Project title: High-definition crystalline Silicon-Germanium structures for Quantum circuits

Project number: K124/2018

Executive Summary

At IKZ, strained ^{28}Si in Si/SiGe heterostructures have been successfully grown by Molecular Beam Epitaxy (MBE) for quantum circuits. Secondary Ion Mass Spectroscopy (SIMS) results indicate the high isotopic enrichment as well as the high chemical purity of the layers. Scanning electron images (SEM), transmission electron micrographs (TEM) and X-ray examination show a good layer and interface morphology. Strain-relaxed SiGe buffer layers (SRB) on Si(100) substrates were grown with Chemical Vapor Deposition (CVD) at the IHP.

Misfit dislocations were identified as a central issue concerning homogeneity of strain in the quantum well layers. A TEM-based Burgers vector analysis reveals that a majority of misfit dislocations are of 60°-type, while the rest are Lomer dislocations. The 60° dislocations further turned out to be split into Shockley partials. Scanning X-ray Diffraction Microscopy (SXDM) has shown high potential in the material characterization of quantum devices with local resolution. Hereby, an analysis technique for synchrotron SXDM data has been developed to calculate the components of the lattice strain tensor in the Si QW layer from the diffraction maps, which has been proved to be useful to understand the nature of defects relevant for quantum applications.

Manufactured samples were delivered to IHT, IQI for further processing. For the development of non-invasive gates (IHT/IQI) process development was carried out, in particular with respect to selective dry etching and chemical mechanical polishing processes. Non-invasive Ohmic contacts have been fabricated using an ultrathin SiN interlayer in order to remove Fermi level pinning. As an alternative, non-invasive Ohmic contacts have also been realized with laser annealing. For local electrical sample characterization, a fabrication process for two-array gate design has been developed. Stable 2DEG accumulation was observed in the CVD grown heterostructures. Thus, one could sequentially form single-electron transistors (SETs) within a 2D grid and characterize the gate-voltages required to form tunnel barriers, as well as to map the local charge noise.

1. Achievement of objectives and milestones

The objectives and milestones of the project are listed below, together with the time table comparing them to the planned schedule. All proposed work packages were finalized, the milestones were achieved and accomplished, where only the last milestone MC4.36 was only partially fulfilled due to difficulties to process the material stacks. However, the research effort to produce coherence figures of IKZ/IHP produced devices is ongoing after project termination.

List of Milestones and Objectives of the Project (Milestone, Partner, Number, Month)
MA1.12 – ^{28}Si of required quantity and purity available
MA2.26 – Suitability of MBE layers is compared with that of CVD (B.1)
MB1.06 – Epi-ready CVD grown virtual SiGe/Si substrates for optimal MBE growth of SiGe
MB2.26 – Suitability of CVD layers is compared with that of MBE (A.2)
    → Layers with superior perfection are preferably delivered to WP C.1-4
MC1.08 – Non-invasive ohmic contacts to SiGe 2DEG with R < 5 kΩ
MC1.12 – Non-invasive interface-engineered ohmic contacts on Si at 1 K with R < 5 kΩ
MC1.17 – Decision on priority of gate fabrication (C1 or C2)
MC2.12 – Non-invasive interface-engineered contacts on Si and Si/SiGe QW (1 K, < 5 kΩ)
MC4.10 – Suitability tests of C.1 devices for key figure testing completed; feedback to C.1.3
MC4.12 – Charge noise of Si/SiGe fabricated with non-invasive gates
MC4.32 – Charge noise figures of full devices; feedback to A, B, C tasks
MC4.36 – Coherence figures T_1 and T_2^* of full devices

2. Activities and obstacles

The activities described here, together with the activities and results presented in the Interim Report, outline the activities during the funding period. Additional Figures can be found in the Attachment.

WP A 1-4

IKZ was able to develop ^{28}Si MBE source material, as well as the growth process of lattice-matched ^{28}SiGe/^{28}Si/^{28}SiGe heterostructures on SiGe virtual substrates by molecular beam epitaxy (MBE) [1]. Furthermore, a plethora of material characterization methods was established to elucidate the nature of defects in such heterostructures. In particular, X-ray and electron microscopy-based methods were applied to shed light on the nature of structural defects, such as dislocations [Thesis1]. A central growth challenge hereby was the development of reliable surface cleaning methods of the virtual substrates, to ensure epitaxy free of pits or mounds. Most successful results were obtained by combination of ex-situ wet chemical cleaning, as well as in-situ annealing, involving atomic hydrogen exposure [1, Thesis1].

