Optically pumped GeSn micro-disks with 16% Sn lasing at 3.1 μm up to 180 K

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Recent demonstrations of optically pumped lasers based on GeSn alloys put forward the prospect of efficient laser sources monolithically integrated on a Si photonic platform. For instance, GeSn layers with 12.5% of Sn were reported to lase at 2.5 μm wavelength up to 130 K. In this work, we report a longer emitted wavelength and a significant improvement in lasing temperature. The improvements resulted from the use of higher Sn content GeSn layers of optimized crystalline quality, grown on graded Sn content buffers using reduced pressure CVD. The fabricated GeSn micro-disks with 13% and 16% of Sn showed lasing operation at 2.6 μm and 3.1 μm wavelengths, respectively. For the longest wavelength (i.e., 3.1 μm), lasing behavior was demonstrated up to 180 K, with a threshold of 377 kW/cm² at 25 K. *Published by AIP Publishing.*

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Germanium tin (GeSn) is a Complementary Metal Oxide Semiconductor (CMOS)-compatible group IV material which can exhibit a direct bandgap,^{1,2} leading to potential applications in photonics³ and microelectronics.⁴ Recent progresses in Chemical Vapor Deposition (CVD)⁵ allowed the demonstration of the first GeSn laser in 2015.³ Later on, high Sn content Ge_{1-x}Sn_x layers with 8.5% to 12.5% of Sn lasing at wavelengths up to 2.5 μm and at temperatures up to 130 K were demonstrated by the same group.⁶ The lasing operation was confirmed in a double heterostructure configuration using a strained Ge cap layer.⁷ However, for such group-IV lasers to become an alternative to a complex integration of III-V lasers on Silicon (Si), their operating temperature is to be substantially increased.

To increase the operating temperature of group-IV semiconductor lasers, two paths are considered promising and are being currently investigated: GeSiSn alloys^{8,9} and tensile strain.^{10–12} However, a straightforward improvement of the GeSn material itself should not be neglected: a higher Sn content in GeSn layers increases the energy difference between the L and the Γ valleys^{5,9,13} and shifts the emitted wavelengths toward the Mid-Infrared (MIR).^{9,14} The wavelength range between 3 μm and 3.5 μm is indeed interesting for sensing applications. There, the atmosphere transparency window overlaps with absorbing lines of various gases.^{15,16} Similarly to the short wave IR (SWIR), standalone III-V lasers have been developed for the MIR,^{17–19} and they are not compatible with CMOS foundries. Therefore, GeSn materials are promising candidates for group IV laser sources fully integrated in SWIR and MIR Si photonic systems, both for sensing applications and on-chip short range optical interconnects.^{20–22}

The challenge in increasing the Sn content in GeSn is to preserve the crystallographic quality of the material. In this

work, we demonstrate lasing operation up to 180 K with optically pumped 16% Sn GeSn microdisk cavities emitting at 3.1 μm . We attribute the significantly higher lasing temperature to (i) a stronger band offset between the L- and the Γ -valleys stemming from a higher Sn content, (ii) a better crystalline quality of the GeSn layers grown on top of step-graded GeSn buffers, and (iii) the carrier confinement inside the lowest bandgap layer, preventing recombination in the more defective GeSn bottom layers.²³

Thick GeSn layers (well above the critical thickness for plastic relaxation) were grown in a 200 mm Epi Centura 5200 Reduced Pressure-Chemical Vapor Deposition (RP-CVD) cluster tool from Applied Materials.^{14,24,25} Digermane (Ge₂H₆) and tin-tetrachloride (SnCl₄) were used as low temperature precursors. All growth processes, including those of the step-graded GeSn buffers, were carried out on top of Ge Strain Relaxed Buffers (SRBs),²² reducing the lattice mismatch to the Si(001) substrates. Four configurations have been investigated: (i) a $\sim$ 430 nm thick GeSn active layer with nominally 13% of Sn (**sample A**), (ii) a 430 nm thick GeSn active layer with nominally 16% of Sn (**sample B**), (iii) a $\sim$ 200 nm thick GeSn 13% active layer grown on top of a GeSn step-graded buffer made of GeSn 8% and 9.5% layers ($\sim$ 110 nm thick each) (**sample C**), (iv) a $\sim$ 180 nm thick GeSn 16% active layer grown on a GeSn step-graded buffer made of GeSn 8%, 9.5%, and 13% layers ($\sim$ 110 nm thick each) (**sample D**) (Fig. S1, [supplementary material](#)). Sn incorporation in the GeSn layers was controlled by changing the growth temperature.^{14,24,26} In the case of Ge_{0.87}Sn_{0.13} layers (sample A), 95% of the 200 mm wafer area presents smooth/mirror-like surface under grazing light with no significant Sn segregation. For Ge_{0.84}Sn_{0.16} layers (sample B), the useful area reduces to around 30% of the 200 mm wafer surface. In the case of the GeSn layers grown on step-graded buffers, the layers were mirror-like over the whole wafer surface. The use of a GeSn step-graded

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TABLE I. Layer properties and band structure parameters calculated at 300 K for micro-disks before and after strain relaxation. (*) Strain in partially suspended micro-disks measured by Raman spectroscopy (supplementary material).

