Heterogeneous growth of single crystals on polycrystals

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This work discloses a surprising, previously unknown heterogeneous growth mode. Namely, large-area, thin sheets of *single-crystalline* Ge were observed to grow laterally on top of a *polycrystalline* Al substrate, covering as many as tens of differently oriented Al grains at low temperatures. The observation of the Ge crystal-growth process by *in situ* heating transmission electron microscopy demonstrates an intriguing type of “faceted” growth: the growth of *single-crystalline* Ge thin sheets proceeding *Al-grain by Al-grain* on top of the polycrystalline Al substrate. The crystalline Ge growth front tends to align along the lines of intersection of the Al grain boundaries with the Al surface. Such an unusual heterogeneous growth mode has been shown to be a consequence of the strong anisotropy of the energy of the crystalline/crystalline (here: c-Ge/c-Al) interfaces.

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I. INTRODUCTION

Heterogeneous epitaxial growth of crystalline thin films on crystalline substrates is a phenomenon of crucial importance for components in numerous modern technologies, such as microelectronic and magnetic storage devices, solid-state lasers, and light-emitting diodes (see Ref. [1] and references therein). It is well known that the heterogeneous epitaxial growth is largely controlled by the (mis)match of the lattices of the film and the substrate, based on which different types of film-substrate orientation relationship may occur (e.g., cube-on-cube [2–4], domain epitaxy [5], Bain orientation relationship [6], etc.). Thereby the orientation, the quality, and the properties of the film can be controlled by selection of suitable single-crystal substrates of specific surface orientations [1,5].

In this paper, a surprising, previously unknown heterogeneous growth mode is disclosed. Large-area, thin sheets of *single-crystalline* Ge (c-Ge) were observed to grow laterally on the surface of a *polycrystalline* Al substrate, covering as many as tens of differently oriented Al grains of the Al substrate. The investigation of the c-Ge growth process by *in situ* transmission electron microscopy (TEM) revealed an intriguing type of “faceted” growth: the growth of single-crystalline Ge thin sheets proceeds *Al-grain by Al-grain* on top of the polycrystalline Al substrate. The c-Ge growth front tends to align along the lines of intersection of Al grain boundaries (GBs) with the surface of the contacting *polycrystalline* Al substrate. Such an unusual heterogeneous growth mode was shown to be a consequence of the pronounced anisotropy of the energy of the crystalline/crystalline (here: c-Ge/c-Al) interfaces. Thereby a route is opened to the controlled growth of single crystals at low temperatures on polycrystalline substrates.

II. EXPERIMENTAL PROCEDURES

Specimen preparation was carried out in an ultrahigh vacuum (UHV) molecular beam epitaxy system (base pressure $< 2 \times 10^{-10}$ mbar). First, a 50-nm-thick polycrystalline Al (c-Al) thin film was deposited at room temperature (RT) on a 20-nm thin amorphous SiO₂ membrane substrate (SiMPore, Inc.) by thermal evaporation from a pyrolytic boron nitride crucible filled with pure Al (purity ≥ 99.95 wt%). Then, without breaking the UHV, a 15-nm-thick amorphous Ge (a-Ge) layer was deposited on top of the Al sublayer at RT by thermal evaporation from a graphite crucible filled with pure Ge (purity ≥ 99.999 wt%). The growth rates of each sublayer were calibrated separately by employing spectroscopic ellipsometry (J.A. Woollam M-2000 ellipsometer) or surface profiling (Veeco Dektak 8 profilometer) measurements of the layer thicknesses for various growth times. The thus-deposited a-Ge/c-Al bilayer specimens were taken out of the growth chamber and directly used for subsequent *in situ* heating analytic TEM investigations.

The *in situ* heating energy-filtered TEM (EF-TEM) experiments were carried out in a Zeiss Sub-eV-Sub-Ångström Microscope, which is operated at 200 keV and equipped with an electrostatic monochromator and a MANDOLINE filter [7]. The specimen was heated *in situ* using a Gatan 652 double-tilt heating stage in the microscope in the temperature range of 100 °C to 150 °C. The temperature of the holder tip was monitored by a calibrated silicon diode mounted on the holder tip, which provides a sensitive temperature response with a precision of 0.5 °C.