The strain in the Si quantum well layers was verified globally by X-ray based methods and locally by Raman mapping. Structural defects, such as dislocations, were identified by transmission electron microscopy (TEM) and electron channelling contrast imaging (ECCI). Misfit dislocation were identified as a central issue concerning homogeneity of strain in the quantum well layers [2, 3].

ECCI was a particularly useful tool to investigate the nature of misfit dislocations, since it is destruction-free and allows for fast feedback. The elongation of misfit dislocations from threading dislocations in Si/SiGe as well as Ge/SiGe heterostructures was observed to be a thermally activated process, as shown in Figure 1. Hereby, MBE allowed to grow heterostructures below thermal activation of dislocation glide, making it possible to study the activation law by ex-situ annealing experiments and ECCI investigations. These results are submitted at Journal of Applied Physics and currently under peer-review.

SIMS investigations conducted by RTG revealed a low concentration of ^{29}Si of only 100 ppm in the epitaxy layers, as well as a low concentration of C and O, but a significant contamination from Au, originating from preceding research conducted in the used MBE chamber. This Au contamination was responsible for the unsuccessful device demonstration on the MBE grown heterostructures. To resolve this issue, a novel MBE system was set up and is in operation since beginning of 2023, and ^{28}Si^{76}Ge heterostructure development for qubits application is ongoing.

WP B 1-3

At IHP, SiGe strain-relaxed buffer layers (SRB) were successfully grown on Si(001) substrates by Reduced Pressure Chemical Vapor Deposition (RP-CVD), which serve as so-called virtual substrates (VS) for MBE growth experiments at IKZ. For this purpose, ten such substrates were supplied by IHP to IKZ.

To improve the uniformity and fabrication yield of semiconductor spin qubits, the relation between the material properties of the heterostructures housing the qubits and the qubit quantum performance has to be determined. Moreover, a feedback loop for the fabrication procedure of qubit heterostructures has been developed. To improve the quality of the Si quantum well (QW), further development has been performed on the graded buffer of the SiGe/Si heterostructures grown at IHP. For the advanced layer stack, which is sketched in Figure 2a, the thickness of the $Si_{1-x}Ge_x$ steps (x = 5, 10, 15, 20, 25 %) in the graded buffer have been increased from previously 50 nm to 500 nm. The increased step thickness leads to a more effective bending of dislocations, achieving a reduction of the threading dislocation density (TDD) from a previous value of > 10^7 cm^{-2} to approx. $2 \cdot 10^6$ cm^{-2} in the upper part of the $Si_{0.7}Ge_{0.3}$ buffer layer. The improvement is apparent in X-ray Diffraction (XRD) measurements, by comparing the width in sample rocking angle ω of the 224 Bragg reflection from the $Si_{0.7}Ge_{0.3}$ for an old sample (SJZ331-09) with a new sample (EJZ059-06), as shown in Figure 2b. Furthermore, in the current heterostructures, the fringe oscillations stemming from interference between the coherent interfaces of the Si QW layer are well resolved in reciprocal space maps (RSM, Figure 2c), demonstrating the high crystalline quality.

Moreover, a procedure geared towards the characterization of qubit heterostructures has been established at IHP, including Atomic Force Microscopy (AFM, Figure 2d) to measure the lateral length scale and amplitude of the cross hatch pattern, Transmission Electron Microscopy (TEM, Figure 2e) to characterize layer thicknesses and interface quality and the quantification of TDD by etch pit counting (EPC, Figure 2f) by Scanning Electron Microscopy (SEM). This will allow for further tuning of CVD process conditions by short-looped feedback towards optimized material properties, in particular surface morphology, interface sharpness, and dislocation density.

As predicted in the intermediate report, Scanning X-ray Diffraction Microscopy (SXDM) has shown high potential in the material characterization of quantum devices with local resolution, as demonstrated by recent publications[4, 6].

