



HAL
open science

Microstructure of GaAs thin films grown on glass using Ge seed layers fabricated by aluminium induced crystallization

D. Pelati, G. Patriarche, L. Largeau, O. Mauguin, L. Travers, F. Brisset, F. Glas, F. Oehler

► To cite this version:

D. Pelati, G. Patriarche, L. Largeau, O. Mauguin, L. Travers, et al.. Microstructure of GaAs thin films grown on glass using Ge seed layers fabricated by aluminium induced crystallization. *Thin Solid Films*, 2020, 694, pp.137737. 10.1016/j.tsf.2019.137737 . hal-02404458

HAL Id: hal-02404458

<https://hal.science/hal-02404458v1>

Submitted on 26 Nov 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Microstructure of GaAs thin films grown on glass using Ge seed layers fabricated by Aluminium Induced Crystallization

D. Pelati^{a,c,d}, G. Patriarche^a, L. Largeau^a, O. Mauguin^a, L. Travers^a, F. Brisset^b, F. Glas^a, F. Oehler^{a,*}

^aCentre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Sud, Université Paris-Saclay, 10 Boulevard Thomas Gobert, 91120 Palaiseau, France

^bInstitut de Chimie Moléculaire et des Matériaux d'Orsay, CNRS, Université Paris-Sud, Université Paris-Saclay, 91405 Orsay, France

^cRIBER SA, 31 rue Casimir Périer, 95870 Bezons, France

^dInstitut Photovoltaïque d'Ile-de-France (IPVF), 18 Boulevard Thomas Gobert, 91120 Palaiseau, France

Abstract

We perform the growth of GaAs epilayers by molecular beam epitaxy (MBE) on Ge pseudo-substrates obtained by the Aluminium Induced Crystallisation (AIC) of thin amorphous Ge layers deposited on silica. Despite the apparent uniformity of the AIC-Ge layer, large domains (more than 50 μm wide) previously thought to be monocrystalline are found to actually consist in smaller grains (500 to 1000 nm wide), separated by low angle grain boundaries. These defects are transferred during epitaxy to the GaAs layer and degrade the quality of the III-V material. In our growth conditions, the MBE results in the selective deposition of thin GaAs layers on Ge with respect to the silica support, but selectivity is progressively lost with increasing layer thickness.

Keywords:

Metal Induced Crystallization, ALILE, Germanium, Aluminium, Gallium Arsenide, Molecular Beam Epitaxy

1. Introduction

Metal induced crystallization (MIC) is a promising technique for the fabrication of crystalline Si or Ge layers on low cost supports. Depending on the nature of the metal, the crystallization of amorphous Ge can occur at very low temperatures, down to 177°C for Au or 211°C for Al, below the respective eutectic temperature of the binary mixtures (Ge-Al 420°C, Ge-Au 361°C)[1, 2, 3]. The aluminium-induced crystallization (AIC) of Ge has attracted much interest due to the peculiar layer inversion process (ALILE) during the crystallization[4]. The low cost of the ALILE technique has primarily prompted solar applications[5, 6, 7, 8], although there have been other outcomes such as transistors[9, 10].

Interestingly, the control of the diffusion of Ge in the metal can lead to the crystallization of oriented grains with normals mainly along the [111] or [100] directions, depending on the nature of diffusion barrier and substrate[11, 12, 13, 14, 15, 16, 17]. The fabrication of flat oriented crystals with large grains

(wider than 50 μm) is interesting and can compete with other approaches such as localized liquid phase crystallization[18] or laser annealing[19, 20].

Yet, the use of MIC layers as oriented crystalline pseudo-substrates for subsequent epitaxy has been rather limited. Thin MIC-Si or MIC-Ge layers on silica have been used as substrates for the growth of GaAs[21, 22] or Ge[23] nanowires, but nanostructures with small footprints are not very demanding in terms of substrate crystallinity[24]. In an attempt to reduce defect density present in the MIC-Ge layer, Toko *et al.* performed the homoepitaxy of planar Ge layers on MIC-Ge pseudo-substrates by molecular beam epitaxy[15] (MBE). Due to the good lattice match between Ge and GaAs (lattice parameters GaAs 5.653Å, Ge 5.658Å), the direct epitaxy of GaAs on such low-cost Ge layers is a promising route for the fabrication of cheaper III-V devices and solar cells. Comparable approaches have already been investigated in the literature, with GaAs solar cells grown on large-grain poly-Ge[25, 26] or Ge buffer on Si(100)[27]. Recently, work by Toko *et al.* has shown a promising photoresponse from GaAs epilayers grown by MBE on MIC-Ge pseudo-substrates[28].

Yet the quality of MIC-Ge pseudo-substrates, which

*
Email address: fabrice.oehler@c2n.upsaclay.fr (F. Oehler)

consist in wide ‘monocrystalline’ domains[13, 15, 28], remains lower than commercial mono-crystalline Ge wafers. In particular no study has looked in details at the microstructure of the so-called ‘mono-crystalline’ MIC-Ge domains and how it affects the subsequent GaAs epitaxy.

2. Materials and methods

We use here a fabrication procedure similar to that of Toko *et al.*[28]. A simplified scheme is presented Fig. 1 and highlights the two main steps: the fabrication of the MIC-Ge pseudo-substrates (Fig. 1.a) and the epitaxy of the GaAs epilayers (Fig. 1.b).

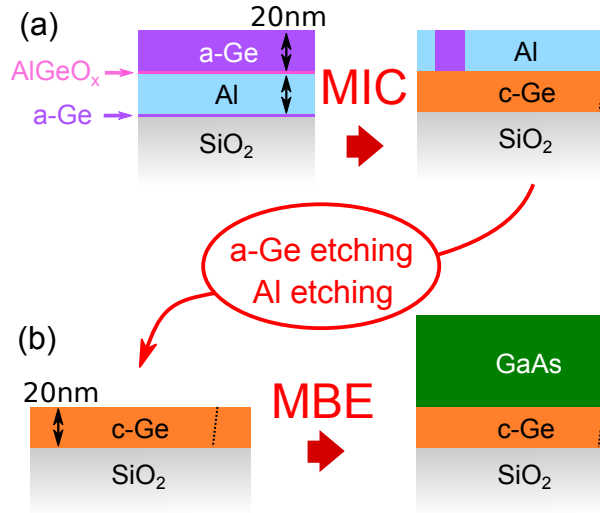


Figure 1: Simplified processing scheme of the samples. (a) Fabrication by MIC (ALILE) of the crystalline (c-Ge) pseudo-substrates starting from 20 nm thick amorphous Ge (a-Ge) and aluminium layers deposited on silica. (b) Growth of GaAs on the Ge pseudo-substrates by molecular beam epitaxy (MBE).

The MIC-Ge pseudo-substrates are fabricated as follows. First, we form a 70 nm thick silica layer on standard Si(100) 2” wafers (Siltronix) by dry oxidation at 1050°C for 15 min, which acts as a virtual amorphous silica substrate for the rest of the process. Samples fabricated from such oxidized Si wafers can be easily cleaved to fit the various equipments used in this study. Then, we deposit the Al and Ge layers by e-beam evaporation using a Plassys MEB550SL, as detailed in our previous work[29].

