Non-isothermal phase-field simulations of laser-written in-plane SiGe heterostructures for photonic applications

Ozan Aktas1✉, Yuji Yamamoto2, Mehmet Kaynak2 & Anna C. Peacock1✉

Advanced solid-state devices, including lasers and modulators, require semiconductor heterostructures for nanoscale engineering of the electronic bandgap and refractive index. However, existing epitaxial growth methods are limited to fabrication of vertical heterostructures grown layer by layer. Here, we report the use of finite-element-method-based phase-field modelling with thermocapillary convection to investigate laser inscription of in-plane heterostructures within silicon-germanium films. The modelling is supported by experimental work using epitaxially-grown Si0.5Ge0.5 layers. The phase-field simulations reveal that various in-plane heterostructures with single or periodic interfaces can be fabricated by controlling phase segregation through modulation of the scan speed, power, and beam position. Optical simulations are used to demonstrate the potential for two devices: graded-index waveguides with Ge-rich (>70%) cores, and waveguide Bragg gratings with nanoscale periods (100–500 nm). Periodic heterostructure formation via sub-millisecond modulation of the laser parameters opens a route for post-growth fabrication of in-plane quantum wells and superlattices in semiconductor alloy films.

1 Optoelectronics Research Centre, University of Southampton, Southampton, UK. 2 IHP-Leibniz-Institut für Innovative Mikroelektronik, Frankfurt (Oder), Germany. ✉email: O.Aktas@soton.ac.uk; acp@orc.soton.ac.uk
Modern-day applications of microelectronics, optoelectronics, and photonics have become undoubtedly possible with the technological advancements in material growth and formation of interfaces between different materials. For example, the development of semiconductor heterostructures, quantum wells, and superlattices, which provide confinement for light and carriers via spatially engineered refractive index and bandgap profiles, has enabled the fabrication of various devices such as heterojunction bipolar transistors, double heterojunction lasers, quantum cascade lasers, distributed Bragg reflectors, semiconductor saturable absorber mirrors, and modulators. Methods commonly used to grow epitaxial layers of semiconductors are molecular beam epitaxy and derivatives of chemical vapor deposition (CVD), which allow for material growth with atomic precision and control. However, these methods implement a layer-by-layer growth approach; therefore, they can only be used to fabricate vertical heterostructures and superlattices, which are limited in total thickness (≤5 μm) due to the constraint of lattice matching and strain issues. Recently, fabrication of lateral heterostructures has been demonstrated in semiconductor alloys by laser processing of amorphous SiGe films. Generally, phase segregation is an undesired phenomenon for epitaxial growth of heterostructures in semiconductor alloy epilayers. This is particularly true for silicon-germanium (SiGe) alloys, which have a large gap between the liquidus and solidus curves in the phase diagram. For stable growth from a melted SiGe alloy where phase segregation results in a Ge-rich liquid phase, solidification speeds have to be smaller than a critical speed, which depends on the local temperature gradient and thermophysical properties of the alloy. Otherwise, constitutional undercooling can emerge ahead of the solid/liquid interface, breaking its stability. Consequently, the interface instability results in cellular or faceted structures during bulk crystal growth, and dendritic structures during laser annealing of SiGe films on glass substrates, leading to a random redistribution of the initial composition. To exploit heat-induced phase segregation for the fabrication of regular in-plane heterostructures with well-defined morphologies, cellular, or dendritic solidification of alloy thin films needs to be avoided by ensuring either low growth speeds or high temperature gradients at the liquid/solid interface. However, low solidification speeds, on the order of 10 μm s⁻¹, allow for stable growth, are impractical in terms of process time. Another alternative is the application of non-conventional thermal sources, like lasers, to post-process semiconductor alloy thin films grown on substrates with high thermal conductivities. Such methods allow for achieving extremely high temperature gradients (10⁶ K m⁻¹), which are required to ensure the stability of the solid/liquid interface. Recently, by using laser processing of amorphous SiGe thin films on silicon substrates, the fabrication of polycrystalline SiGe microstripes was demonstrated with compositional profiles that can be tuned by applying different scan speeds in the range of 0.1–100 mm s⁻¹. Additionally, it is known from classical theory of solidification that transient effects such as acceleration of the liquid/solid interface can be used to form compositional microstructures, and this was recently demonstrated by laser processing of SiGe core silica cladding fibers.

Over the past two decades, phase-field models have been extensively applied to study the formation of cellular and dendritic structures during solidification of alloys, where a phase-field parameter ϕ(r, t) is used to track the liquid/solid interface. Here, we extend these studies to show that it can also be exploited for modeling non-dendritic solidification of laser-melted semiconductor alloys. A three-dimensional (3D) finite-element-method (FEM)-based phase-field model was used to investigate non-isothermal solidification with thermocapillary convection, and motional effects on phase segregation at the moving liquid/solid interface. The simulation results are supported with experimental data, which were obtained by optical microscopy and scanning transmission electron microscopy (STEM) with energy-dispersive X-ray spectroscopy (EDX), using Si₉₀Ge₁₀ alloy thin films epitaxially grown on silicon substrates. Based on this, we introduce a concept for post-process laser writing of in-plane heterostructures with single or periodic interfaces in semiconductor alloy epilayers. Our work focuses on SiGe epilayers because of the increasing interest in Ge/SiGe heterostructures in micro/nanostructures, and their tantalizing applications, and ii) waveguide Bragg gratings for wavelength filtering in the mid-IR spectrum.