The analysis technique for SXDM data, obtained at the Synchrotron beamline ID01/ESRF, has been developed to calculate the components of the lattice strain tensor in the Si QW layer from the diffraction maps. In Figure 3, maps of the in-plane strain $\varepsilon_{\mathrm{yy}}$, surface normal strain $\varepsilon_{\mathrm{zz}}$ and shear strain $\varepsilon_{\mathrm{yz}}$ around a “QuBus” test structure are shown. We observe local modulations on a magnitude of approx. $5 \cdot 10^{-4}$ - $1 \cdot 10^{-3}$ of these three strain components caused by three different effects:

1. The Ti/Pd electrodes are acting as stressors, inducing strain modulation in the QW layer. This effect is particularly strong along the edges of the electrodes
2. Likewise, strain modulations are observed along the edges of the Ni micromagnet deposited on top of the Ti/Pd electrodes
3. Line-like defects are observed in the strain maps that are assumed to either originate from the misfit dislocation network in the relaxed $Si_{0.7}Ge_{0.3}$, or from misfit segments in the Si QW layer.

It is expected that the magnitude of the effects (1) and (2) will increase when the device is cooled down to cryogenic temperatures for QuBus operation, due to the mismatch of the thermal expansion coefficients between Si(Ge) and Pd or Ni respectively. Preliminary band structure calculations indicate that strain modulations in the range observed here cause fluctuations of several meV in the potential level of the Si conduction band minimum, which is comparable to the charging energy of electrostatic quantum dots and thus may form an obstacle to e.g. coherent electron shuttling. Thus, we have been able to observe and identify causes of inhomogeneity in the lattice strain landscape of quantum devices and make a prediction of their impact on device performance. Furthermore, more complex QuBus devices fabricated with e-beam lithography have been investigated by SXDM. Preliminary analysis of the diffraction maps shown in Figure 4 demonstrates that it is possible to resolve small gates with less < 100 nm width. Further manuscripts for publication of these and other Synchrotron datasets are in preparation, and their description and analysis will form the body of a PhD Thesis to be submitted this year.

WP C 1-4

Compared to the Interim report, we explored a wide space of laser-annealing parameters (laser-power *P*, scan velocity *v* and direction) for 3 Si/SiGe heterostructure from IHP and one commercial reference from LSRL. As a first step color change and topology after recrystallization is evaluated by optical-microscope inspection (Figure 5a). The laser-power *P* is the most critical parameter and its operation window for the LSRL reference is caused by stepwise versus linear alloy content grading in the SiGe buffer (latter for LSRL). The reflectivitiesof all heterostructures at laser wavelength were approx. equal. We measure the contact resistance to the SiGe 2DEG by gated Hall-Bars at 4 K (Figure 5b). We have achieved reproducible contact resistances of *R*~600 Ohm, thus much better than our goal of 5 kOhm (MC1.08 reached). Recrystallization by laser-annealing leads mostly to lower *R* compared to global annealing by rapid thermal annealing (RTP). We saw first indications that electron mobility in the QW is enhanced if the contacts are laser-annealed.

Furthermore, we fabricated the simulated test-device (see intermediate report) by a two-layer e-beam lithography process (Figure 6a). We observed stable 2DEG accumulation in new IHP heterostructure compared to the ones of the intermediate report. Thus, we could sequentially form single-electron transistors (SETs) within a 2D grid and characterized the gate-voltages required to form the tunnel barriers (Figure 5b-e, MC4.10). The homogeneity of the IHP material is better than the one used as reference material. The reference test structure was cooled down to 20 mK to form up to 14 SETs at different positions and to map the local charge noise (Figure 6f), in order to show that the method for MC4.32 works. The noise $S^{1/2}$(1Hz) varies between 0.91 to 5.57 µeV/sqrt(Hz) and is higher than the best value of 0.47 µeV/sqrt(Hz) we published earlier.

3. Results and successes

SiGeQuant was instrumental in initiating the new research direction of materials development for semiconductor-based quantum technologies at the IKZ. This was cemented by founding a new junior research group “SiGe-based Quantum Materials and Heterostructures” lead by Dr. Kevin-Peter Gradwohl and establishing a new MBE system for isotopically enriched SiGe epitaxy only.

Based on SiGeQuant, the IKZ and the project partners are participating in the BMBF cooperation project QUASAR until end of 2024. A research collaboration related to SiGe buffer development with the German company Siltronic AG has been established running until end of 2024. IKZ became partner in the Canadian Consortium on Quantum Simulation with Spin Qubits (CQS2Q), a consortium funded by the Research Council of Canada, running until 2028. IHP and IQI became partners in the European Horizon 2020 project Quantum Large Scale Integretion in Silicon (QLSI), running until 2024. Further project applications at the DFG and the BMBF are submitted/in preparation.