Before crystallization, the MIC-Ge sample consists in a 1.0 nm Ge underlayer[30], followed by 20 nm of Al and a 1.0 nm thick Ge layer which is exposed to air (for 10 min) to form a AlGeO_x diffusion barrier[31, 32], and

a final 20 nm thick Ge top layer. The MIC is performed in a programmable hot plate (Harry Gestigkeit) flushed with nitrogen gas by heating the sample at 270-300 °C during 15 to 20 h. After the annealing, the remaining amorphous Ge is selectively removed using reactive ion etching (CCP/RIE Nextral NE100, using O₂ and SF₆ for 4 min)[33]. Then the top Al layer is removed chemically using H₃PO₄ at 100°C during approximately 60 s. The change of the surface reflectivity, from the bright Al layer to the darker crystalline Ge material (c-Ge), is used to control the wet etch duration.

Before introduction in the Molecular Beam Epitaxy (MBE) chamber (RIBER 32) the MIC-Ge pseudo-substrates are further cleaned using H₂O₂ (30 s) and HF 1% (30 s) treatments, with intermediate rinse in deionized water. The MIC-Ge substrates are then out-gassed at 400°C in a dedicated ultra high vacuum chamber, before transfer to the main growth chamber (base pressure 1.0 10⁻¹⁰ Torr). The GaAs epitaxy is performed using parameters previously optimized for the twin-free epitaxy of GaAs(111) on Ge(111) monocrystalline wafers[34].

Here, gallium is provided by a standard effusion cell, while arsenic is delivered by a valved cracker cell (RIBER VAC500) producing As₄ tetramers (cracker temperature 600°C). The Ga beam equivalent pressure (BEP) is set to 1.5 10⁻⁷ Torr, corresponding to a growth rate of ~1.5 Å.s⁻¹ on GaAs (100). The ratio of arsenic to gallium (BEP_{As₄} : BEP_{Ga}) is approximately 60:1. The substrate temperature is 625-635°C, as measured by thermocouple and optical pyrometry, during all the growth procedure.

The samples are characterized before and after growth using Scanning Electron Microscopy (SEM, FEI Magellan). The local crystalline orientation is measured using Electron Backscatter Diffraction (EBSD, ZEISS Supra 55 VP with Hikari/OIM TSL EDAX detector). The layers are also characterized by Transmission Electron Microscopy (TEM, FEI Titan THEMIS) using Energy Dispersive X-ray Spectroscopy (EDX, Bruker Super-X). Focused Ion Beam (FIB, FEI Helios) is used to prepare the TEM foils.

3. Results

3.1. Growth

Figure 2 presents the surface morphology before and after the GaAs growth. The initial MIC-Ge surface is shown Fig. 2.a, in which the silica support appears dark. The growth of a 40 nm thick GaAs layer (at 630°C for 4.5 min) creates large density of triangular terraces and

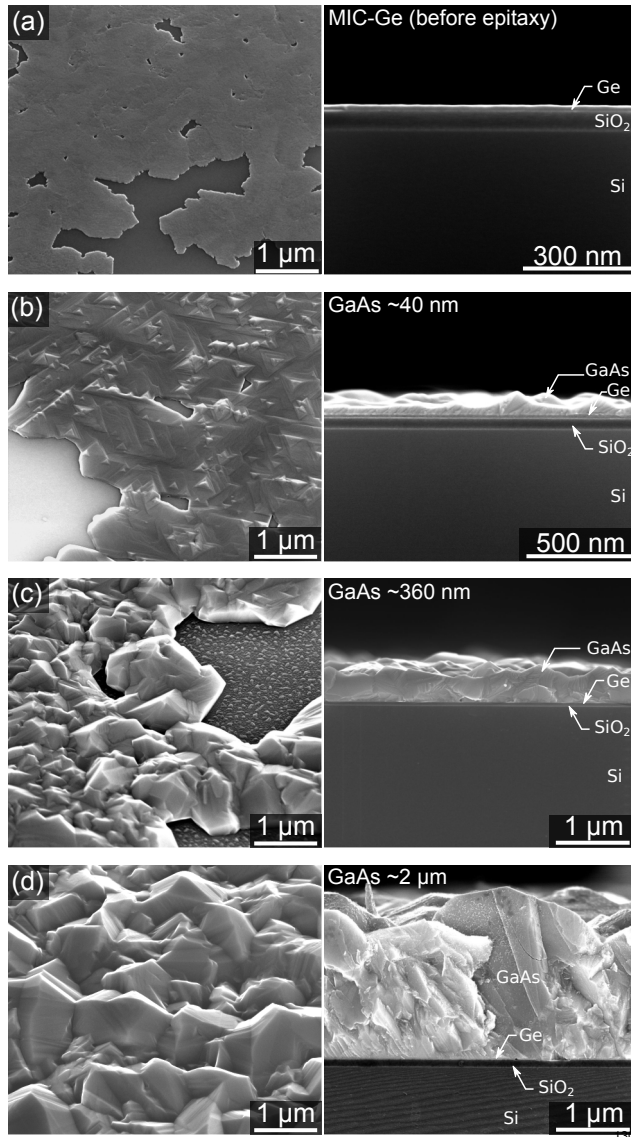


Figure 2: SEM image of the samples before and after GaAs growth of increasing thickness. (a) Bird's eye view (45° tilt) and cross sectional image of the initial MIC-Ge layer. (b-d) Similar images after 40 nm, 360 nm and 2 μm thick GaAs growth on MIC-Ge pseudo-substrates.

120 small pyramidal hillocks, Fig. 2.b. The triangular shape
 121 of the hillocks is compatible with a (111) orientation of
 122 the layer. For this short duration, the MBE growth of
 123 GaAs is selective and no GaAs crystal nucleates on the
 124 silica surface.

125 For 60 nm thick GaAs layer (Fig. 2.c), grown for
 126 40 min, the surface becomes rougher and small GaAs
 127 crystals appear on the SiO₂ layer. For even thicker GaAs
 128 layers (2 μm), grown for approximately 200 min, the surface
 129 is very rough and the growth is not selective at all