III. RESULTS AND DISCUSSION

Plan-view energy-filtered TEM images of an as-deposited 15-nm a-Ge/50-nm c-Al specimen are shown in Fig. 1(a), taken at zero electron-energy loss and at electron-energy loss $\Delta E = -15$ eV (corresponding to the bulk plasmon absorption of Al [8]). Since the Ge sublayer is amorphous, the

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FIG. 1. (a) Plan-view zero-loss (left-hand side) and energy-filtered (at $\Delta E = -15$ eV) TEM images of an as-deposited 15-nm a-Ge/50-nm c-Al bilayer specimen, showing the columnar grain structure of the polycrystalline Al film. (b) Plan-view zero-loss (left-hand side) and energy-filtered (at $\Delta E = -15$ eV; right-hand side) TEM images of a 15-nm a-Ge/50-nm c-Al bilayer after *in situ* heating at 150 °C for 15 min. In the energy-filtered ($\Delta E = -15$ eV) TEM image, Al-rich regions appear bright and Ge-rich regions appear dark.

contrast in these TEM images reveals clearly the (columnar) polycrystalline grain structure of the c-Al sublayer. After *in situ* annealing of the bilayer at 150 °C for 15 min, crystallization of a-Ge has occurred, as evidenced by the corresponding TEM images shown in Fig. 1(b) (see what follows). The here-observed a-Ge crystallization temperature of 150 °C is much lower than the bulk crystallization temperature of a-Ge, which is as high as 500 °C [9]. Such a phenomenon is known as so-called metal-induced crystallization [10,11]. The EF-TEM image at $\Delta E = -15$ eV is shown at the right-hand side of Fig. 1(b), in which Al-rich regions appear bright and Ge-rich regions appear dark. The Ge-rich regions are crystalline, as follows from the diffraction contrast observed in the corresponding zero-loss TEM image shown at the left-hand side of Fig. 1(b).

Each separate c-Ge region (c-Ge thin “sheet”), which is about 0.5–1 μm large and 20-nm thick, is *single crystalline* as demonstrated by high-resolution transmission electron microscopy (HRTEM) (see, e.g., Fig. 2, and Figs. S1–S3 of the Supplemental Material [12]). The single-crystalline c-Ge sheets have developed laterally on top of the polycrystalline Al substrate, apparently *Al-grain by Al-grain*, with *curved and “faceted”* growth fronts which run more or less along the lines of intersection of the Al GBs with the Al surface [see, e.g., Fig. 1(b)]. The surfaces of some of the Al grains are “bypassed” by the heterogeneously growing c-Ge sheet, leading to the formation of holes in the grown c-Ge sheet.

FIG. 2. Plan-view HRTEM analyses of a 15-nm a-Ge/50-nm c-Al bilayer after *in situ* heating at 150 °C for 15 min. The image shows two representative, separately developed large thin “sheets” of single-crystalline Ge (1# and 2#). The lateral growth of the thin c-Ge sheets became stagnant at certain lines of intersection of the Al grain boundaries with the Al surface (see insets *i*, *ii*, and *iii*).

The “shadows”/presence of many Al grains at (behind) the projected location of the c-Ge thin sheets can be discerned in Fig. 1(b) [see also Figs. 3(a) and 3(b)]. Energy-dispersive x-ray spectroscopy measurements of the same specimens showed that the original a-Ge has largely been depleted (on average ~ 98 at.% Al, ~ 2 at.% Ge) at the projected location outside the c-Ge sheets, whereas Al and Ge both occur (~ 65 at.% Al, ~ 35 at.% Ge) at the projected location where c-Ge sheets have developed.

A detailed HRTEM analysis of the c-Ge thin sheets formed upon *in situ* annealing of the a-Ge/c-Al specimen at 150 °C for 15 min is given in Fig. 2. The representative image shows two separately developed large thin Ge sheets (denoted 1 and 2). Both Ge sheets in the image have been shown to be single crystalline but of different crystal orientation. A Ge GB has formed at the location where the two Ge sheets have come into contact (marked by the green dashed line in Fig. 2). Evidently, the lateral growth of the thin c-Ge sheets has become stagnant at certain lines of intersection of the Al GBs with the surface of the contacting Al substrate, leading to the formation of “faceted” and curved growth fronts of the c-Ge sheets. Two Al grains can be seen at the (lateral) location between the two c-Ge sheets (see center of Fig. 2). Two small “bypassed” Al grains are observable within the c-Ge sheet 1.