Within the project activities several PhD student were employed, which published several first authorship papers. Hereby, Yujia Liu successfully defended her thesis in March 2023 with great distinction “magna cum laude” [Thesis1]. Two further PhD student are about to submit their thesis.

A list of the research publications arising within the project activities can be found below:

[1] Liu, Yujia, et al. "Growth of 28Si Quantum Well Layers for Qubits by a Hybrid MBE/CVD Technique." ECS Journal of Solid State Science and Technology 12.2 (2023): 024006.

[2] Liu, Yujia, et al. "Role of critical thickness in SiGe/Si/SiGe heterostructure design for qubits." Journal of Applied Physics 132.8 (2022): 085302.

[3] Gradwohl, Kevin-P., et al. "Strain relaxation of Si/SiGe heterostrucutres by a geometric Monte Carlo approach." physica status solidi (RRL)–Rapid Research Letters (2022).

[4] Corley-Wiciak, Cedric, et al. "Nanoscale Mapping of the 3D Strain Tensor in a Germanium Quantum Well Hosting a Functional Spin Qubit Device." ACS Applied Materials & Interfaces (2023).

[5] Liu, Yujia, et al. "28Silicon-on-insulator for optically interfaced quantum emitters." Journal of Crystal Growth 593 (2022): 126733.

[6] Corley-Wiciak, Cedric, et al. "Lattice deformation at the sub-micron scale: X-ray nanobeam measurements of elastic strain in electron shuttling devices." Submitted (2023). arXiv:2304.09120

[Thesis1] Liu, Yujia. "28 Si, Ge Epitaxy for Qubits." (2023).

4. Equal opportunities, career development and internationalisation

Our effort to recruit female and international staff to our research team was successful with the employment of Ms Yujia Liu from China as a PhD student at IKZ, Mr Ketan Anand from India at IHP, and Ms Isabelle Sprace as Master student. Ms Sprave continues as a PhD student at IQI at the end of the project. The founding of a junior research group at IKZ allowed for a generational change, and supports a new PhD student at IKZ working on this topic.

5. Structures and collaboration

The structure of the cooperation consisting of IKZ, IHP, and IHT/IQI as well as the time schedule corresponded to the work packages of the project application. The research efforts and governance of the activities were organized by the project leader Dr. Torsten Boeck (IKZ) up to August 2022 and due to health reasons assumed by Dr. Kevin-Peter Gradwohl (IKZ). At IHP, Dr. Wolfgang Klesse acted as project officer until he left IHP in February 2022, whereupon this position was assumed by Mr. Cedric Corley-Wiciak.

Within the research on ^{28}SiGe structures for quantum circuits, a wide collaboration network beyond the project activities could be established. This includes, just to name a few, a close collaboration with Prof. Sven Rogge from UNSW, Australia and Prof. Nils Curson from UCL, Great Britain, Dr. Andreas Reiserer, Max-Planck für Quantenoptik, Germany, Dr. Peter Storck from Siltronic AG, Germany, Prof. Oussama Moutanabbir, Polytechnique Montreal, Canada, Dr. Giordano Scappucci, TU Delft, Netherlands, Prof. Michele Virgilio, Universita di Pisa, Italy, Dr. Vincent Mourik, FZ Jülich, Germany, as well as collaboration with industry, such as IBM Research Europe, Zürich, Switzerland.

Hereby, the IKZ strategically positioned itself for the long-term development and research on such heterostructures by founding a Junior Research Group “SiGe-based Quantum Materials and Heterostructures” and investing 1.2 million € into a novel MBE system developed within a close collaboration to the German company Dr. Eberl MBE Komponenten GmbH. This MBE system is now in operation and designed for the epitaxy of nuclear spin-free ^{28}Si and ^{76}Ge only.

6. Quality assurance

The proven in-house referee system of the IKZ is used for quality assurance of scientific publications of the project partners. Process data and characterization results are communicated via a safety-certified European database. IHP has ensured open access for all its publications and both in-house and Synchrotron measurements results are being stored at the IHP servers. The raw data of the latter is also publicly available from the large-scale facilities. No animal testing has been conducted.