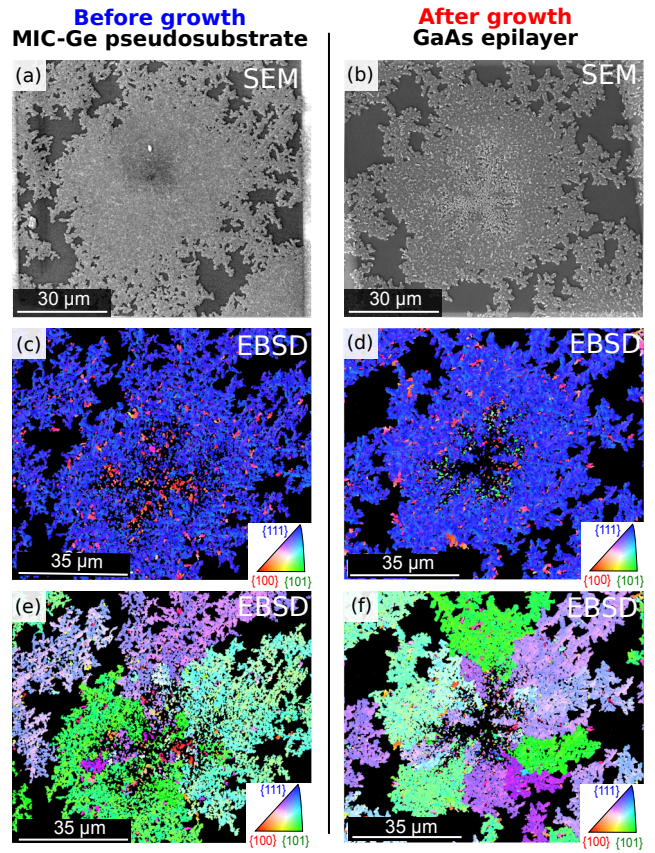


Figure 3: Different zones from the initial MIC-Ge pseudo-substrate (before growth, left) and the GaAs epilayer (after growth, right). (a)(b) SEM top views of the MIC-Ge and GaAs surfaces, respectively. (c)(d) Corresponding EBSD maps of the *out of plane* crystal orientation. The color is assigned from the inverse pole figure in insert. (e)(f) Corresponding EBSD maps of the *in plane* crystal orientation. Same color scale as in (c-d).

(Fig. 2.d).

3.2. Crystal orientation

Figure 3 presents SEM and EBSD characterizations of the sample before and after 40 nm thick GaAs growth. Before growth (Fig. 3.a) MIC-Ge pseudo-substrate is structured in large Ge islands on SiO₂ (more than 50 μm diameter, 20 nm thick) which are mostly (111) oriented, as indicated by the dominant blue color in the *out of plane* EBSD (Fig. 3.c). Although the island shown in Fig. 3.a appears homogeneous, it is actually made of three separate dendritic crystals with different *in plane* orientations (Fig. 3.e). The central core is a defective zone with mixed orientation[29]. Due to the small grain size and poor crystal quality in this area, the corresponding zones appears black in the EBSD maps, Fig. 3.c and 3.e. Between the Ge islands, the amorphous

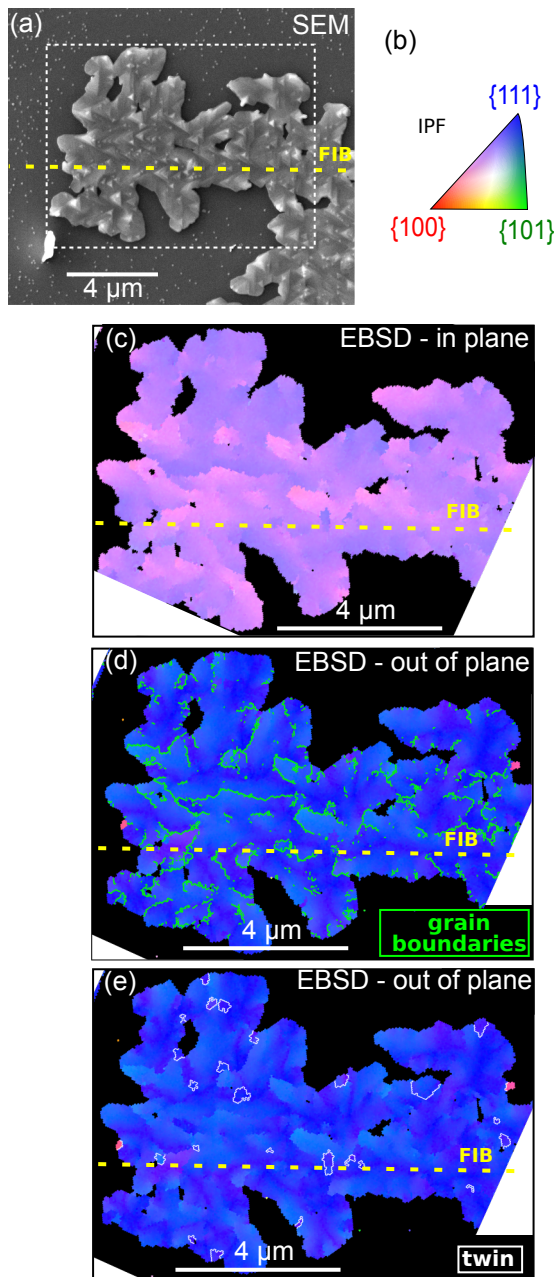


Figure 4: SEM and EBSD characterizations of an area with uniform *in plane* orientation. The yellow dashed line marks the position of the FIB cut used to extract the TEM foil. (a) Top view SEM image showing the zone selected for EBSD analysis (white rectangle). (b) Inverse pole figure (IPF) color scheme used in the EBSD maps. (c) EBSD map of the *in plane* crystal orientation. (d) *Out of plane* EBSD map showing grain boundaries (0-10°) in green. (e) *Out of plane* crystal orientation, twin boundaries are marked in white.

146 silica substrate does not yield any diffraction signal and
 147 also appears black on the EBSD maps.

148 Figure 3.b presents the morphology after the growth
 149 of a thin GaAs layer (40 nm). As already seen in
 150 Fig. 2.b, this short GaAs deposition is selective with respect
 151 to the silica support and the GaAs crystals only grow
 152 on the MIC-Ge islands. EBSD characterization
 153 (Fig. 3.d) shows that the preferential (111) orientation
 154 of the MIC-Ge layer (Fig. 3.c, blue color) is conserved
 155 in the GaAs layer. The central defective core and the
 156 dendritic crystals with different *in plane* orientations are
 157 also visible in the GaAs crystal orientation (Fig. 3.f), as
 158 observed in the MIC-Ge layer before growth (Fig. 3.e).

159 By zooming on the termination of a large dendrite
 160 (Fig. 4.a) we can focus on an apparently monocrys-
 161 talline area with uniform *in plane* orientation (Fig. 4.c)
 162 and (111) crystal orientation (Fig. 4.d). At this scale,
 163 small correlated color in the *in plane* and *out of plane*
 164 EBSD maps shifts hint at the presence of small adjacent
 165 grains with slightly different crystal orientation.
 166 To reveal this grainy structure, we highlight in bright
 167 green local misorientations (0-10°) between neighboring
 168 points (Fig. 4.d). The obtained set of grain bound-
 169 aries drastically reduces the typical grain size from sev-
 170 eral tens of microns to one micron approximately. Fig-
 171 ure 4.e highlights in white color another set of misori-
 172 entation (60° *in plane* rotation), which corresponds to
 173 twinned domains for (111)-oriented cubic crystals. The
 174 typical lateral size of twinned domains is also of the
 175 order of one micron or smaller.

176 3.3. Microstructure

177 To better understand the microstructure of the MIC-
 178 Ge and GaAs layers, we perform a FIB cut in the
 179 previously analyzed ‘monocrystalline’ area (see yellow
 180 dashed line in Fig. 4). Figure 5 shows g_{002} dark field
 181 images from the TEM cross section oriented along the
 182 $[1\bar{1}0]$ zone axis. This type of image is sensitive to com-
 183 positional changes and permits to differentiate GaAs
 184 from and Ge, despite the close lattice match and crys-
 185 tal structure.