Micro-disks fabricated on			Before strain relaxation		After strain relaxation	
Sample	Growth buffer	x_{Sn} (%)	ε_{II} (%)	$E_{\text{GL}}-E_{\text{GR}}$ (eV)	ε_{II} (%)	$E_{\text{GL}}-E_{\text{GR}}$ (eV)
A	Ge SRB	13.07	-0.56	0.030	$\sim 0^*$	0.095
B	Ge SRB	16.41	-0.76	0.063	$\sim 0^*$	0.152
C	Step graded GeSn	12.87	-0.66	0.015	$\sim 0^*$	0.092
D	Step graded GeSn	16.05	-0.80	0.053	$\sim 0^*$	0.146

structure improved the quality of the stacks, especially for very high Sn concentrations (i.e., 16%).²³ All optical characterizations were performed on mirror-like areas.

X-ray diffraction (XRD) measurements indicate that the top GeSn layers are partially relaxed (see the [supplementary material](#)). The residual compressive strain and the Sn content of the top layers (see Table I) were extracted from Omega-2Theta scans around the (004) XRD order and reciprocal space maps (RSM) around the (224) order (Fig. S1, [supplementary material](#)) taking into account the small positive deviation from a straightforward interpolation between the lattice parameter of pure Ge and Sn.²⁷ The GeSn top layers were slightly less relaxed, and the residual compressive strain was higher for step-graded samples (samples C and D) than for their SRB counterparts (samples A and B). To fully relax the residual strain, suspended micro-disks were fabricated in those layers.^{6,28} Strain values at the edges of suspended micro-disks were measured by Raman spectroscopy (Fig. S2, [supplementary material](#)). The residual strain and the Sn content in the GeSn layers laying on top of the four samples are provided in Table I for unsuspended layers (before elastic strain relaxation) and for suspended micro-disks (i.e., after strain relaxation). The theoretical bandgap was determined using the conventional deformation potential theory,^{29,30} with a linear interpolation between the deformation potential of pure Ge and pure Sn^{3,9} and a bowing parameter at 2.4 eV found in Ref. 12. The theoretical bandgaps for the L (E_{GL}) and Γ valleys (E_{GR}) at 300 K are added in Table S1 of the [supplementary material](#). A good agreement is found with the measured Γ bandgaps (E_{GR}) at 300 K provided in Table S1 ([supplementary material](#)). The larger compressive strain in samples C and D using a step-graded buffer results in a smaller energy difference between the L and Γ bandgaps, of only 15 meV in sample C compared to 30 meV in sample A and 53 meV in sample D compared to 63 meV in sample B. However, a larger Sn content of the samples D and B compared to samples C and A favorably increases energy differences between the L and Γ bandgaps, to compare with $k_{\text{B}}T = 25$ meV at room temperature. During the fabrication of micro-disks, the residual compressive strain is released, increasing the difference between the L and Γ bandgaps to around 90 meV for micro-disks with 13% Sn and to around 150 meV for micro-disks with 16% Sn (Table I).

We evaluated the optical quality of our layers and the directness of the bandgaps through temperature-dependent photoluminescence (PL) (see [supplementary material](#)). Figures 1(a)–1(c) show the integrated intensity, the energy,

and the Full Width at Half Maximum (FWHM) of the PL peaks extracted from the spectra provided in Figs. S3a–S3d of the [supplementary material](#), respectively. The integrated intensity and FWHM from a 0.7 μm thick reference Ge layer³¹ showing indirect band-gap behaviour has been added for comparison in Fig. 1(a). When the measurement temperature decreases from 300 K to 20 K, the Γ -valley luminescence of all GeSn samples strongly increases. This trend is a clear indication that the bandgap is direct.³ By contrast, the PL intensity of the Ge on the Si sample, therefore with an indirect bandgap, is much lower and decreases when the sample is cooled-down. Interestingly, the integrated PL intensity of sample C (GeSn 13% layer on a GeSn step-graded buffer) increases significantly only for temperatures below 150–200 K. This is likely due to the very small energy difference between the L and Γ valleys, i.e., only 15 meV (Table I), allowing for thermal escape of electrons from the Γ conduction band valley into the L valley at room temperature ($k_{\text{B}}T \sim 25$ meV). An additional proof of direct bandgap

FIG. 1. (a) Integrated PL intensity, (b) energy, and (c) full width at half maximum of the PL peaks as a function of the measurement temperature for samples A–D (Table I) together with the integrated PL intensity of a Ge SRB grown on Si in the same CVD reactor.

FIG. 2. (a) 45° tilted view of the SEM image of a GeSn micro-disk; (b) zoom on the etched sidewall for the same 45° tilt, and (c) a 5° tilt view of that micro-disk.

behavior is a significantly larger FWHM at room temperature for sample C: a shoulder in the PL spectra is indeed observed (Fig. S3, [supplementary material](#)), which is likely coming from the L-valley.