**Results**

**Laser writing of in-plane longitudinal SiGe heterostructures with tunable compositionally-graded profiles at various constant scan speeds.** We begin our investigations of laser-driven heterostructure formation with some preliminary experimental work by applying laser processing on homogenous 575 nm thick SiGe epilayers (50% Ge). The Si₉₀Ge₁₀ epilayers were epitaxially grown on silicon substrates by reduced pressure CVD. For laser processing, we used a continuous wave laser operating at 532 nm and an experimental setup schematically shown in Supplementary Fig. 2. The laser beam with a power of 2.5 W was focused to a spot with a diameter of 5 μm on the surface of the Si₉₀Ge₁₀ epilayer by a 10× microscope objective. Laser processing was conducted at the same power with different constant scan speeds in the range of 0.1–200 mm s⁻¹. Scanning the stage under the fixed laser beam produces a traveling molten zone. Depending on the scan speed, the laser-driven liquid/solid interface produces microstripes solidified with varying Ge compositions, as shown in the optical microscope image in Fig. 1a. The laser-written SiGe microstripes for the low scan speeds of 0.1 and 1 mm s⁻¹ are barely distinguishable; however, wider microstripes emerge with a yellow color as the scan speed increases and Ge accumulates on the surface.

**Phase-field simulations of laser-written in-plane longitudinal SiGe heterostructures.** We carried out 3D FEM-based non-

COMMUNICATIONS PHYSICS | https://doi.org/10.1038/s42005-021-00632-1 | www.nature.com/commsphys
The other parameters used in the simulations were: optical power (red) travels along the scan direction dragging the Ge-rich liquid, and solidifies at the trailing edge with a scan-speed-dependent redistribution of the initial composition $x_0 = 0.5$. After a short Si-rich transient (blue), an indefinitely long steady-state region emerges with a Ge-rich ($x > 0.5$) strip core (yellow) and a Si-rich ($x < 0.5$) under-cladding. The phase-field modeling provides good qualitative agreement when compared to the experimental observations of the scan-speed-dependent Ge redistribution. For example, both the experimental data given in Fig. 1a and the simulation results given in Fig. 1b show that the Ge-rich strip core on the top surface widens as the scan speed increases. For further qualitative comparison, a laser-processed SiGe epilayer was cleaved in the middle, and its cleaved edge was polished at an angle to reveal the cross-section of a longitudinal heterostructure laser-written at a scan speed of $200 \text{ mm s}^{-1}$. The simulated redistribution of the composition on the top surface and in the cross-section (see Fig. 1c) agrees well with the experimental result shown in the optical microscope image in Fig. 1d. The Ge-rich top surface (yellow) and boundary of the Si-rich cladding (blue) are clearly observable via the natural color contrast.

The simulation results show that the laser beam absorbed by the SiGe epilayer increases the surface temperature up to 1475 °C, resulting in a molten zone and a thermocapillary-driven convection. The size of the molten zone, and thermocapillary-driven flow pattern were found to be quite similar for the experimental scan speed range, as the maximum experimental scan speed $v$ of 200 mm s$^{-1}$ is one order of magnitude smaller than the maximum flow speed $u_{max} = 2.3 \text{ m s}^{-1}$. In Fig. 2a, the phase-field parameter $\phi(r)$ shows the solid regions (blue) and molten zone (red) traveling at a scan speed of 1 mm s$^{-1}$. The solid/liquid interface (rainbow color) has a depth-dependent thickness ranging between 60–125 nm, due to the different thermal gradients, and mushy zone between the liquidus and solidus curves in the phase diagram. The thermocapillary convection within the molten zone pushes the liquid radially outwards from the center (see Fig. 2b), forming a Ge-rich stagnant layer (darker red) close to the liquid/solid interface, where the flow speed decreases to 0 m s$^{-1}$ from the maximum value of 2.3 m s$^{-1}$. Therefore, distribution of the composition in the cross-section and on the top surface of a laser-written microstripe. An in-plane longitudinal SiGe heterostructure was formed with a Ge-rich strip core and Si-rich under-cladding along the scan direction. The image size in the transverse direction was rescaled accordingly to match the width of the laser-written microstripe in d. d Optical microscope image showing the angle-polished cross-section and top facet of a longitudinal SiGe heterostructure laser-written at 200 mm s$^{-1}$ for qualitative comparison with the simulation result.

Fig. 1 Laser writing of in-plane longitudinal SiGe heterostructures via scan-speed-dependent spatial redistribution of the composition. a Optical microscope image showing laser-written microstripes on a Si$_{0.5}$Ge$_{0.5}$ thin film (epi-SiGe) epitaxially grown on Si substrates, for scan speeds in the range of 0.1–200 mm s$^{-1}$. b Results of finite-element-method (FEM)-based phase-field simulations for laser-driven phase segregation are given as composition color maps showing the Ge molar fraction $x(r)$ for the short transient (blue) and indefinitely-long steady-state regions (yellow/orange) solidified behind the laser-induced molten zone (red) traveling at a constant scan speed in the range of 1–500 mm s$^{-1}$. c Simulated spatial redistribution of the alloy composition in the cross-section and on the top surface of a laser-written microstripe. d Laser-written SiGe microstripes
the Ge-rich molten zone is not homogeneous (see Fig. 2c). At high scan speeds, a build-up of Ge occurs at the trailing edge (see Fig. 2d), because of insufficient diffusion-limited transport preventing complete mixing in the annular stagnant layer, as in the case for low scan speeds. By leveraging the scan-speed-dependent phase segregation, different Ge profiles in the laser-processed region can thus be obtained.