186 In a defect free region (Fig. 5.b), we observe a smooth
 187 MIC-Ge layer with a thickness of 20 nm, which is
 188 equal to the nominal thickness of the initial amorphous
 189 Ge layer (see methods). The thickness of the GaAs
 190 layer is 40 nm, as expected from the MBE growth rate
 191 ($1.5 \text{ \AA} \cdot \text{s}^{-1}$) and duration (4.5 min).

192 While no defect is visible in this smooth area
 193 (Fig. 5.b), pyramidal hillocks and vertical defects are
 194 found in zone with thicker (~60 nm) GaAs (Fig. 5.c).
 195 These defects originate from the MIC-Ge layer (orange
 196 arrow) and extend to the whole GaAs thickness (green
 197 arrows). The area bordering these vertical defect are

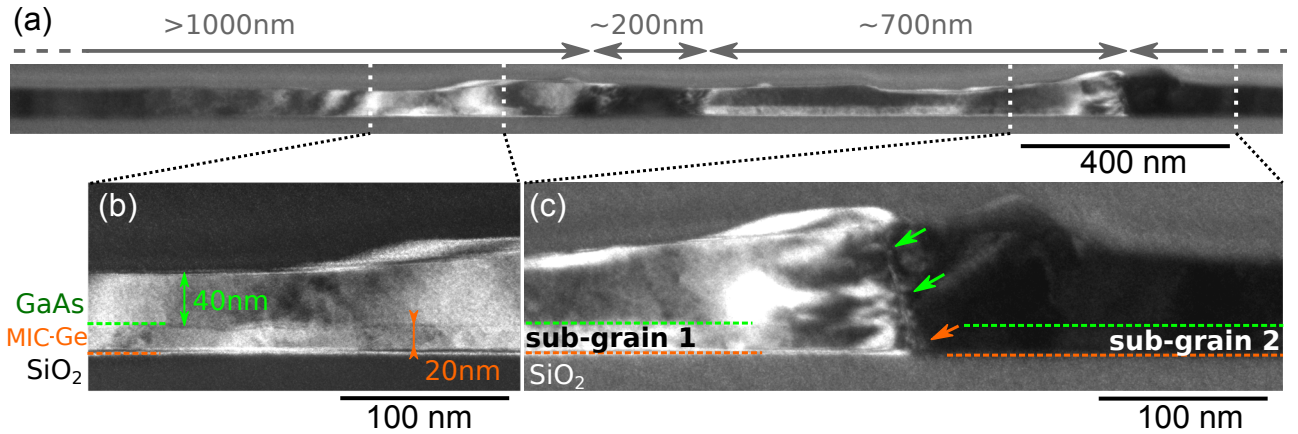


Figure 5: Dark field cross-sectional g_{002} TEM image of the GaAs / MIC-Ge pseudo-substrate near the $[1\bar{1}0]$ zone axis. (a) Large scale view showing the zones selected for further analysis. The lateral size of the sub-grains is indicated above the image. (b) Details of a smooth area showing the thickness of each layer on the SiO_2 support. (c) Details of a pyramidal hillock. A defect from the MIC-Ge pseudo-substrate (orange arrow) extends to the GaAs layer (green arrows).

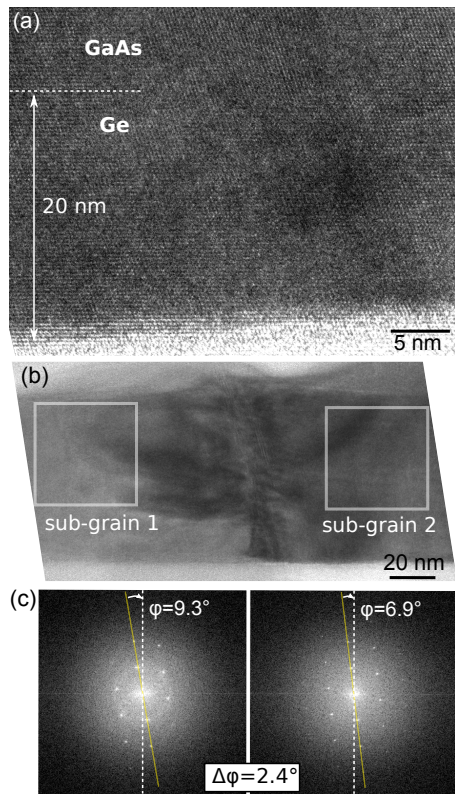


Figure 6: HR-TEM images of the zones analyzed in Fig. 5. (a) Atomically resolved image of the smooth area (see Fig. 5.b). (b) Image of the defective zone with two sub-grains (see Fig. 5.c). The left and right squares mark the zones used to perform FFT. (c) FFT patterns of the left and right sub-grain. Both GaAs crystals are cubic with a relative angular misalignment of $\Delta\varphi=2.4^\circ$.

198 highly contrasted. Sub-grain #1 (left of the defect) ap-
 199 pears in light color while sub-grain #2 (at right) is dark.
 200 The lateral size of the sub-grains is between 200 and
 201 1000 nm, or possibly larger (see Fig. 5.a).

202 Figure 6 presents high resolution TEM (HR-TEM)
 203 images of the zones previously analyzed in Fig. 5. The
 204 smooth area is detailed in Fig. 6.a. There, we observe
 205 the continuation of the cubic crystal lattice from the
 206 MIC-Ge pseudo-substrate to the GaAs layer. Figure 6.b
 207 shows HR-TEM image of the defective area of Fig. 5.c
 208 . Two zones are selected in this image to compute a
 209 fast Fourier transform (FFT). This FFT (Fig. 6.c) reveals
 210 that two standard cubic patterns with a relative misori-
 211 entation of $\Delta\varphi=2.4^\circ$ between the two crystals. This
 212 demonstrates that the extended defect of Fig. 5.c and
 213 Fig. 6.b is a low-angle grain boundary (LAGB).

214 4. Discussion

215 Beside a few reports on nanowire growth[21, 22, 23],
 216 we could only find an two attempts at planar layer
 217 epitaxy on MIC substrates: homoepitaxy of Ge on
 218 MIC-Ge(111)[15] and hetero-epitaxy of GaAs on MIC-
 219 Ge(111)[28]. Contrary to Toko *et al.*[28], who report
 220 good photoresponse from GaAs layers grown on MIC-
 221 Ge, the disrupted morphology of our thick GaAs layers
 222 (Fig. 2) indicates that defects degrades the III-V material
 223 quality, at length scales much smaller to the appar-
 224 ent domain size (over $20\mu\text{m}$) determined by large scale
 225 EBSD (Fig. 3).