A detailed EF-TEM analysis of a c-Ge sheet formed upon heating at 150 °C for 15 min is presented in Figs. 3(a)–3(c). It confirms indeed that the c-Ge sheet is growing on top of the polycrystalline Al substrate *Al-grain by Al-grain like* [see, in particular, the EF-TEM image at $\Delta E = -15$ eV in Fig. 3(b)]. The stagnation of the growth of the c-Ge sheet [its growth front is outlined in green in Figs. 3(a) and 3(b)] at the projected locations of many Al GBs is clearly observable. The formed

FIG. 3. (a)–(c) Plan-view (a) zero-loss, (b) energy-filtered at $\Delta E = -15$ eV TEM images and (c) map of the FWHM of the plasmon-loss peak, taken from a 15-nm a-Ge/50-nm c-Al bilayer after *in situ* heating at 150 °C for 15 min. (d), (e) Plan-view (d) zero-loss, (e) energy-filtered at $\Delta E = -15$ eV TEM images, and (f) map of the FWHM of the plasmon-loss peak, taken from a 15-nm a-Ge/50-nm c-Al bilayer after *in situ* heating at 100 °C for 15 min. Note that the FWHM of the plasmon-loss peak of Al is much narrower than that of Ge, and thereby the Ge-rich regions (i.e., the c-Ge sheets) can be very well discerned (in red) in the FWHM mappings in (c) and (f).

c-Ge sheet shown in the center of Fig. 3(b) has covered more than ten Al grains.

This intriguing heterogeneous growth mode has also been observed at an even lower temperature of only 100 °C. The EF-TEM analysis of a 15-nm a-Ge/50-nm c-Al specimen annealed at 100 °C for 15 min is shown in Figs. 3(d)–3(f). Similar to the c-Ge sheets formed at 150 °C, the c-Ge sheets formed at 100 °C appear to grow also *Al-grain by Al-grain*, and thus show a “faceted” grain front. As compared to the results at 150 °C, the c-Ge sheet morphology at 100 °C is more branchlike [compare Figs. 3(d)–3(f) and Figs. 3(a)–3(c)]. This may indicate that, at the lower temperature, less Al grains are “active” (i.e., associated with a relatively low energy barrier which can be overcome; see discussion below) for the Al-induced crystallization of a-Ge.

The observed heterogeneous growth mode (i.e., growth of *single* crystals on *polycrystals*) has not been reported before. The mechanism for such an unusual crystal-growth mode is discussed in the following; see Fig. 4.

The first step of the heterogeneous growth is the nucleation of a Ge crystallite on top of an Al grain [grain 0, Fig. 4(a)]. This *interface-mediated* nucleation of c-Ge has been shown to be possible to occur at the a-Ge/c-Al interface at very low temperatures (≥ 50 °C) on the basis of interface thermodynamic calculation [9,13]. The critical thickness above which the bulk Gibbs energy overcompensates the increase of the interface energies upon crystallization is smaller than the thickness of a-Ge adjacent to the metal of weakened covalent bonds and thus of increased atomic mobility [14]. Then, as shown in Fig. 4(b), the c-Ge may grow laterally towards and on top of the surface of the Al grains (grains 1–7) surrounding grain 0. The crystallization of a-Ge will be retarded by a relatively high interface energy of the formed c-Ge/c-Al interface due to

crystallization of a-Ge ($\gamma_{\text{Ge}}^i|_{\text{Al}}$) [15], with an *average* value of about 0.35 J m^{-2} at 100 °C [13], as compared to that of the original a-Ge/c-Al interface ($\gamma_{\text{Ge}}^i|_{\text{Al}}$) [15], $\sim 0.05 \text{ J m}^{-2}$ at 100 °C [13]. Depending on the interface energy of c-Ge with different Al grains ($\gamma_{\text{Ge}}^i|_{\text{Al}}$, $i = 1 - 7$), the growth of c-Ge proceeds preferentially across the Al grains associated with *lower* interface energies $\gamma_{\text{Ge}}^i|_{\text{Al}}$ (in this drawing, grains 2, 4, and 5) and becomes stagnant at the Al GBs towards Al grains associated with relatively high $\gamma_{\text{Ge}}^i|_{\text{Al}}$ [in this drawing, grains 1, 3, 6, and 7; see also Fig. 4(d) for a schematic cross-sectional view]. Subsequently, as shown in Fig. 4(c), the c-Ge continues to grow, now from grains 2, 4, and 5, towards the surface of adjacent Al grains associated with relatively low $\gamma_{\text{Ge}}^i|_{\text{Al}}$, thus forming a faceted thin sheet morphology of c-Ge as observed in Figs. 2 and 3. Since $\gamma_{\text{Ge}}^i|_{\text{Al}}$ increases with decreasing temperatures [13], at relatively low temperatures only few (say, one or two) of the adjacent Al grains could be energetically favorable enough for the growth of the c-Ge sheet above. This would lead to the formation of more branch-shaped c-Ge, as observed in Figs. 3(d)–3(f) ($T = 100$ °C). At higher temperatures, more (say, three to five) of the adjacent Al grains could energetically accept to be covered with c-Ge, i.e., allow the growth of the c-Ge sheet on top. This would lead to the formation of more compacted c-Ge sheets, as observed in Figs. 2 and 3(a)–3(c) ($T = 150$ °C).