The Ge concentration profiles calculated by the phase-field simulations along different directions at the top surface and cross-section are given in Fig. 3a–c for the in-plane longitudinal SiGe heterostructures laser-written at different scan speeds. When the laser beam melts the surface and induces phase segregation at the liquid/solid interface, the excess Ge abruptly diffuses into the molten zone, due to the higher solubility of Ge in the liquid phase. As the laser beam moves in the scan direction, more Ge accumulates in the molten zone up to a steady-state composition. Therefore, the Ge concentration of the initially solidified material in the transient region is very low (see Fig. 3a). However, it increases remarkably as the steady-state composition is reached in the Ge-rich molten zone. The length of the transient region highly depends on the scan speed. The higher Ge content in the stagnant layer close to the liquid/solid interface can also be seen in Fig. 3a. A maximum Ge molar fraction \( x \) up to 0.7 mol is reached on the top surface in the steady-state region for the scan speed of 200 mm s\(^{-1}\). Higher scan speeds result in the levelling off of the Ge composition, widening the width of the Ge-rich strip core, as shown in Fig. 3b for the scan speed of 500 mm s\(^{-1}\). The phase-field gradient \( \nabla \phi \) has the highest value at the bottom of the molten zone, which increases the strength of phase segregation according to Eq. (20). Thus, more Ge diffuses away from the liquid/solid interface, and the most Si-rich region in the undercladding occurs at the bottom close to the SiGe/Si interface (see Fig. 3c). The effect of laser driven phase segregation on the dispersion of the Ge molar fraction can be clearly seen by calculating histograms of the composition within the entire solidified volume in the steady-state region for different scan speeds (Fig. 3d). These results complement our understanding of the formation of in-plane longitudinal heterostructures using laser processing.

Our FEM-based approach was not limited by the RAM memory (<100 GB), but by the total time (>2 days) required to run each phase-field simulation with thermocapillary convection. It is for this reason that we were only able to simulate laser processing using laser-written stripes with smaller sizes (2 \( \mu \)m wide and 10 \( \mu \)m long) compared to the experimentally measured sizes (5.7 \( \mu \)m wide and 4 cm long). Therefore, this does not allow for a one-to-one quantitative comparison between the experiments and the simulation results given in Fig. 3a–c. However, to reduce the total simulation time significantly so that these can be conducted with the experimental cross-sectional dimensions, the thermocapillary-driven convection can be neglected in the phase-field simulations by not incorporating the Navier–Stokes equations. This is justified as the absence of the convection (see Supplementary Fig. 3) would only result in a slightly higher Ge content (\( \Delta x < 0.02 \) mol) at the center of the top surface. This difference is experimentally insignificant as the achievable compositional resolution is typically on the order of 0.05 mol (5\%)\(^{28}\).

To conduct the comparison, the cross-section of a SiGe microstripe laser-written at a scan speed of 200 mm s\(^{-1}\), was analysed using STEM with EDX. A high-angle annular dark-field (HAADF) image, which was taken with \( \times 14000 \) magnification of the cross-section at 200 keV, shows the material contrast in Fig. 4a, due to the redistribution of the composition in the melted region, as given by the two-dimensional (2D) EDX map in Fig. 4b. Our simulated result, neglecting thermocapillary convection but using the actual experimental thickness of the SiGe epilayer (575 nm) and width of the laser-written stripe (5.7 \( \mu \)m), is shown in Fig. 4c, revealing the 2D redistribution of Ge in the cross-section. The 1D concentration depth profile was also calculated and compared with the EDX data in Fig. 4d, which was compensated for the presence of other impurity elements, such as oxygen and carbon. The simulated 2D and 1D Ge profiles agree quantitatively well with the EDX data, which has an error margin
Fig. 3 Simulation results showing the Si$_{1-x}$Ge$_x$ composition profiles along different directions on various surfaces, and dispersion of the composition for scan speeds in the range of 1–500 mm s$^{-1}$. Inset figures show the surface and direction (dotted lines) where the composition profile was taken. Ge molar fraction $x(r)$ along the a longitudinal and b transverse directions on the top surface, and c along the vertical direction at the cross-sections of laser-written SiGe microstripes. d Normalized volume histogram showing the scan-speed-dependent dispersion of the initial composition $x_0 = 0.5$ within the whole solidified volume in terms of the Ge molar fraction. The bin size for histograms is $\Delta x = 0.01$. Inset figure shows the laser-processed volume in the steady-state region, where the histograms were calculated.

Fig. 4 Elemental analysis of an in-plane longitudinal SiGe heterostructure laser-written at a scan speed of 200 mm s$^{-1}$. a High-angle annular dark-field image (HAADF) showing the material contrast at the cross-section of the laser-processed region. b Energy-dispersive X-ray spectroscopy (EDX) and c phase-field simulation results showing spatial redistribution of the alloy composition at the cross-section for 2D comparison. The color scale applies to both figures in b, c. d Quantitative comparison between the experimental (blue solid line) and simulated (red dashed line) Ge concentration along the depth profile across the cross-section, as marked by a white arrow in a. The experimental error margin in the EDX data is 5%.
of ±5%. It is worth noting that even in the case of laser processing at a scan speed of 200 mm s⁻¹, where the maximum compositional segregation occurs, the Ge concentration profile is smoothly changing across the cross-section, as can be seen in Fig. 4d. However, an abrupt transition occurs within a 25 nm range at the melt/solid boundary, jumping from the lowest Ge content (25%) to the initial Ge content (50%) of the SiGe epilayer.

Modal analysis of in-plane longitudinal SiGe heterostructures that function as laser-written graded-index waveguides. Similar to the double heterostructure semiconductor lasers, where optical guiding is achieved in a high-index core sandwiched between two lower index claddings, a single longitudinal heterostructure formed with a Ge-rich stripe core and a surrounding Si-rich cladding can provide confinement for light. To illustrate this concept, we calculated the optical modes supported by a longitudinal heterostructure laser-written at a scan speed of 200 mm s⁻¹ by a focused beam of 2 µm in diameter. The 2D Ge molar fraction at the cross-section of the laser-written SiGe microstripe, which was obtained by a phase-field simulation, is given in Fig. 5a. The corresponding refractive index profile \( n(x) \) at a wavelength of 2 µm was calculated by using \( n(x) = 3.451 + 0.2951x + 0.3419x^2 \), which is a quadratic function of the Ge molar fraction 38.