226 4.1. Absence of antiphase boundaries

227 Compared to commercial ‘epiready’ monocrystalline
228 Ge wafers, the surface of our MIC-Ge pseudo-substrates
229 is very rough: nanometer sized variations of the MIC-
230 Ge thickness can be seen directly by cross-sectional
231 TEM in Fig. 5, even in the defect-free area. Due to
232 the small thickness of the MIC-Ge layer (20 nm) fur-
233 ther chemical or mechanical polishing is proscribed. It
234 is thus critical that the GaAs epitaxy is not sensitive to
235 the Ge surface roughness. Yet, even the standard epitaxy
236 case of polar GaAs on non-polar Ge is known to require
237 a carefully reconstructed double-step Ge(100) surface
238 and a specific wafer miscut[35, 36]. The direct GaAs
239 epitaxy on (100)-oriented MIC-Ge layers is thus likely
240 to hold high densities of antiphase boundaries (APB).

241 The identification of APB from EBSD maps is a dif-
242 ficult task, as the standard IPF color scheme (Fig. 4.b)
243 is insensitive to 180° rotations, so that it cannot dis-
244 criminate grains with opposite orientations. Note that
245 this applies to both *out of plane* (Fig. 4.d) and *in plane*
246 (Fig. 4.c) EBSD maps.

247 Fortunately, the EBSD data (Fig. 3 and 4) show
248 that the main orientation of our MIC-Ge layers is
249 (111), similarly to most layers fabricated by ALILE on
250 silica[13, 30, 15]. On commercial ‘epiready’ Ge(111)
251 wafers, the polarity of GaAs epilayers grown by MBE
252 is uniquely determined and insensitive to the surface
253 roughness, so that no antiphase domain form during the
254 growth[37, 34]. Our TEM analysis (Fig. 5) did not re-
255 veal any APB in our GaAs layers, despite the lower sur-
256 face quality of our MIC-Ge pseudo-substrates compared
257 to commercial Ge(111) wafers. The growth of GaAs
258 on (111)-oriented MIC-Ge templates remains ABP-free
259 and it is thus a promising combination for the fabrica-
260 tion of low-cost III-V devices on silica.

261 4.2. Selective growth of GaAs on MIC-Ge(111)/SiO₂

262 The use of a silica support for the MIC-Ge layers is
263 particularly relevant since SiO₂ masks are already used
264 in the selective area growth of Ge[38] or GaAs[39, 40]
265 by MBE. Despite the relatively high substrate tempera-
266 ture (~630°C), we only observe a partial selectivity for
267 the GaAs growth between the MIC-Ge islands and the
268 SiO₂ support, which is progressively lost as the deposi-
269 tion duration increases from 4.5 to 40 min (Fig. 2). It
270 is possible that the exposed silica still host some residual
271 Al or Ge contaminations from the deposition of the ini-
272 tial MIC structure, despite the thorough chemical clean-
273 ing of the MIC-Ge layers (see methods). A solution to
274 this partial selectivity issue is to fabricate MIC-Ge lay-
275 ers with a high surface coverage[13, 15] so that no SiO₂
276 is exposed to GaAs during the MBE growth[28].

277 4.3. Epitaxial relationship GaAs/MIC-Ge(111)

278 By focusing on the thin GaAs layer (~40 nm) for
279 which the growth is still fully selective (Fig. 2), we
280 can learn more about the epitaxial relationship between
281 the GaAs layer and the MIC-Ge pseudo-substrate. The
282 transfer of the large scale (111) crystal texture from
283 the MIC-Ge template to the GaAs epilayer (Fig. 3) is
284 a good indication of the epitaxy relationship. This hy-
285 pothesis is confirmed by cross-sectional HR-TEM im-
286 ages (Fig. 6), where we observe the continuation of the
287 crystal lattice from the cubic MIC-Ge grain to the GaAs
288 cubic crystal. The growth of GaAs (111) on the MIC-
289 Ge(111) thus proceeds as on standard monocrystalline
290 Ge substrates[37, 41, 34] and preserve in GaAs the same
291 *in plane* and *out of plane* orientations as the Ge sub-
292 strate. The absence of visible defect by TEM at the
293 GaAs/Ge(111) interface (Fig. 5) demonstrates that our
294 substrate fabrication and cleaning procedures enable the
295 defect-free epitaxy of GaAs(111) on MIC-Ge(111).

296 4.4. Twinning in GaAs(111) epilayers

297 As the GaAs growth proceeds in epitaxy with the
298 MIC-Ge substrate, it seems likely that the crystal ori-
299 entation of MIC-Ge grains is exactly that of GaAs epi-
300 layer. Yet it is possible that extended defects in the
301 GaAs layer alter this initial epitaxy relationship.

302 The color scheme of the EBSD maps stems from a
303 standard IPF (Fig. 4.b). As stated before, this color cod-
304 ing does not differentiate between 180° rotations, so that
305 it is not only insensitive to antiphase domains but also
306 to twin defects. However twin boundaries can be re-
307 vealed by showing the local 60° *in plane* misorienta-
308 tions in (111)-oriented domains (Fig. 4.e).

309 From previous work[34] and literature[41] using
310 commercial monocrystalline Ge(111) wafers, we know
311 that the MBE growth of GaAs on Ge(111) is prone to
312 twinning and that the twin density can be minimized by
313 adapting the V:III ratio during growth. The Figure 4.e
314 shows that twinned area fraction is relatively low (3%)
315 using MIC-Ge pseudo-substrates. Yet, we cannot fully
316 replicate the growth of twin-free GaAs layers obtained
317 on commercial Ge(111) wafers, even we use the same
318 optimized growth conditions[34]. It is also possible that
319 twinning occurs in the MIC-Ge layer, during the Ge
320 crystallization, but we did no investigate this hypothe-
321 sis further due to the low density of twinned domains
322 compared to other extended defects.

323 4.5. Microstructure of MIC-Ge pseudo-substrates

324 We previously studied the macrostructure of our
325 MIC-Ge layers on silica using *in situ* optical

326 microscopy[29]. This study showed that the defec- 378
327 tive core of each Ge island crystallizes from a para- 379
328 sitic MIC process, while the radially expanding (111)- 380
329 oriented dendrites are created by the standard ALILE 381
330 mechanism. These large Ge have different *in plane* 382
331 orientations (Fig. 3.e), and it is generally accepted that they 383
332 are monocrystalline[15, 30, 31], as supported by large 384
333 scale EBSD maps. 385

334 However, one can usually find grains with a differ- 386
335 ent orientation, typically close to (100), in the other- 387
336 wise (111)-oriented dendritic crystal (see red dots vs 388
337 blue patches in Fig. 3.c). By zooming on the termina- 389
338 tion of a large dendrite (Fig. 4), we avoid such localized de- 390
339 fects and we can assess the quality of the main fraction 391
340 of the GaAs epilayer. Hence, whereas the GaAs ap- 392
341 pears homogeneous at large scale, it breaks down into 393
342 small sub-micronic domains at small scale. Beside a 394
343 minor fraction of twinned domains (Fig. 4.e), we ob- 395
344 serve correlated color shifts in the *in plane* (Fig. 4.c) 396
345 and *out of plane* (Fig. 4.d) EBSD projections, some 397
346 of which are identified as grain boundaries (Fig. 4.d). 398
347 However EBSD characterization is usually performed 399
348 on flat surfaces, which are usually obtained after fine 400
349 mechanical polishing or ion milling. Considering the 401
350 disrupted morphology of our 40 nm thick GaAs layers 402
351 (Fig. 4.a), the small angular misorientations highlighted 403
352 in Fig. 4.d could be EBSD artefacts due to rough surface 404
353 of the sample. It is therefore important to complement 405
354 the EBSD data by another characterization technique. 406