After growth stagnation of c-Ge at the intersection of an Al GB with the Al surface, it is energetically more preferred that the Ge atoms of the a-Ge on the adjacent Al grain [grain 1 in the drawing of Figs. 4(d) and 4(e)] now diffuse along the a-Ge/c-Al interface [14] towards the c-Ge on grain 0, causing growth of the c-Ge sheet on grain 0 perpendicular to the surface (i.e., thickening) and depletion of a-Ge on Al grain 1, as experimentally observed (Figs. 1 and 3).

FIG. 4. Schematic illustration of the growth mechanism of a single-crystalline sheet of Ge (red) on top of a polycrystalline Al substrate, (a)–(c) a top view, (d), (e) a cross-sectional view: (a) first c-Ge is nucleated locally on top of an Al grain (grain 0); (b) the c-Ge grows towards (and on top of) the surface of the Al grains (grains 1–7) surrounding grain 0. Depending on the interface energy of c-Ge on different Al grains ($\gamma_{\langle \text{Ge} \rangle | \langle \text{Al} \rangle}^i$), the growth of c-Ge proceeds preferentially across the Al grains with lower $\gamma_{\langle \text{Ge} \rangle | \langle \text{Al} \rangle}^i$ (in this drawing, grains 2, 4, and 5), and becomes stagnant at the Al GBs towards Al grains with very high $\gamma_{\langle \text{Ge} \rangle | \langle \text{Al} \rangle}^i$ (in this drawing, grains 1, 3, 6, and 7). (c) The c-Ge continues to grow, now from grains 2, 4, and 5, towards the surface of adjacent Al grains with relatively low $\gamma_{\langle \text{Ge} \rangle | \langle \text{Al} \rangle}^i$, thus forming a faceted thin sheet morphology of c-Ge, as, e.g., observed in Fig. 3. (d) A cross-sectional view: the growth front of c-Ge reaches the location of intersection of an Al GB with the Al surface. If the interface energy of c-Ge on the adjacent Al grain (grain 1), $\gamma_{\langle \text{Ge} \rangle | \langle \text{Al} \rangle}^1$, is relatively high, the growth of c-Ge will become stagnant at the Al GB between grains 0 and 1. (e) Upon the c-Ge growth stagnation at the Al GB, Ge atoms in the a-Ge layer on top of Al grain 1 will diffuse along the a-Ge/Al interface towards the c-Ge sheet on Al grain 0, thus leading to the growth of a c-Ge sheet perpendicular to the surface and the depletion of a-Ge on Al grain 1.

A key point of the above growth mechanism is the *anisotropy* of the energy of the crystalline/crystalline interface, which involves that for a single-crystalline Ge sheet (i.e., with a fixed crystal orientation), the c-Ge/c-Al interface energy is much dependent on the orientation of the underlying c-Al. The orientations of the (c-Al) grains in the polycrystalline (Al) film grown on an amorphous substrate have been known to be close to *random* [16]. Already more or less straightforward considerations make clear that the surface energy of a crystalline solid is significantly anisotropic, with relatively low surface energy for the more close-packed surfaces [17]. However, such treatments become much more complex considering the interface energy of a crystalline/crystalline interface [18–22].