The process temperatures, which are high enough to melt the surface, result in biaxial tensile strains within the laser-processed SiGe films, due to the difference between the thermal expansion coefficients of Ge and Si. Residual tensile strains as high as 0.6% can be reached28. Therefore, photo-elastic effects, which are strain-induced changes in the refractive index \( n \), need to be incorporated39. We found that an increase of 3% in the refractive index within the laser-processed strained region gives the best agreement with the optical experiments in terms of matching the mode profiles. The resulting refractive index profile, which includes the photo-elastic effects, is shown in Fig. 5b. The simulated fundamental mode profiles for the transverse electric (TE) and transverse magnetic (TM) polarized light at a wavelength of 2 µm are shown in Fig. 5c, d, respectively.

For a proof-of-principle demonstration, we used a 1 cm long SiGe microstripe that was written using a laser with a spot size of 5 µm at a scan speed of 200 mm s⁻¹. The end facets were prepared for optical coupling by manual cleaving, and a 2 µm fiber laser was first free space coupled into the graded-index SiGe waveguide by using a 60× objective. The light at the output was then collimated by a 40× objective and imaged by an infrared camera to capture the images of the TE and TM modes shown in Fig. 5e, f, respectively. A schematic for the experimental setup used for optical coupling is given in Supplementary Fig. 4. In agreement with the optical simulations, we observed that only the fundamental TE and TM modes were guided through these laser-written SiGe waveguides at a wavelength of 2 µm. Moreover, optical guiding was observed even for the waveguide written at the lowest scan speed of 0.1 mm s⁻¹, where the least compositional segregation occurs within the laser-processed region. This observation confirms our assumption that the residual tensile strains increase the refractive index in the laser-processed regions to allow for optical guiding. The TM mode was found to be lossy; therefore, it leaks from the strip core decreasing the intensity of the propagating light. More confinement is necessary for the TM mode to achieve optical guiding at much longer wavelengths, such as in the molecular fingerprint regime (6–20 µm wavelength). The depth of the melted region is naturally limited by the thickness of the SiGe epilayer, which can be grown on a Si substrate with a thickness up to 10 µm using CVD methods. By increasing the laser power and using longer wavelengths (>532 nm), the deposited energy and optical penetration can be increased to deepen the melted region closer to the SiGe/Si interface.

Conventional waveguides depend on patterning of silicon-on-insulator (SOI) wafers by e-beam lithography and etching. Although this approach provides compact components for integrated photonics in the telecom range, the silica buffer oxide layer in SOI waveguides suffers from strong absorption above 3.6 µm, hindering photonic applications in the mid-IR range. Therefore, graded-index SiGe waveguides have recently received increased interest for mid-IR applications in integrated photonics40,41, due both to the low optical losses (1–2 dB cm⁻¹) in the wavelength range of 2–8 µm and their tunable optical properties via compositional engineering. However, these graded-index SiGe waveguides fabricated by epitaxial growth have two drawbacks: i) The graded refractive index can only be obtained in the growth direction perpendicular to the substrate surface, and a step index profile in the plane for mode confinement still requires patterning of the surface similar to the SOI waveguides. ii) Waveguides with different graded index profiles on the same
Substrate require different growth processes for each of them, increasing the cost and complexity of the fabrication. In contrast, for the laser-written graded-index SiGe waveguides, an index grading both in the transverse and vertical directions at the cross-section is simultaneously achieved via compositional segregation of the alloy within the melt region during laser processing. Moreover, this graded index profile is tunable by controlling the laser scan speed, allowing fabrication of customized waveguides using the same homogeneous SiGe epilayer grown on a Si substrate. A tunable index profile can be leveraged for dispersion engineering in nonlinear optical applications. Furthermore, compositional segregation of Si and Ge enables a refractive index difference $\Delta n$ on the order of $10^{-4}$, which is higher compared to those typically obtained ($10^{-6}$) in laser-written waveguides in bulk unary semiconductors via pulsed-laser-induced modifications. Furthermore, laser writing of SiGe waveguides does not require lithographic patterning, which can lead to scattering losses due to the roughness of the etched waveguide boundaries, and it can be used to write components with complex shapes such as directional couplers, Y-junctions, S-bends, and spirals, if 2-axis motorized stages are used.

Phase-field simulations of in-plane periodic transverse heterostructures (superlattices) fabricated by modulation of the laser processing parameters. As the steady-state region within the laser-written microstripes is indefinitely long (depending only on the total scan range), the laser processing parameters can be modulated to create transient effects superimposed on the constant scan speed of the stage during laser processing. To investigate the idea, we first performed simulations with the modulation of the stage scan speed to produce periodic heterostructures in the steady-state region (see Fig. 1b). A 77.6 kHz square wave modulation of the stage scan speed $v$ between 25–50 mm s$^{-1}$ with a duty cycle of 1/3 resulted in a SiGe superlattice with a period of $\Lambda = 430$ nm (see Fig. 6a). The resulting periodic composition profile is embedded into the Ge-rich strip core and diminishes close to the surrounding Si-rich cladding (down to a depth of 300 nm), as shown in the inset figure. The composition profile (blue) along the longitudinal direction on the top surface shows the profile of the Ge molar fraction, where the stationary background composition level is determined by the initial scan speed (see Fig. 3a). It is possible to obtain a higher background composition level up to the maximum Ge molar fraction of $x = 0.7$ by increasing the initial scan speed; however, this requires application of higher modulation frequencies to achieve the same period. To demonstrate a smaller period and a different back-ground composition level, we performed a 621.1 kHz modulation of the stage scan speed $v$ between 100–200 mm s$^{-1}$, which forms a SiGe superlattice with a period of $\Lambda = 220$ nm, as shown in Fig. 6b. Although smaller periods down to 100 nm could be achieved, the finiteness of the mesh (minimum 20 nm) limits the resolution of the simulated heterostructures.