7. Additional resources

The estimated value of in-kind resources of the total project is approx. 982k€. Budgetary financing of several employees in the IKZ account for approx. 250k€ , as well as the investment for a new MBE system of around 1.2 million €. More information can be found in the attached Tables. But as described in Chapter 5, for the IKZ, these costs seem justified because SiGeQuant has opened the door to a new, future-oriented research direction for the institute enabling collaboration with excellent partners.

8. Outlook

SiGeQuant has opened to door for the project partners to enter the research field on SiGe-based quantum materials and has revealed several still open problems and research questions, which will be subject of upcoming research proposals:

First, a central realization was the significant influence of crystal defects such as dislocations in the SiGe buffers or in the QW layers on the device performance, as well as the related short comings of the technology. However, the process to control these defects to the desired degree has not been achieved yet, and is an open problem. Moreover, dislocations deep in the buffer layers and the gate electrodes of the device are observed to induce local strains. This leads to an inhomogeneous potential landscape in the QW and may pose an obstacle to the controlling of gates with shared voltages. Furthermore, reliable control of the valley splitting in SiGe heterostructures is a central unresolved issue, and essential for a reliable SiGe-based qubit technology. In particular, heterostructure design and the associated epitaxy processes are key proposals to control the valley splitting.

Attachment - Tables and Figures

workpackages	month																																			
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36
IKZ																																				
A.1 Material for MBE targets from monoisotopic 28Si	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	**MA1.12**																								
A.2 MBE growth of SiGe layer stack				🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	**MA2.26**			🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩
A.3 structural characterization by TEM					▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨
A.4 Electrical characterization					▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	
IHP																																				
B.1 Development and productin of SiGe VS	🟩	🟩	🟩	🟩	🟩	**MB1.06**			🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	
B.2 CVD growth of SiGe layer stack						🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	**MB2.26**			🟩	🟩	🟩	🟩	🟩	🟩	🟩	
B.3 XRD characterization	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨
IHT / IQI																																				
C.1 Non-invasive gate fabrication by spacer process	🟩	🟩	🟩	🟩	🟩	**MC1.08**			🟩	🟩	🟩	**MC1.12**			🟩	🟩	**MC1.17**			🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩
C.2 Non-invasive ohmic contacts				🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	**MC2.12**			🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩
C.3 Non-invasive gate fabrication by wafer bonding																			▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨	▨
C.4 Local electrical sample characterization							🟩	**MC4.10**			🟩	**MC4.12**			🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	**MC4.32**			🟩	**MC4.36**		

Table 1: List of work packages associated with the project partners and respective milestones.