According to Ref. [23], the $\langle A \rangle | \langle B \rangle$ interface energy is given by

$$\gamma_{\langle A \rangle | \langle B \rangle} = \gamma_{\langle A \rangle | \langle B \rangle}^{\text{mis}} + \gamma_{\langle A \rangle | \langle B \rangle}^{\text{chem}}, \quad (1)$$

where the mismatch energy term $\gamma_{\langle A \rangle | \langle B \rangle}^{\text{mis}}$ originates from the structural mismatch between the lattices of $\langle A \rangle$ and $\langle B \rangle$, and the chemical energy term $\gamma_{\langle A \rangle | \langle B \rangle}^{\text{chem}}$ originates from the chemical interaction of materials $\langle A \rangle$ and $\langle B \rangle$.

If A and B are the same material, the $\gamma_{\langle A \rangle | \langle A \rangle}^{\text{chem}}$ term is zero, and it follows for the grain-boundary energy $\gamma_{\langle A \rangle}^{\text{GB}}$,

$$\gamma_{\langle A \rangle}^{\text{GB}} \equiv \gamma_{\langle A \rangle | \langle A \rangle} = \gamma_{\langle A \rangle | \langle A \rangle}^{\text{mis}}. \quad (2)$$

The chemical interaction energy term $\gamma_{\langle A \rangle | \langle B \rangle}^{\text{chem}}$ depends on the atomic density at the interface plane [23] and should therefore be anisotropic.

The mismatch term $\gamma_{\langle A \rangle | \langle B \rangle}^{\text{mis}}$ is often discussed in literature to be quasi-isotropic for a “general” (i.e., except coincident-site-lattice interfaces) high-angle solid-solid interfaces (including GBs), with an average value of [23]

$$\gamma_{\langle A \rangle | \langle B \rangle}^{\text{mis}} = \frac{1}{6} (\gamma_{\langle A \rangle}^{\text{S}} + \gamma_{\langle B \rangle}^{\text{S}}) \quad (3)$$

and thus

$$\gamma_{\langle A \rangle}^{\text{GB}} = \frac{1}{3} \gamma_{\langle A \rangle}^{\text{S}}. \quad (4)$$

Recent theoretical work [19,20] has, however, indicated the occurrence of significant GB energy anisotropy in face-centered-cubic and body-centered-cubic metals: the GB energy $\gamma_{\langle A \rangle}^{\text{GB}}$ depends strongly on the misorientation angle and the GB plane orientation, with an average value close to that predicted according to Eq. (4). Analogously, the $\gamma_{\langle A \rangle | \langle B \rangle}^{\text{mis}}$ should also be strongly anisotropic.

As a result of the above considerations, it must be expected that both $\gamma_{\langle A \rangle | \langle B \rangle}^{\text{mis}}$ and $\gamma_{\langle A \rangle | \langle B \rangle}^{\text{chem}}$, and hence $\gamma_{\langle A \rangle | \langle B \rangle}$, are (strongly) anisotropic. Indeed, using the methods described in Refs. [23,24], it is calculated that a semicoherent Al(111)/Ge(111) interface has an energy of about 0.24 J m^{-2} , which is significantly smaller than the average energy of about 0.35 J m^{-2} for “general” $\langle \text{Al} \rangle / \langle \text{Ge} \rangle$ interfaces [13].

Reversely, the heterogeneous growth mode observed in this project provides strong experimental proof for the occurrence of crystalline/crystalline $\langle A \rangle / \langle B \rangle$ interface energy anisotropy.

IV. CONCLUSIONS

An unusual heterogeneous crystal-growth mode is disclosed by which relatively large *single*-crystalline thin sheets can be grown on top of *polycrystals*. The growth of the single-crystalline thin sheets proceeds laterally on top of the polycrystalline substrate, in a substrate-*grain-by-grain* manner. Such a growth mode is controlled fundamentally by the anisotropy of the crystalline/crystalline interface energies. This growth mode may enable the development of single-

crystalline devices at low temperatures on polycrystalline substrates.