Using the GeSn layers described earlier as the active media, we fabricated suspended micro-disk optical resonators.^{6,28} This approach enables to (i) increase the offset between Γ and L valleys of the active GeSn layers by relaxing the strain (Table I) and (ii) confine optical modes in the GeSn layers. The layers were patterned using e-beam lithography and an anisotropic dry etching with Cl_2/N_2 gasses. The GeSn layers were then under-etched using a selective dry etching recipe based on CF_4 .^{32,33} The dry etching recipe selectively etched the Ge SRBs and the 8% Sn layers, leaving intact the higher Sn content GeSn layers above. This way, most of the misfit dislocations at the Ge/GeSn interfaces were removed. The thickness of sample C after the under-etching step (Sn 13% on the step graded GeSn buffer) is then 305 nm, to be compared to ~ 430 nm of sample A (Fig. S3 inset, [supplementary material](#)). The 400 nm thickness of sample D after the under-etching step (composed by

the 180 nm thick GeSn 16% active layer, the two ~ 110 nm thick GeSn 13% and 9.5% layers) is by contrast close to the 430 nm thickness of sample B (Fig. S3 inset, [supplementary material](#)). The surface of the micro-disks was not passivated. Figure 2 shows tilted Scanning Electron Microscopy (SEM) views of a typical micro-disk with an $8\text{ }\mu\text{m}$ diameter, smooth sidewall [Figs. 2(a) and 2(b)] and a $3.8\text{ }\mu\text{m}$ under-etch [Fig. 2(c)].

To test the lasing performances of the micro-disks, we used a 1064 nm Nd:YAG pulsed pump laser with a pulse width of 0.6 ns and a repetition rate of 50 kHz. The pump was focused on micro-disks with a spot diameter of $8\text{ }\mu\text{m}$. Figure 3(a) shows the laser-output versus pumping-laser-input (L-L) for a $8\text{ }\mu\text{m}$ micro-disk fabricated in the GeSn layer with 16% Sn content grown on the GeSn step-graded buffer (i.e., sample D), together with emission spectra at three different pumping levels at 25 K. The lasing threshold is measured at 377 kW/cm^2 at 25 K. Figure S4 ([supplementary material](#)) presents the lasing spectra of all samples at 25 K at their corresponding threshold power. Whispering-gallery-modes were identified by measuring the free spectral range (FSR) at 102 nm in different spectra. A good agreement was obtained with the calculated FSR from the group index of the first mode calculated using Lumerical software modelling. The quality factor of the cavities could not be extracted as the linewidth of the laser peak is limited by the resolution of the spectrometer. Figure 3(b) shows laser spectra evolution as a function of temperature for sample D. The spectra from 50 K to 200 K were acquired with a lower spectral resolution. As the temperature increases from 50 K to 180 K, a mode switching was observed. Indeed, the optical gain shifted rapidly with the temperature, while the cavity modes evolved at a much slower rate. At 200 K, a clear broadening of the emitted spectrum is observed compared to lower temperature spectra. The spectra of Fig. 3(b) have been measured at 4.3 MW/cm^2 , and L-L curves at 180 K are shown in Fig. 3(c). We attribute the high temperature working operation improvement to a larger energy splitting between Γ - and L-valleys in higher Sn-content layers (16% in our case compared to 12.5% in Refs. 3, 6, and 7). We expect the thresholds of our lasers to be reduced using surface passivation and carrier confinement. In addition,

FIG. 3. GeSn micro-disk characterization: (a) L-L curve of a $8\text{ }\mu\text{m}$ micro-disk fabricated in the 16% GeSn layer grown on the GeSn graded buffer at 25 K (sample D); (b) Evolution of the laser spectra as a function of temperature (spectra from 50 K to 200 K acquired with a low spectral resolution); (c) L-L curve of a $8\text{ }\mu\text{m}$ micro-disk fabricated in the 16% GeSn layer grown on the GeSn graded buffer at 180 K (sample D).

capping of the top GeSn layers with lower Sn content layers (i.e., the use of double heterostructures) should enhance carrier confinement.

In summary, we have studied partially relaxed GeSn layers with very high Sn contents (up to 16%) grown either on Ge SRBs or on step-graded GeSn buffers. Both buffer strategies led to a direct bandgap for Ge_{0.87}Sn_{0.13} and Ge_{0.84}Sn_{0.16} layers. After under-etching, both buffer strategies yielded optically pumped laser emission at 2.6 μm (for 13% of Sn) and 3.1 μm (for 16% of Sn). However, only GeSn layers grown on step-graded buffers presented mirror-like surfaces under grazing light with no significant Sn segregation over the whole 200-mm wafer surface, and only a combination of 16% Sn GeSn layers and step-graded GeSn buffers led to a laser operation behavior up to 180 K. The Ge_{0.84}Sn_{0.16} microdisk cavities emitted at 3.1 μm with a lasing threshold of 377 kW/cm² at 25 K. An increase of the Sn content grown on GeSn step-graded buffers is, thus, a path towards room-temperature GeSn lasers.

See [supplementary material](#) for XRD and PL characterizations of GeSn layers, strain characterization via Raman spectroscopy and lasing spectra, and threshold power analysis at 25 K.

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