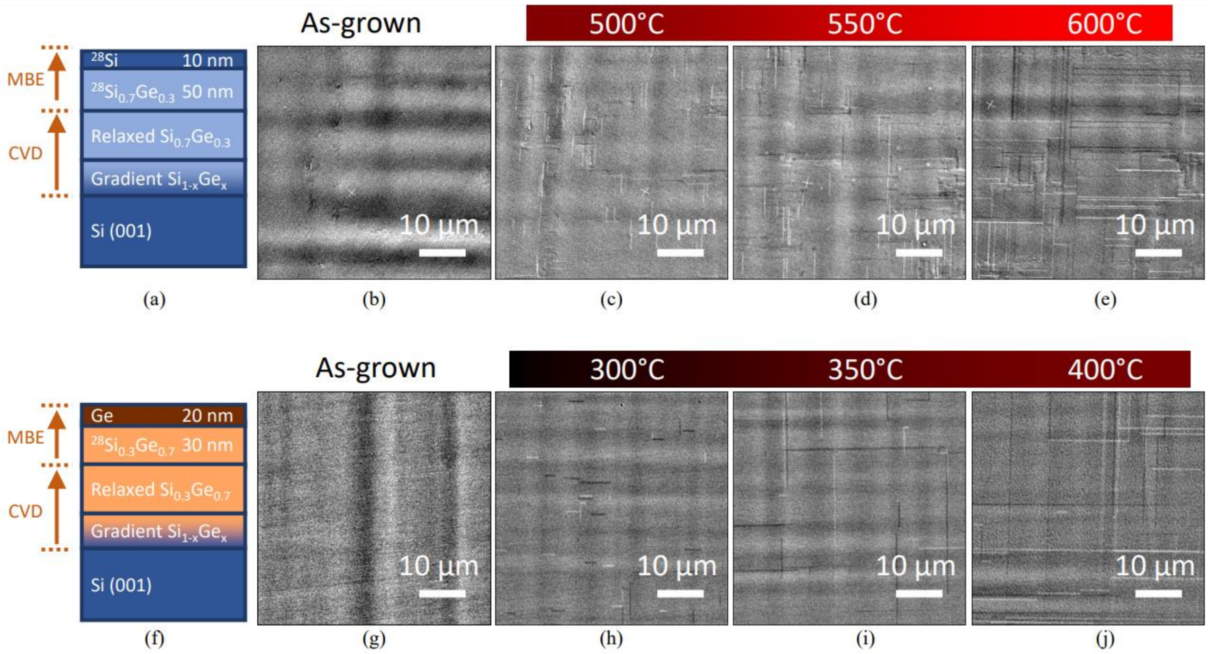

Figure 1: Schematic cross sections of (a) a strained Si layer on relaxed $^{28}Si_{0.7}Ge_{0.3}$ or (f) the strained Ge layer on relaxed $^{28}Si_{0.3}Ge_{0.7}$ grown at IKZ. ECCI images show the dislocations in the strained Si and Ge layers: (b) the as-grown Si layer presents no misfit dislocation; the misfit dislocations form in the Si layer through the annealing at temperatures from 500°C to 600°C (c - e) for 10 min. Similarly, the as-grown Ge layer presents no misfit dislocation (g); the misfit dislocations form in the Ge layer through the annealing at temperatures from 300°C to 400°C (h - j) for 10 min. The annealing temperatures are indicated above the ECCI images.

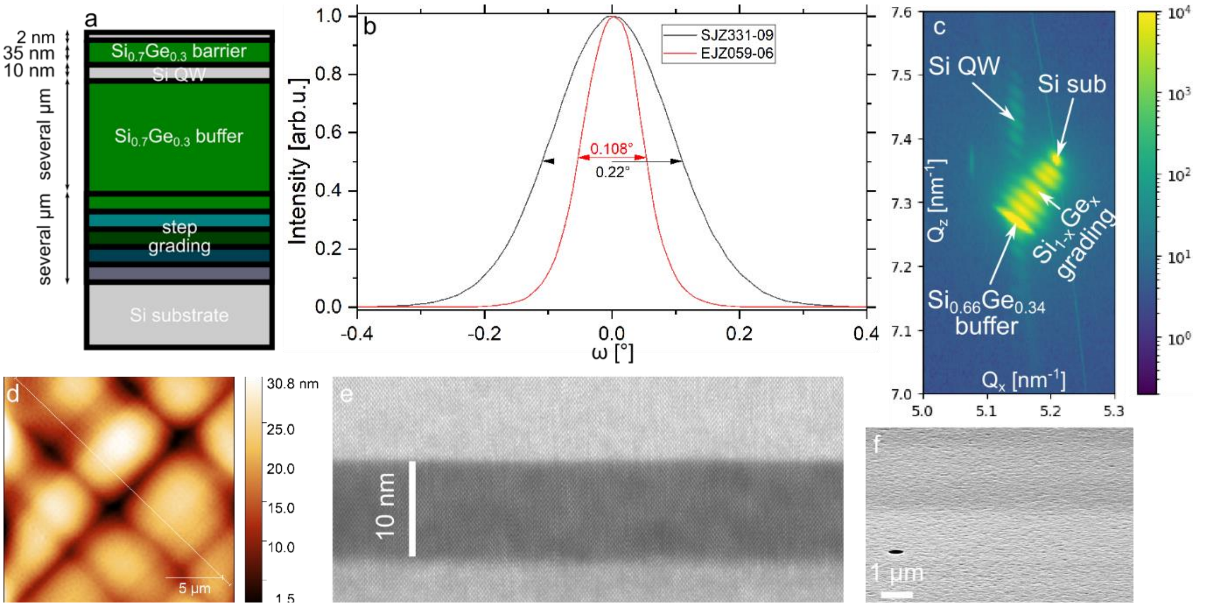

Figure 2: (a) Schematic cross section of the improved CVD heterostructure layer stack. (b) 224 rocking curves of the SiGe buffer of an old (black curve) and a new (red) sample. (c) 224 RSM of a new sample, showing the oscillations from coherent interfaces of the Si QW layer. (d) AFM topography map of the sample surface, showing the cross-hatch pattern. (e) TEM image in cross section of the Si QW layer. (f) Plane-view SEM image after a SECCO etch, with a pit from a single dislocation.