Modulation of the scan speed by controlling the movement of the stage, where the SiGe sample is fixed, might be experimentally limited by the maximum achievable acceleration due to the inertia of the stage mechanism. Much faster motional transients for the laser-induced molten zone can be realized by modulating either power or position of the laser beam using electro-optic modulators/deflectors or acousto-optic scanners, while the stage moves independently at a constant scan speed. We performed 20% modulation (reduction) of the laser power $P_0 = 200$ mW with a duty cycle of 1/3 at 61.0 kHz to obtain an in-plane transverse SiGe superlattice with a period of $\Lambda = 402$ nm (see Fig. 6c). In this case, we were able to obtain a greater change in the Ge molar fraction in the range of $\Delta x = 0.55–0.78$ mol, via the higher acceleration of the solidification boundary when the spot size suddenly shrinks. Another approach for modulation is switching the position of the focused laser beam back and forth in the scan direction while the stage moves at a constant scan speed. A 61.1 kHz sawtooth modulation of the beam position with a movement of 200 nm resulted in a SiGe superlattice with a period of $\Lambda = 410$ nm (see Fig. 6d). Slowing the speed of the laser beam in the return cycle helps to lower the Ge molar fraction (down to $x_0 = 0.5$) between the two maxima of the composition. Periodic heterostructure formation via modulation of the beam position and power are shown in Supplementary Movies 1 and 2, respectively.

Optical simulations for transmission and reflection spectra of waveguide Bragg gratings based on in-plane transverse SiGe superlattices. Waveguide Bragg gratings, which consist of alternating layers with different refractive indices, is one of the basic elements used in integrated photonics today. Primary methods used to fabricate Bragg gratings are material removal by etching, and local refractive index modification induced by ultra-fast lasers. Laser-written in-plane transverse SiGe superlattices can be exploited as a waveguide Bragg grating if the period $\Lambda$ of the grating satisfies the first-order Bragg condition $\lambda = 2n_{eff} \Lambda$, where $\lambda$ is the wavelength of interest, and $n_{eff}$ is the effective index of the reflected optical mode.

We simulated an in-plane transverse SiGe superlattice laser-written at a constant stage scan speed of 25 mm s$^{-1}$, using a sawtooth modulation of the laser beam position with a movement of 200 nm (see Fig. 7a). A 10-period Bragg grating with a period of $\Lambda_0 = 285$ nm was chosen for a filtering application at a wavelength range around 2 μm. The total length of the Bragg grating was kept short, due to the computational constraints of the FEM-based optical simulations in 3D. However, to increase the coupling between propagating and counter-propagating modes in such a short waveguide Bragg grating, we ensured that the maximum refractive index modulation was achieved in the periodic composition profile ($\Delta x = 0.45–0.75$ mol), as shown in Fig. 7b. The calculated transmission and reflection spectra of the 10-period waveguide Bragg grating are shown in Fig. 7c. Further details regarding implementation of the optical simulations are given in Supplementary Note 2.

A dip in the transmission spectrum was observed at a wavelength of 2038.9 nm for the reflected TE mode with an effective index of $n_{eff} = 3.5244$. By using the Bragg condition, we found that this corresponds to a period $\Lambda = 289.3$ nm, which is close to the simulated period $\Lambda_0 = 285$ nm. A slight mismatch is expected due to the coarser size of the mesh (40 nm) used for the optical simulations. Additionally, we noticed that the reflection was suppressed, which is desired for a notch filter application. The reduction in the reflection is common for single mode waveguides with asymmetric gratings, like a tilted fiber Bragg grating, which couple the reflected light into higher order lossy modes. As our graded-index SiGe waveguides support only the fundamental TE mode at a wavelength of 2000 nm, we expect high losses for the reflected higher order modes. A waveguide Bragg grating with a high number of periods (>1000) and a longer total length (>300 μm) would be straightforward to fabricate to achieve better rejection ratios in the stop band, and thus could find ready use in integrated photonics applications.

Discussion

Laser-driven phase segregation in homogeneous Si$_{0.3}$Ge$_{0.5}$ epilayers allows for achieving Ge concentrations remarkably higher ($x > 0.7$) and lower ($x < 0.25$) than the initial concentration within the strip core and under-cladding, respectively. To obtain higher
Ge concentrations, SiGe epilayers with an initially higher Ge content can be used. According to our phase-field simulations, laser processing of a SiGe epilayer with an initial 80% Ge content at a scan speed of 200 mm s$^{-1}$ enables a 95% Ge content to be reached at the top surface of the strip core. The initial Ge content can be further increased, however, the strength of the phase segregation decreases due to narrowing of the gap between the liquid and solidus curves in the phase diagram. In contrast, using transient effects such as the acceleration of the liquid/solid boundary via modulation of the laser processing parameters has a stronger effect in terms of reaching a higher Ge content. This could potentially lead to the fabrication of in-plane transverse heterostructures having pure Ge in Ge-rich SiGe epilayers, which are in high demand for development of spin-based quantum technologies$^{46}$, as well as for fabrication of quantum wells and superlattices, which could be used as building blocks of photonic and optoelectronic devices including photodetectors and modulators.

To further control the transverse patterning, laser beam shapers can be used to obtain non-circular beam shapes. For example, in-plane traverse heterostructures with straight interfaces perpendicular to the scan direction, required for quantum well-like structures, could be written using a rectangular-shaped laser beam. Although the transverse size of the periodic heterostructures is determined by the diameter of the focused laser spot, the spatial period is not diffraction-limited. This is because it is not the spot area$^{47}$, but the displacement of the 2D liquid/solid interface in the scan direction that determines the period, which in principle can be as small as the modulation frequency of the laser processing parameters can be increased. Thus, the width of the quantum wells that could be written into the epilayers is determined by multiplication of the constant scan speed and modulation period $\Delta t$. For example, to fabricate a quantum well with a width of 20 nm, the modulation period should be 1 $\mu$s for a scan speed of 20 mm s$^{-1}$. This corresponds to a modulation frequency of 1 MHz, which is within the capabilities of state-of-
the-art electro-optic laser beam modulators and deflectors. However, the estimated average thickness of the mushy solid/liquid interface is 90 nm, which depends on the local thermal gradients, and mushy zone range at $x_0 = 0.5$. Therefore, we expect that the width of the quantum wells could be decreased further by setting the spatial period of the modulation smaller than 90 nm. However, this would compromise the modulation depth $\Delta x$ in composition, which is the maximum compositional difference between the well and barrier regions. It is worth noting that these laser-written in-plane transverse heterostructures do not have an atomically-sharp interface, but a gradual one, which could be useful to fabricate graded confining potentials.