355 Using cross-sectional TEM, we link the position of 407
356 the triangular hillocks observed by SEM (Fig. 4.a) 408
357 with those of extended defects inside the GaAs layer 409
358 (Fig. 5). Further analysis (Fig. 6) demonstrates that the 410
359 observed extended defects are low angle grain bound- 411
360 aries (LAGBs). Combining both TEM analyses (Fig. 5- 412
361 6) with the SEM overview (Fig. 4), we hypothesize that 413
362 each hillock may be associated to a LAGB. This high 414
363 density of LAGB is coherent with the estimated position 415
364 of grain boundaries computed by EBSD (Fig. 4.d). The 416
365 typical grain size determined by TEM, (200-1000 nm, 417
366 Fig. 5) matches the length scale of the small tonal 418
367 changes in the EBSD maps (Fig. 4.c) and the sub-grain 419
368 width of Fig. 4.d. The inner structure of a LAGB con- 420
369 sisting in a dense 1D array of dislocations[42], the high 421
370 density of LAGBs also explains the poor optical quality 422
371 of our GaAs epilayers (not shown). 423

372 The Fig. 5.a and Fig. 6.b clearly show that the LAGB 424
373 originates from the MIC-Ge layer. This implies that 425
374 the epitaxial growth merely extends the defective mi- 426
375 crostructure of the MIC-Ge pseudo-substrate to the 427
376 GaAs layer. Compared to other attempt at planar epi- 428
377 taxy using MIC-Ge pseudo-substrates[15, 28], we ob-

serve defect-free regions (MIC-Ge and GaAs layers) 429
which span more than several hundreds of nanometer 430
(Fig. 5), with only a couple of LAGB over one micron. 431
This suggests that the quality of our MIC-Ge material is 432
better if not comparable to the state of the art. 433

Moreover, we note that the small orientation shifts 434
characterized by EBSD (Fig. 4), which we link to the 435
defective LAGB microstructure (Fig 6), are visible in 436
most of the literature on crystalline Ge layers fabri- 437
cated using ALILE[15, 30, 31, 13] or gold-induced 438
crystallization[43]. It is thus likely that the formation 439
of LAGB is intrinsic to the metal-assisted crystalliza- 440
tion of thin Ge layers, as regards the published results 441
on (111)-oriented MIC-Ge layers on silica. 442

443 5. Conclusion

We have successfully performed the heteroepitaxy 444
of GaAs by MBE on (111)-oriented MIC-Ge pseudo- 445
substrates fabricated on SiO₂ supports. The epitaxy re- 446
lationship between GaAs and MIC-Ge is the same as 447
for standard growth on monocrystalline bulk Ge wafers. 448
No extended defect nucleates at the GaAs/Ge growth 449
interface, which indicates that our cleaning procedures 450
are adequate. The low fraction of twinned GaAs crys- 451
tals indicates that our MBE growth conditions are close 452
to optimal values. However, the epitaxial growth trans- 453
fers the defective microstructure of the MIC-Ge pseudo- 454
substrate to the GaAs epilayer so that the final III-V 455
material is of poor quality. Despite an apparent homo- 456
geneity at large scale and EBSD maps similar to the 457
current literature, we find that our MIC-Ge layers are 458
composed of slightly misaligned small crystallites (200 459
to 1000 nm wide), even in apparently ‘monocrystalline’ 460
domains. This defective microstructure appears as small 461
tonal variations in standard EBSD maps. The observed 462
high density of low-angle grain boundaries, hence dense 463
dislocation arrays, impact the final III-V material qual- 464
ity and may limit the performance of III-V devices fab- 465
ricated on MIC-Ge pseudo-substrates. 466

467 Acknowledgement

The authors thank the Institut Photovoltaïque d’Ile 468
de France (IPVF) for financial support under frame- 469
work project E.3. The authors also acknowledge ANR 470
Investissement d’Avenir program (TEMPOS Project 471
ANR-10-EQPX-50) for having funded the acquisition 472
of the NANOTEM platform and the TEM-STEM (FEI 473
Titan Themis) used in this work. 474