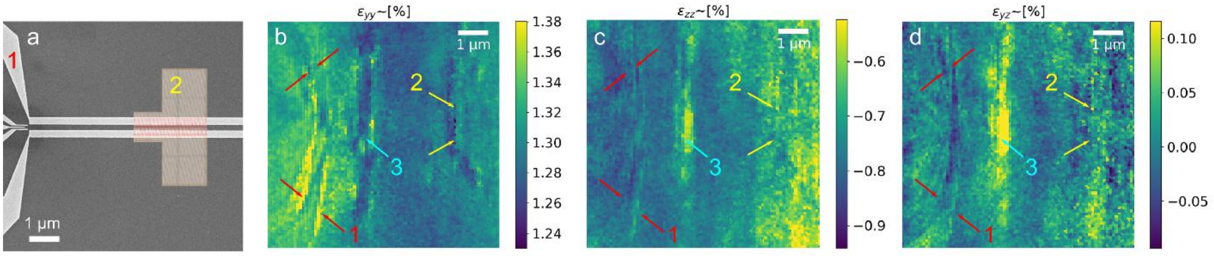

Figure 3: (a) Plane-view SEM image of a QuBus test structure with a sketch of the micromagnet; Strain maps in the Si QW layer: (b) ε_{yy}, (c) ε_{zz}, (d) ε_{yz}. In the measured area, there are strain modulations caused by (1) the QuBus electrodes, (2) the micromagnet, (3), misfit dislocations.

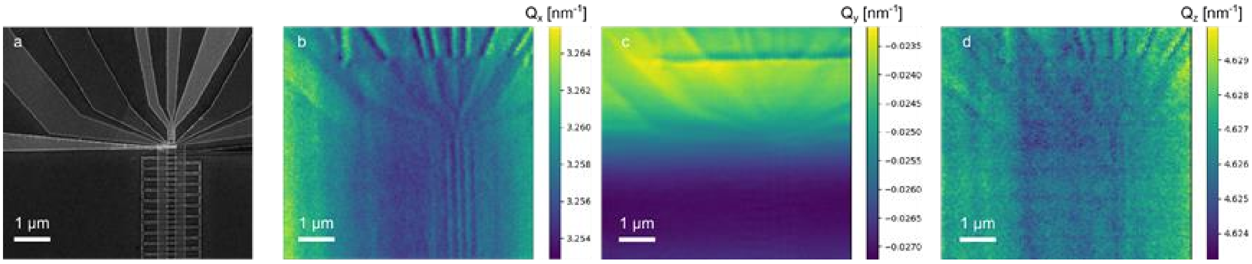

Figure 4: (a) Plane-view SEM image of a QuBus with small gate pitch; diffraction maps in the Si QW layer: (b) Q_x, (c) Q_y, (d) Q_z.