For the fabrication of active devices based on in-plane heterostructures, a doping process is required. Conventionally, doping can be realized after laser processing by either ion-implantation or thermally assisted diffusion from a dopant source. However, this would compromise the crystalline structure and Ge profile of the superlattices, decreasing the compositional contrast between the well and barrier regions. SiGe epilayers can also be doped during epitaxial growth via CVD. Laser post-processing can be then applied to these initially doped SiGe epilayers to fabricate in-plane doped heterostructures. The most appealing aspect of this strategy would be simultaneous activation of the dopants without requiring an additional annealing process, though it could also result in a redistribution of the dopants within the heterostructures. Another useful avenue to explore would be to populate un-doped Ge-rich heterostructures with charge carriers by either using modulation doping or gate-induced accumulation from the ohmic contacts. Finally, laser-driven phase segregation with modulation of the laser process parameters could also be applied to fabricate in-plane heterostructures in other pseudo-binary systems, such as metal alloys (Ni–Cu), ternary semiconductor alloys (Al$_x$Ga$_{1-x}$As), quaternary alloys of transition metal dichalcogenides (Mo$_x$W$_{1-x}$S$_y$Se$_{1-y}$)$_{14}$, and ceramics (Al$_2$O$_3$–Cr$_2$O$_3$), which have phase diagrams similar to that of SiGe alloy.

**Methods**

**Calculation of the equilibrium phase diagram for SiGe alloys.** We used the molar Gibbs free energy $F$ to calculate the equilibrium phase diagram of SiGe alloys. By fitting it to the experimental phase diagram data, we estimated the parameters in the molar Gibbs free energy $F(\phi,T,x)$, which is given at a constant volume and a constant pressure for a regular solution model of binary alloys by:

$$F(\phi,T,x) = E(\phi, T, x) - TS(\phi, T, x),$$

$$\phi = \frac{(1-x)\phi_S(T, x) + x\phi_L(T, x)}{\phi_S(T, x) + \phi_L(T, x)} + \frac{RT(1-x)\ln(1-x) + x\ln(x) + w(x)\phi(1-x)}{}$$  \hspace{1cm} (1)

where $E(\phi, T, x)$ is the total internal energy, $S(\phi, T, x)$ is the total entropy, $T$ is the temperature, $x$ is the molar fraction of Ge, $R$ is the ideal gas constant, $\phi$ is the phase-field representing the solid ($\phi = 0$) and liquid ($\phi = 1$) phases. $\phi_S(T)$ and $\phi_L(T)$ are the Gibbs free energies in units of 1 mol$^{-1}$ for pure Si and Ge, respectively. The molar Gibbs free energy for a pure element $A$ (Si or Ge) is given by:

$$\phi_S(T) = \frac{\phi_S(T, x)}{\phi_S(T, x) + \phi_L(T, x)} + \frac{RT(1-x)\ln(1-x) + x\ln(x) + w(x)\phi(1-x)}{}$$  \hspace{1cm} (2)

where $\phi_S(T)$ and $\phi_L(T)$ are the internal energy and entropy for the pure element $A$, respectively; $H_m$ is the height of the energy barrier between the liquid and solid phases at the melting temperature $T_{m,n}$. $L_{G}$ is the latent heat of melting; $g$ is the double-well potential function with minima at $\phi = 0$ and $\phi = 1$; and $p(\phi) = \phi(3 - 2\phi)$ is an interpolating function, which satisfies $p(\phi = 0) = 0$ and $p(\phi = 1) = 1$. The height of the energy barrier $H_m$ is assumed to be the same for Si and Ge, due to the similar atomic properties.

In the case of phase coexistence at thermal equilibrium, the chemical potential for each element in the solid (S) and liquid (L) solutions are equal, and given by:

$$\mu_A(\phi = 0, T, x) = \mu_A(\phi = 1, T, x),$$

$$\mu_A(\phi = 0, T, x) = \mu_A(\phi = 1, T, x),$$

where $x_0$ and $x_1$ are the Ge molar fractions in the solid and liquid solutions of Si$_x$Ge$_{1-x}$, respectively. By using Eqs. (2–7) can be rewritten explicitly in terms of the regular solution parameters, latent heats of melting, and melting temperatures of the alloy elements, as given below:

$$\ln \left(1 - x_0 \right) + \frac{L_{G}}{RT} \left(1 - \frac{1}{T_{m,n}}\right) + \frac{1}{RT} \left(w_0 x_0^2 - w_0 x_1^2\right),$$

Fig. 7 Laser-written in-plane transverse SiGe superlattice on a silicon substrate as a waveguide Bragg grating. a Composition color map shows the periodic Ge molar fraction $x$ on the top surface of an in-plane transverse SiGe superlattice written at a constant stage scan speed of 25 mm s$^{-1}$, using a sawtooth wave modulation of the laser beam position (back and forth movement of 200 nm) in the scan direction. Calculated optical intensity profile of a transverse electric (TE) polarized mode is shown as superimposed on the output facet of the simulated waveguide Bragg grating. b Ge molar fraction profile as a function of position along the center line on the top surface showing a 10-period grating with a period of 285 nm. c Calculated transmission and reflection spectra of the 10-period SiGe waveguide Bragg grating by using finite-element-method (FEM)-based optical simulations. The reflection data (red bottom line) is magnified ten times for clarity.
with a dynamic viscosity of the liquid SiGe alloy, which was estimated using the surface tensions of liquid Si and Ge. These surface tensions control the fluid motion in the molten pool due to the thermocapillary effect. Furthermore, the density of the liquid SiGe phases are taken as $\rho_{\text{Si}} = 2.33 \times 10^3 \text{ kg m}^{-3}$ and $\rho_{\text{Ge}} = 5.39 \times 10^3 \text{ kg m}^{-3}$. These terms depend on the composition and temperature, as implied by Eq. (25). The terms in the square brackets in Eq. (26) are defined as $\beta_a$ and $\beta_b$, which are the solid and liquid phases of the alloy, respectively, to obtain a compact form for the energy density, which is rewritten as:

$$e(x, \phi, T) = (1-x)e_{\text{Si}}(x, T) + x e_{\text{Ge}}(T) + w(\phi)t(1-x).$$