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- [1] H. Brune, in *Surface and Interface Science* (Wiley-VCH Verlag GmbH & Co, KGaA, Weinheim, Germany, 2013), p. 421.
 - [2] F. C. Frank and J. H. van der Merwe, *Proc. R. Soc. Lond. A* **198**, 216 (1949).
 - [3] J. W. Matthews and A. E. Blakeslee, *J. Cryst. Growth* **27**, 118 (1974).
 - [4] E. A. Fitzgerald, *Mater. Sci. Rep.* **7**, 87 (1991).
 - [5] J. Narayan and B. C. Larson, *J. Appl. Phys.* **93**, 278 (2003).
 - [6] J. Buschbeck, I. Opahle, M. Richter, U. K. Röbber, P. Klaer, M. Kallmayer, H. J. Elmers, G. Jakob, L. Schultz, and S. Fähler, *Phys. Rev. Lett.* **103**, 216101 (2009).
 - [7] C. T. Koch, W. Sigle, R. Höschen, M. Rühle, E. Essers, G. Benner, and M. Matijevic, *Microsc. Microanal.* **12**, 506 (2006).
 - [8] R. F. Egerton, *Electron Energy-Loss Spectroscopy in the Electron Microscope*, 2nd ed. (Plenum Press, New York, 1996).
 - [9] Z. M. Wang, L. P. H. Jeurgens, J. Y. Wang, and E. J. Mittemeijer, *Adv. Eng. Mater.* **11**, 131 (2009).
 - [10] Z. M. Wang, J. Y. Wang, L. P. H. Jeurgens, and E. J. Mittemeijer, *Phys. Rev. Lett.* **100**, 125503 (2008).
 - [11] *Metal-Induced Crystallization: Fundamentals and Applications*, edited by Z. M. Wang, L. P. H. Jeurgens, and E. J. Mittemeijer (Pan Stanford, Singapore, 2015).
 - [12] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.95.094109> for plan-view bright-field/dark-field TEM and HRTEM characterization of the c-Ge nanosheets.
 - [13] Z. M. Wang, J. Y. Wang, L. P. H. Jeurgens, and E. J. Mittemeijer, *Phys. Rev. B* **77**, 045424 (2008).
 - [14] Z. M. Wang, L. P. H. Jeurgens, W. Sigle, and E. J. Mittemeijer, *Phys. Rev. Lett.* **115**, 016102 (2015).
 - [15] In this paper, “ $\langle \rangle$ ” denotes the crystalline phase whereas “ $\{ \}$ ” denotes the amorphous phase.
 - [16] X-ray diffraction pole-figure measurements performed in this project have shown that the deposited Al film has a weak (111) texture with fully random in-plane orientations.
 - [17] I. Galanakis, N. Papanikolaou, and P. H. Dederichs, *Surf. Sci.* **511**, 1 (2002).
 - [18] A. P. Sutton and R. W. Balluffi, *Interfaces in Crystalline Materials* (Oxford University Press, Oxford, UK, 1995).
 - [19] D. L. Olmsted, S. M. Foiles, and E. A. Holm, *Acta Mater.* **57**, 3694 (2009).
 - [20] G. S. Rohrer, *J. Mater. Sci.* **46**, 5881 (2011).
 - [21] V. V. Bulatov, B. W. Reed, and M. Kumar, *Acta Mater.* **65**, 161 (2014).
 - [22] P. R. M. van Beers, V. G. Kouznetsova, M. G. D. Geers, M. A. Tschopp, and D. L. McDowell, *Acta Mater.* **82**, 513 (2015).
 - [23] L. P. H. Jeurgens, Z. M. Wang, and E. J. Mittemeijer, *Int. J. Mater. Res.* **100**, 1281 (2009).
 - [24] F. Reichel, L. P. H. Jeurgens, and E. J. Mittemeijer, *Phys. Rev. B* **74**, 144103 (2006).

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Figure S1. Plan-view **(a)** bright-field and **(b)** corresponding dark-field TEM images of a 15-nm a-Ge/50-nm c-Al bilayer after *in situ* heating at 150 °C for 15 min. The dark-field image has been taken by positioning the objective aperture on a diffraction spot corresponding to c-Ge. The dark-field TEM has confirmed the single-crystalline structure of a representative, large c-Ge sheet developed on top of polycrystalline Al.

Figure S2. Plan-view HRTEM image of a 15-nm a-Ge/50-nm c-Al bilayer after *in situ* heating at 150 °C for 15 min. The image confirms the single-crystalline structure of a large Ge sheet developed on top of polycrystalline Al.

Figure S3. Another plan-view HRTEM image of a 15-nm a-Ge/50-nm c-Al bilayer after *in situ* heating at 150 °C for 15 min. The image confirms the single-crystalline structure of a large Ge sheet developed on top of polycrystalline Al.