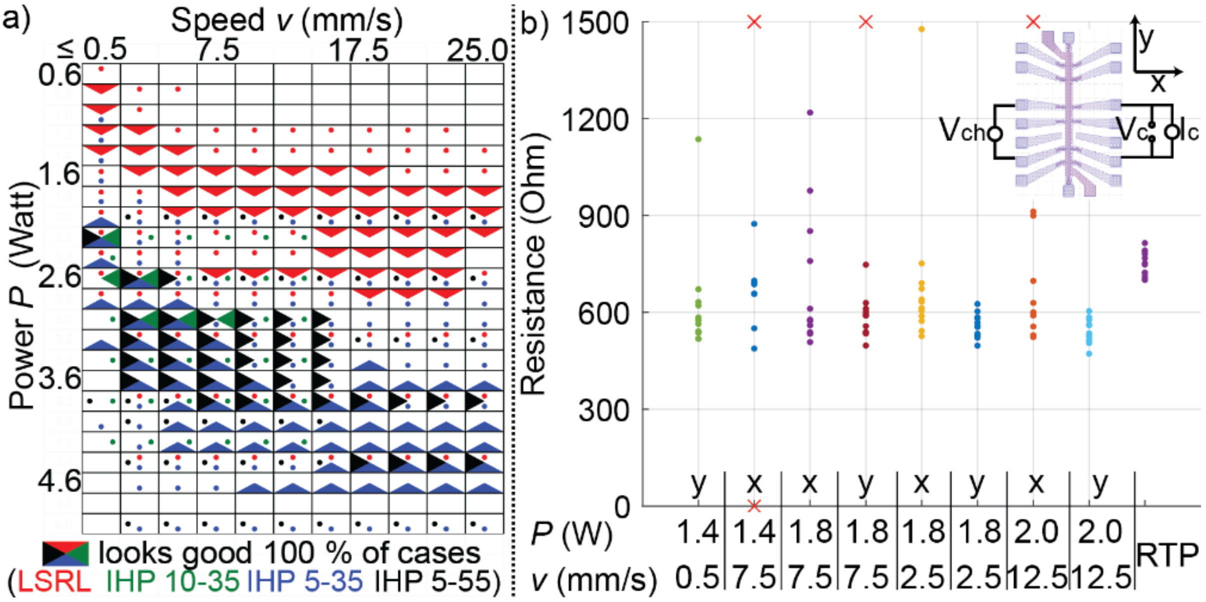

Figure 5: Non-invasive ohmic contacts by laser annealing. a) Optical inspection of all ohmics after annealing as a function of laser power and scan velocity for 4 heterostructures (encoded by colors). Triangles indicate a success rate of 100%, while dots indicated tested but unsuitable parameters. b) Contact resistance to QW as function of scan direction (x, y), laser power and scan velocity. Schematic of used gated-Hall-bar as inset. Bright violet areas are laser-annealed.

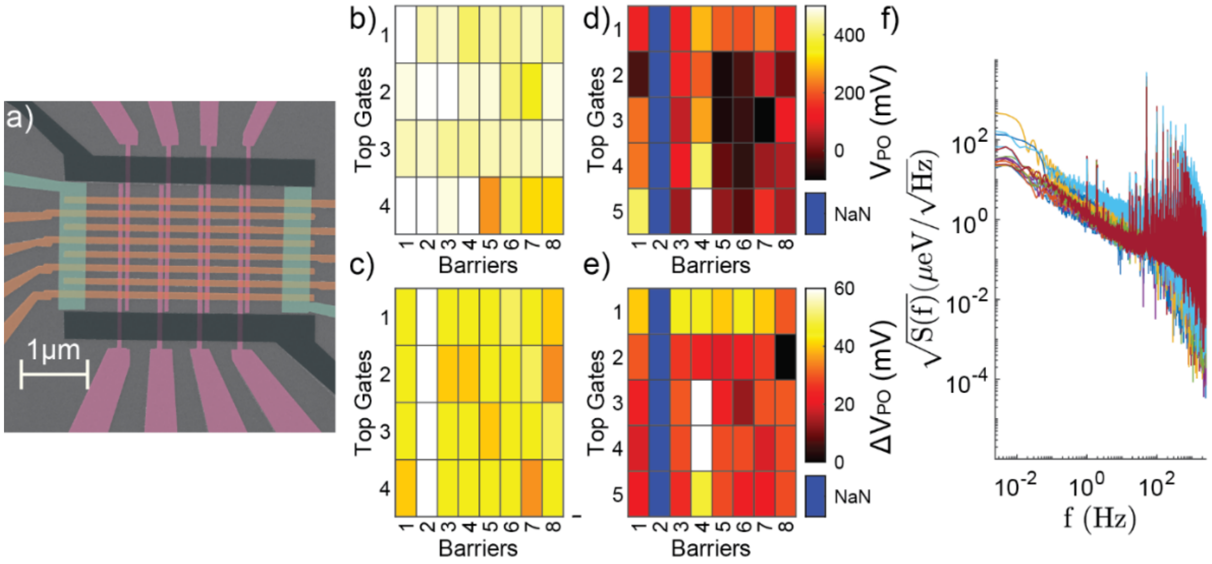

Figure 6: Characterization by transport. a) False-colored scanning-electron micrograph transport test-structure. b, c) Barrier pinch-off voltage VPO and its hysteresis VPO mapped for an IHP wafer at 4 K. d, e) Same as b,c for MBE grown reference sample. Blue color is used, if the barrier cannot be formed due to e.g. broken gate. f) Charge noise spectrum of 14 SETs formed in the device at 20 mK.