By using Eqs. (1, 2, and 19), the energy density of a partially-melted alloy can be written as:

$$e(x, \phi, T) = (1-x)e_{\text{Si}}(x, T) + x e_{\text{Ge}}(T) + w(\phi)t(1-x).$$

The terms depend on the composition and temperature, as implied by Eq. (26). The composition is determined by the initial Ge fraction $x_0$ of the initially-homogeneous alloy. To simplify the implementation of the simulations, we assume the x-dependent terms in $\beta_a$ and $\beta_b$ stay constant in the following derivation. Furthermore, the density $\rho$ of the SiGe alloy is also assumed to be constant (temperature independent) and the same for both phases. For derivation of a heat transport equation, by inserting Eq. (27) into Eq. (17), we obtain

$$\rho \frac{\partial T}{\partial t} = \nabla \cdot (\lambda \nabla T) + \rho \left[ (1-x)C_\text{Si} T + x C_\text{Ge} T + w(\phi) \right] \nabla x - \frac{1}{\rho} \left[ T \nabla \cdot (\rho^\text{latent} \nabla \phi) + T \nabla \phi \right].$$

Here, $k = M_T T$ is the thermal conductivity, and $c_p = c_p' + c_p''$ are the specific heat capacities of the solid and liquid phases, respectively. The terms in the square parentheses can be defined as an apparent heat capacity $c_p'$:

$$c_p = c_p'(1 - \phi) + c_p'' + \frac{\partial \phi}{\partial T}.$$
where \( L \) is the latent heat of melting for the SiGe alloy, which is estimated as
\[ L = aL_{\text{fl}} + (1 - a)L_{\text{Li}} \]
using Vegard’s law. By incorporating the thermocapillary-driven convection with a velocity field \( \mathbf{u} \) and a heat source, the final form of the heat transport equation in Eq. (29) can be written as
\[ \rho L \frac{dT}{dt} + \nabla \cdot (\mathbf{u} \rho L) = \nabla \cdot (\kappa \nabla T) + Q. \]

Here, the external heat source \( Q \), due to the volumetric absorption of the Gaussian laser beam, is given by the Beer–Lambert law in terms of the absorption coefficient \( \alpha \) and laser intensity \( \mathbf{I} \) as
\[ Q = \alpha \mathbf{I} \cdot \mathbf{E} \cdot \mathbf{n} = \frac{2(1 - R_p)}{\pi r_c^2} (1 - e^{-2\alpha r_c^2 + \mathbf{y}^T / \mathbf{r}_c^2}) \mathbf{E}^2 \mathbf{E}^2, \]
where \( R_p = 0.4 \) is the reflection coefficient, \( P_0 \) is the optical power, \( r_c \) is the beam radius, and coordinate \( X = t \) is given in terms of the time \( t \) and constant scan speed \( v \).

The shape and size of the liquid/solid interface, as the boundary of the laser-induced molten zone, is completely represented by the phase-field \( \phi \), of which evolution drives the phase segregation and non-dendritic solidification. In non-isothermal conditions, where the temperature field \( T(r, \theta) \) significantly changes within the SiGe epitaxy, the thickness of the mushy solid/liquid interface has a position-dependent value, because it is determined by the local temperature gradient \( \nabla T \) and the temperature range (mushy zone) between the liquidus and solidus curves in the phase diagram at a particular Ge content. For example, the convection with a velocity \( \mathbf{u} \) is induced molten zone, is completely represented by the phase-field equation via the apparent heat capacity \( C_{\text{app}} \), as well as to formulate the phase-field model, we assume that it is driven by a unit-area parabolic function, which moderates the capture and release of the latent heat as
\[ \frac{d\phi}{dT} = \begin{cases} \frac{1}{\mathbf{T}_m} \left( 1 - \frac{T - T_m}{T_m - T_l} \right), & 0 < T < T_m \text{(Solid phase)} \\ \frac{1}{\mathbf{T}_m} \left( 1 - \frac{T - T_m}{T_m - T_l} \right), & T_m < T < T_l \text{(Mushy zone)} \\ 0, & T > T_l \text{(Liquid phase)} \end{cases} \]

where \( T_m = (T_l + T_s) / 2 \) is the melting temperature of the alloy, and \( T_l = (T_s + T_m) / 2 \) is the temperature range for the mushy zone. In this way, the coupled evolution of temperature and phase-field parameter was determined by solving the heat transport equation in COMSOL. This helped us to reduce the number of differential equations as well as to formulate the phase-field model with variables completely governed by the known thermophysical properties of the SiGe alloys and laser processing parameters.

Data availability
The datasets generated during the current study are available in the University of Southampton Institutional Research Repository at https://doi.org/10.5258/SOTON/D1701.