424 **References**

- 425 [1] S. Gaudet, C. Detavernier, A. J. Kellock, P. Desjardins,
426 C. Lavoie, Thin film reaction of transition metals with germa-
427 nium, *Journal of Vacuum Science & Technology A* 24 (3) (2006)
428 474–485 (2006). doi:10.1116/1.2191861.
- 429 [2] W. Knaepen, C. Detavernier, R. V. Meirhaeghe, J. J. Sweet,
430 C. Lavoie, In-situ X-ray diffraction study of metal induced crys-
431 tallization of amorphous silicon, *Thin Solid Films* 516 (15)
432 (2008) 4946–4952 (2008). doi:10.1016/j.tsf.2007.09.037.
- 433 [3] W. Knaepen, S. Gaudet, C. Detavernier, R. L. V. Meirhaeghe,
434 J. J. Sweet, C. Lavoie, In situ x-ray diffraction study of
435 metal induced crystallization of amorphous germanium, *Journal of Applied Physics* 105 (8) (2009) 083532 (2009).
436 doi:10.1063/1.3110722.
- 437 [4] O. Nast, S. R. Wenham, Elucidation of the layer exchange mech-
438 anism in the formation of polycrystalline silicon by aluminum-
439 induced crystallization, *Journal of Applied Physics* 88 (1)
440 (2000) 124–132 (2000). doi:10.1063/1.373632.
- 441 [5] O. Nast, S. Brehme, S. Pritchard, A. G. Aberle, S. R. Wenham,
442 Aluminium-induced crystallisation of silicon on glass for thin-
443 film solar cells, *Solar Energy Materials and Solar Cells* 65 (1-4)
444 (2001) 385–392 (2001). doi:10.1016/s0927-0248(00)00117-3.
- 445 [6] I. Gordon, D. V. Gestel, K. V. Nieuwenhuysen, L. Carnel,
446 G. Beaucarne, J. Poortmans, Thin-film polycrystalline silicon
447 solar cells on ceramic substrates by aluminium-induced crys-
448 tallization, *Thin Solid Films* 487 (1-2) (2005) 113–117 (2005).
449 doi:10.1016/j.tsf.2005.01.047.
- 450 [7] D. V. Gestel, I. Gordon, J. Poortmans, Aluminum-induced
451 crystallization for thin-film polycrystalline silicon solar
452 cells: Achievements and perspective, *Solar Energy Materials and Solar Cells* 119 (2013) 261–270 (2013).
453 doi:10.1016/j.solmat.2013.08.014.
- 454 [8] O. Shekoofa, J. Wang, D. Li, Y. Luo, C. Sun, Z. Hao,
455 Y. Han, B. Xiong, L. Wang, H. Li, P-silicon thin film
456 fabricated by magnetron sputtering and aluminium induced
457 crystallization for schottky silicon solar cells, *Materials Science in Semiconductor Processing* 71 (2017) 366–373 (2017).
458 doi:10.1016/j.mssp.2017.06.008.
- 459 [9] R. Chen, W. Zhou, M. Zhang, M. Wong, H.-S. Kwok, High-
460 performance polycrystalline silicon thin-film transistors based
461 on metal-induced crystallization in an oxidizing atmosphere,
462 *IEEE Electron Device Letters* 36 (5) (2015) 460–462 (2015).
463 doi:10.1109/led.2015.2409858.
- 464 [10] K. Moto, K. Yamamoto, T. Imajo, T. Suemasu, H. Nakashima,
465 K. Toko, Polycrystalline thin-film transistors fabricated on high-
466 mobility solid-phase-crystallized Ge on glass, *Applied Physics Letters* 114 (21) (2019) 212107 (2019). doi:10.1063/1.5093952.
- 467 [11] A. Okada, K. Toko, K. O. Hara, N. Usami, T. Suemasu,
468 Dependence of crystal orientation in Al-induced
469 crystallized poly-Si layers on SiO₂ insertion layer thick-
470 ness, *Journal of Crystal Growth* 356 (2012) 65–69 (2012).
471 doi:10.1016/j.jcrysgro.2012.07.015.
- 472 [12] R. Numata, K. Toko, N. Saitoh, N. Yoshizawa, N. Usami,
473 T. Suemasu, Orientation control of large-grained Si films on
474 insulators by thickness-modulated Al-induced crystallization,
475 *Crystal Growth & Design* 13 (4) (2013) 1767–1770 (2013).
476 doi:10.1021/cg4000878.
- 477 [13] K. Nakazawa, K. Toko, N. Saitoh, N. Yoshizawa, N. Usami,
478 T. Suemasu, Large-grained polycrystalline (111) Ge films on in-
479 sulators by thickness-controlled Al-induced crystallization, *ECS Journal of Solid State Science and Technology* 2 (11) (2013) Q195–Q199 (2013). doi:10.1149/2.007311jss.
- 480 [14] K. Toko, R. Numata, N. Saitoh, N. Yoshizawa, N. Usami,
481 T. Suemasu, Selective formation of large-grained, (100)- or
482 (111)-oriented Si on glass by Al-induced layer exchange,
483 *Journal of Applied Physics* 115 (9) (2014) 094301 (2014).
484 doi:10.1063/1.4867218.
- 485 [15] K. Toko, K. Nakazawa, N. Saitoh, N. Yoshizawa, T. Sue-
486 masu, Improved surface quality of the metal-induced crystal-
487 lized Ge seed layer and its influence on subsequent epitaxy,
488 *Crystal Growth & Design* 15 (3) (2015) 1535–1539 (2015).
489 doi:10.1021/acs.cgd.5b00060.
- 490 [16] J.-H. Park, T. Suzuki, M. Kurosawa, M. Miyao, T. Sadoh,
491 Nucleation-controlled gold-induced-crystallization for selective
492 formation of Ge(100) and (111) on insulator at low-temperature
493 (250C), *Applied Physics Letters* 103 (8) (2013) 082102 (2013).
494 doi:10.1063/1.4819015.
- 495 [17] N. Oya, K. Toko, N. Saitoh, N. Yoshizawa, T. Suemasu, Effects
496 of flexible substrate thickness on Al-induced crystallization of
497 amorphous Ge thin films, *Thin Solid Films* 583 (2015) 221–225
498 (2015). doi:10.1016/j.tsf.2015.03.072.
- 499 [18] S. Hu, P. W. Leu, A. F. Marshall, P. C. McIntyre, Single-
500 crystal germanium layers grown on silicon by nanowire seed-
501 ing, *Nature Nanotechnology* 4 (10) (2009) 649–653 (2009).
502 doi:10.1038/nnano.2009.233.
- 503 [19] S. Kühnappel, S. Gall, B. Rech, D. Amkreutz, Towards
504 monocrystalline silicon thin films grown on glass by liquid
505 phase crystallization, *Solar Energy Materials and Solar Cells*
506 140 (2015) 86–91 (2015). doi:10.1016/j.solmat.2015.03.030.
- 507 [20] K. Toko, T. Tanaka, T. Sadoh, M. Miyao, Formation of single-
508 crystalline Ge stripes on quartz substrates by SiGe mixing-
509 triggered liquid-phase epitaxy, *Thin Solid Films* 518 (6) (2010)
510 S179–S181 (2010). doi:10.1016/j.tsf.2009.10.083.
- 511 [21] Y. Cohin, O. Mauguin, L. Largeau, G. Patriarche, F. Glas,
512 E. Søndergård, J.-C. Harmand, Growth of vertical GaAs
513 nanowires on an amorphous substrate via a fiber-textured
514 Si platform, *Nano Letters* 13 (6) (2013) 2743–2747 (2013).
515 doi:10.1021/nl400924c.
- 516 [22] D. Ren, I. M. Høiaas, J. F. Reinertsen, D. L. Dheeraj, A. M.
517 Munshi, D.-C. Kim, H. Weman, B.-O. Fimland, Growth opti-
518 mization for self-catalyzed GaAs-based nanowires on metal-
519 induced crystallized amorphous substrate, *Journal of Vacuum Science & Technology B, Nanotechnology and Microelectronics: Materials, Processing, Measurement, and Phenomena* 34 (2) (2016) 02L117 (2016). doi:10.1116/1.4943926.
- 520 [23] K. Toko, M. Nakata, W. Jevasuwan, N. Fukata, T. Suemasu,
521 Vertically aligned Ge nanowires on flexible plastic films syn-
522 thesized by (111)-oriented Ge seeded vapor-liquid-solid growth,
523 *ACS Applied Materials & Interfaces* 7 (32) (2015) 18120–
524 18124 (2015). doi:10.1021/acsami.5b05394.
- 525 [24] V. Dhaka, T. Haggren, H. Jussila, H. Jiang, E. Kauppinen,
526 T. Huhtio, M. Sopanen, H. Lipsanen, High quality GaAs
527 nanowires grown on glass substrates, *Nano Letters* 12 (4) (2012)
528 1912–1918 (2012). doi:10.1021/nl204314z.
- 529 [25] M. G. Mauk, Low-cost III-V compound semiconductor solar
530 cells, in: *Handbook of Research on Solar Energy Systems and Technologies*, IGI Global, 2012, pp. 254–293 (2012).
531 doi:10.4018/978-1-4666-1996-8.ch010.
- 532 [26] R. Venkatasubramanian, B. C. O. Quinn, E. Siivola, B. Keyes,
533 R. Ahrenkiel, 20% (AM1.5) efficiency GaAs solar cells on sub-
534 mm grain-size poly-Ge and its transition to low-cost substrates,
535 in: *Conference Record of the Twenty Sixth IEEE Photovoltaic Specialists Conference - 1997*, IEEE, 1997, pp. 811–814 (1997).
536 doi:10.1109/pvsc.1997.654211.
- 537 [27] Y. Wang, Z. Ren, M. Thway, K. Lee, S. F. Yoon, I. M.
538 Peters, T. Buonassisi, E. A. Fitzgerald, C. S. Tan, K. H.
539 Lee, Fabrication and characterization of single junction GaAs
540 solar cells on Si with As-doped Ge buffer, *Solar Energy Materials and Solar Cells* 172 (2017) 140–144 (2017).

