavelengths within the strained-Ge emission band and all the measured luminescence peaks, with a maximum discrepancy of less than 3% (which can be mostly attributed to the lack of material dispersion in the PWEM simulations$^{40}$).

In particular, the arrow in each plot of Figs. 3(a)–3(c) indicates the calculated wavelength of the main TM resonance of the corresponding cavity. This feature is especially important, because the strain-enhanced Ge luminescence is predominantly TM polarized (particularly at long wavelengths), and therefore will preferentially excite TM cavity modes. As a result, in each photonic-crystal sample of Fig. 3 a particularly pronounced emission peak is observed very close to the calculated $\Gamma$-point TM resonance indicated by the arrow. The expected red shift of this feature with increasing periodicity of the pillar array is clearly observed in the figure. In fact, the whole high-strain PL spectra also broaden with increasing array period, with their long-wavelength edge increasing roughly from 2.0 to 2.1 towards 2.2 $\mu$m in Figs. 3(a)–3(c), respectively. This observation supports the notion that the measured luminescence involves the direct excitation and subsequent outcoupling of the photonic-crystal cavity modes. As the period is increased, these modes are collectively red shifted so that more and more light can be efficiently emitted at longer and longer wavelengths within the strained-Ge emission band. Interestingly, the cavity resonances can also be tuned by increasing the applied gas pressure, which increases the separation between neighboring pillars in the array (i.e., the period) by the same fractional amount as the tensile strain introduced in the Ge NM. This behavior is illustrated in the inset of Fig. 3(c), where a $\sim 0.5\%$ increase in strain produces a proportional red shift in the peak wavelength.

The strain-induced enhancement in PL efficiency can be estimated from the output power spectral density plots $P(h\nu)$ of Fig. 3, by comparing the emitted photon flux (proportional to $\frac{dh\nu}{dh\nu}/h\nu$, where $h\nu$ is the photon energy) with and without stress. For the unpatterned device of Fig. 3(d), this is obtained with the device of Fig. 3(c), whose main TM mode (indicated by the arrow) is especially well matched with the TM-polarized strained-Ge emission (peaked around 2 $\mu$m for the maximum strain levels near 1.5% considered in this figure$^{27}$).

Finally, we note that, while the cavity resonances of Fig. 3 are generally well resolved, they also remain relatively broad, with measured quality factors of less than 100. Similar values were reported in prior measurements of spontaneous emission from (rigid) III–V semiconductor photonic-crystal slabs.$^{44}$ No evidence of spectral narrowing was observed with the devices of Fig. 3, even though carrier densities near transparency are expected at the high strain and pumping conditions used in these measurements.$^{15,40}$ The underlying losses cannot be attributed to optical absorption in the array, either by free carriers or by band-tail states, as the pillars consist of nominally undoped high-quality crystalline Si. Instead, we believe that the main limiting factor is provided by scattering losses in the Ge NMs, arising from defects in the GOI template material. Such scattering centers can be seen in the microscope image of Fig. 2(a), where they appear as randomly positioned bright-color spots. Similar distributions of defects (with typical separations on the order of tens of microns) are also observed in the unprocessed GOI samples. The implication is that the structural quality of the GOI Ge film is of critical importance in the development of strained-Ge NM lasers. In fact, as shown in a recent report,$^{30}$ reducing the defect density in the Ge film would also allow for the introduction of higher levels of tensile strain, as defects tend to act as crack initiation sites under mechanical stress.

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40See supplementary material at http://dx.doi.org/10.1063/1.4954188 for a detailed description of the simulations of the photonic-crystal cavity modes, the strain measurements by Raman microscopy, the photoluminescence measurement setup, and the lasing condition for the device structures developed in this work.
41For the PI film thickness of 4 μm and refractive index near 1.7, Fabry-Perot fringes with a spacing of approximately 100 meV in photon energy may also be introduced in the PL spectra. However, no obvious modulation with such period is observed in the spectra of Fig. 3 [including the spectra from the unpatterned device of Fig. 3(d)], indicating weak multiple reflections in these PI films.