Received: 13 October 2020; Accepted: 17 May 2021; Published online: 11 June 2021

References
1. Allerov, Z. I. Nobel Lecture: the double heterostructure concept and its applications in physics, electronics, and technology. Rev. Mod. Phys. 73, 767–782 (2001).
2. Paul, D. J. Si/GeSi heterostructures: from material and physics to devices and circuits. Semicond. Sci. Technol. 19, R75–R108 (2004).
3. Paul, D. J. The progress towards terahertz quantum cascades on silicon substrates. Laser Photonics Rev. 4, 310–325 (2010).
4. Bhattacharya, P. K. & Dutta, D. K. Quantum well optical devices and materials. Annu. Rev. Mater. Sci. 23, 79–123 (1993).
5. Chaisakul, P. et al. Recent progress on Ge/SiGe quantum well optical detectors, emitters, and emitters for optical interconnects. Photonics 6, 24 (2019).
6. Tournié, E. & Trampert, A. MBE growth and interface formation of compound semiconductor heterostructures for optoelectronics. Phys. Stat. Solids 244, 2683–2696 (2007).
7. Cai, Z., Liu, B., Zou, X. & Cheng, H.-M. Chemical vapor deposition growth and applications of two-dimensional materials and their heterostructures. Chem. Rev. 118, 6091–6133 (2018).
8. Yu, Y., Fan, R. & Yang, P. Block-by-block growth of single-crystalline Si/Ge/Si superlattice nanowires. Nano Lett. 22, 83–86 (2002).
9. Fang, X. et al. Heterostructures and superlattices in one-dimensional nanoscale semiconductor heterostructures. J. Mater. Chem. 19, 5683–5689 (2009).
10. Zhang, Z. et al. Robust epitaxial growth of two-dimensional heterostructures, multiheterostructures, and superlattices. Science 357, 788–792 (2017).
11. Yu, Y., Fan, R. & Yang, P. Block-by-block growth of single-crystalline Si/Ge/Si superlattice nanowires. Nano Lett. 22, 83–86 (2002).

Communications Physics | https://doi.org/10.1038/s42005-021-00632-1 | www.nature.com/commsphys

COMMUNICATIONS PHYSICS | (2021) 4:132 | https://doi.org/10.1038/s42005-021-00632-1 | www.nature.com/commsphys

11
38. Barritault, P. et al. Mlines characterization of the refractive index profile of SiGe gradient waveguides at 2.15 μm. Opt. Express 21, 11506–11515 (2013).
39. Rho, H., Jackson, H. E. & Weiss, B. L. Mapping of local stress distributions in SiGe/Si optical channel waveguide. J. Appl. Phys. 90, 276–282 (2001).
40. Brun, M. et al. Low loss SiGe graded index waveguides for mid-IR applications. Opt. Express 22, 508–518 (2014).
41. Ramirez, J. M. et al. Graded SiGe waveguides with broadband low-loss propagation in the mid-infrared. Opt. Express 26, 870–877 (2018).
42. Wang, X. et al. Nanosecond laser writing of straight and curved waveguides in silicon with shaped beams. J. Laser Appl. 32, 022002 (2020).
43. Martinez-Jimenez, G. et al. Photonic micro-structures produced by selective etching of laser-cristallized amorphous silicon. Opt. Mater. Express 9, 2573–2581 (2019).
44. Liu, Q. et al. On-chip Bragg grating waveguides and Fabry-Perot resonators for long-wave infrared operation up to 8.4 μm. Opt. Express 26, 34366–34372 (2018).
45. Ams, M., Dekker, P., Gross, S. & Withford, M. J. Fabricating waveguide Bragg gratings (WBGs) in bulk materials using ultrashort laser pulses. Nanophotonics 6, 743–763 (2017).
46. Scappucci, G. et al. The germanium quantum information route. Nat. Rev. Mater. https://doi.org/10.1038/s41578-020-00262-2 (2020).
47. Igo, J., Gabel, M., Yu, Z.-G., Yang, L. & Gu, Y. Photodefined in-plane heterostructures in two-dimensional In₅Se₆ nanolayers for ultrathin photodiodes. ACS Appl. Nano Mater. 2, 6774–6782 (2019).
48. Bi, Z. & Sekerka, R. F. Phase-field model of solidification of a binary alloy. Phys. A 261, 95–106 (1998).
49. Boettinger, W. J., Warren, J. A., Beckermann, C. & Karma, A. Phase-field simulations of solidification. Annu. Rev. Mater. Res. 32, 163–194 (2002).
50. De Oliveira, M. J. Binary mixtures in Equilibrium Thermodynamics (eds. Rhodes, W. T., Stanley, H. E. & Needs, R.) Ch. 10 (Springer, 2013).
51. Chan, C. L., Mazumder, J. & Chen, M. M. Effect of surface tension gradient driven convection in a laser melt pool: three-dimensional perturbation model. J. Appl. Phys. 64, 6166–6174 (1988).
52. Zhang, Q. Z. et al. Numerical investigation on selective emitter formation by laser doping for phosphorous-doped silicon solar cells. J. Laser Appl. 29, 022003 (2017).
53. Ricci, E. et al. Surface tension and density of Si-Ge melts. J. Chem. Phys. 140, 214704 (2014).
54. Penrose, O. & Fife, P. C. Thermodynamically consistent models of phase-field type for the kinetics of phase transitions. Phys. D 43, 44–62 (1990).
55. Steinbach, I. Phase-field models in materials science. Model. Simul. Mater. Sci. Eng. 17, 073001 (2009).

Acknowledgements
The authors acknowledge the use of the IRIDIS-5 High-Performance Computing Facility at the University of Southampton, and support of the Engineering and Physical Sciences Research Council (EPSRC) (EP/P000940/1 and EP/N013247/1) for the completion of this work.

Author contributions
O.A. and A.C.P. conceived the research. O.A. conducted the simulations and theoretical calculations. Y.Y. and M.K. prepared the epitaxially grown SiGe/Si samples and conducted STEM/EDX analysis. O.A. performed the laser processing of the SiGe samples, their imaging by optical microscopy, and optical coupling experiments for the laser-written graded-index waveguides. O.A. and A.C.P. interpreted the results and wrote the manuscript. All authors contributed to the discussion and review.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s42005-021-00632-1.

Correspondence and requests for materials should be addressed to O.A. or A.C.P.

Peer review information Communications Physics thanks the anonymous reviewers for their contribution to the peer review of this work. Peer reviewer reports are available.

Reprints and permission information is available at http://www.nature.com/reprints

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2021