- 553 doi:10.1016/j.solmat.2017.07.028. 618
- 554 [28] T. Nishida, K. Moto, N. Saitoh, N. Yoshizawa, T. Suemasu, 619
- 555 K. Toko, High photoresponsivity in a GaAs film synthe- 620
- 556 sized on glass using a pseudo-single-crystal Ge seed layer, 621
- 557 Applied Physics Letters 114 (14) (2019) 142103 (2019). 622
- 558 doi:10.1063/1.5091714. 623
- 559 [29] D. Pelati, G. Patriarche, O. Mauguin, L. Largeau, F. Brisset, 624
- 560 F. Glas, F. Oehler, In situ optical monitoring of new path- 625
- 561 ways in the metal-induced crystallization of amorphous Ge, 626
- 562 Crystal Growth & Design 17 (11) (2017) 5783–5789 (2017). 627
- 563 doi:10.1021/acs.cgd.7b00799. 628
- 564 [30] R. Numata, K. Toko, K. Nakazawa, N. Usami, T. Sue- 618
- 565 masu, Growth promotion of Al-induced crystallized Ge films 619
- 566 on insulators by insertion of a Ge membrane below the 620
- 567 Al layer, Thin Solid Films 557 (2014) 143–146 (2014). 621
- 568 doi:10.1016/j.tsf.2013.08.040. 622
- 569 [31] R. Numata, K. Toko, N. Oya, N. Usami, T. Suemasu, Struc- 623
- 570 tural characterization of polycrystalline Ge thin films on insula- 624
- 571 tors formed by diffusion-enhanced Al-induced layer exchange, 625
- 572 Japanese Journal of Applied Physics 53 (4S) (2014) 04EH03 626
- 573 (2014). doi:10.7567/jjap.53.04eh03. 627
- 574 [32] D. Pelati, O. Mauguin, L. Largeau, F. Brisset, F. Glas, F. Oehler, 628
- 575 Kinetics and crystal texture improvements on thin germanium 618
- 576 layers obtained by aluminium induced crystallization using oxy- 619
- 577 gen plasma, Surface and Coatings Technology 343 (2018) 121– 620
- 578 126 (2018). doi:10.1016/j.surfcoat.2017.10.034. 621
- 579 [33] D. V. Gestel, I. Gordon, A. Verbist, L. Carnel, G. Beaucarne, 622
- 580 J. Poortmans, A new way to selectively remove Si islands from 623
- 581 polycrystalline silicon seed layers made by aluminum-induced 624
- 582 crystallization, Thin Solid Films 516 (20) (2008) 6907–6911 625
- 583 (2008). doi:10.1016/j.tsf.2007.12.122. 626
- 584 [34] D. Pelati, G. Patriarche, O. Mauguin, L. Largeau, L. Travers, 627
- 585 F. Brisset, F. Glas, F. Oehler, GaAs (111) epilayers 628
- 586 grown by MBE on Ge (111): Twin reduction and polar- 618
- 587 ity, Journal of Crystal Growth 519 (2019) 84–90 (2019). 619
- 588 doi:10.1016/j.jcrysgro.2019.05.006. 620
- 589 [35] P. Pukite, P. Cohen, Suppression of antiphase domains in 621
- 590 the growth of GaAs on Ge(100) by molecular beam epitaxy, 622
- 591 Journal of Crystal Growth 81 (1-4) (1987) 214–220 (1987). 623
- 592 doi:10.1016/0022-0248(87)90393-9. 624
- 593 [36] R. M. Sieg, S. A. Ringel, S. M. Ting, E. A. Fitzgerald, R. N. 625
- 594 Sacks, Anti-phase domain-free growth of GaAs on offcut (001) 626
- 595 Ge wafers by molecular beam epitaxy with suppressed Ge out- 627
- 596 diffusion, Journal of Electronic Materials 27 (7) (1998) 900–907 628
- 597 (1998). doi:10.1007/s11664-998-0116-1. 618
- 598 [37] S. Koh, T. Kondo, Y. Shiraki, R. Ito, GaAs/Ge/GaAs sublattice 619
- 599 reversal epitaxy and its application to nonlinear optical devices, 620
- 600 Journal of Crystal Growth 227-228 (2001) 183–192 (2001). 621
- 601 doi:10.1016/s0022-0248(01)00660-1. 622
- 602 [38] Q. Li, J. L. Krauss, S. Hersee, S. M. Han, Probing interactions 623
- 603 of Ge with chemical and thermal SiO₂ to understand selective 624
- 604 growth of Ge on Si during molecular beam epitaxy, The Jour- 625
- 605 nal of Physical Chemistry C 111 (2) (2007) 779–786 (2007). 626
- 606 doi:10.1021/jp062966o. 627
- 607 [39] S. Plissard, G. Larrieu, X. Wallart, P. Caroff, High yield of self- 628
- 608 catalyzed GaAs nanowire arrays grown on silicon via gallium 618
- 609 droplet positioning, Nanotechnology 22 (27) (2011) 275602 619
- 610 (2011). doi:10.1088/0957-4484/22/27/275602. 620
- 611 [40] A. M. Munshi, D. L. Dheeraj, V. T. Fauske, D. C. Kim, 621
- 612 J. Huh, J. F. Reinertsen, L. Ahtapodov, K. D. Lee, B. Heidari, 622
- 613 A. T. J. van Helvoort, B. O. Fimland, H. Weman, Position- 623
- 614 controlled uniform GaAs nanowires on silicon using nanoim- 624
- 615 print lithography, Nano Letters 14 (2) (2014) 960–966 (2014). 625
- 616 doi:10.1021/nl404376m. 626
- 617 [41] Y. Kajikawa, Y. Son, H. Hayase, H. Ichiba, R. Mori, K. Ushi- 627
- rogouchi, M. Irie, Suppression of twin generation in the growth 628
- of GaAs on Ge (111) substrates, Journal of Crystal Growth 477 618
- (2017) 40–44 (2017). doi:10.1016/j.jcrysgro.2016.12.062. 619
- [42] D. Hull, D. J. Bacon, Introduction to dislocations (Fifth Edition), 620
- Butterworth-Heinemann, 2011 (2011). doi:10.1016/B978-0-08- 621
- 096672-4.00019-0. 622
- [43] T. Sadoh, J.-H. Park, R. Aoki, M. Miyao, Low-temperature 623
- (300C) formation of orientation-controlled large-grain (10mi- 624
- cron) ge-rich SiGe on insulator by gold-induced crys- 625
- tallization, Thin Solid Films 602 (2016) 3–6 (2016). 626
- doi:10.1016/j.tsf.2015.10.057. 627