Electronic Supplementary Material (ESI) for CrystEngComm.

Supplementary for:

Growth mechanism of aluminium-induced solid phase epitaxial (AI-SPE) $Si_{0.5}Ge_{0.5}$ layer using in-situ heating transmission electron microscopy

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Table S1. The Si and Ge parameters excerpt from reference 1 for thermodynamic analysis[1]. Based on Si and Ge parameters, the energies of $Si_{0.75}Ge_{0.25}$, $Si_{0.50}Ge_{0.50}$, and $Si_{0.25}Ge_{0.75}$ are approximately value depending on stoichiometry[2]. The detailed formulas are given in reference 1 and summarized in following.

Crystalline phase	$\langle Si \rangle$	$\langle Si_{0.75}Ge_{0.25} \rangle$	$\langle Si_{0.50}Ge_{0.50} \rangle$	$\langle Si_{0.25}Ge_{0.75} \rangle$	$\langle Ge \rangle$	$\langle Al \rangle$
$\gamma^s(T_0)(J\,m^{-2})$	1.37	1.27	1.17	1.07	0.97	1.03
$T_0(K)$	493	488	483	478	473	500
V_m $(\times 10^{-6} m^3 mol^{-1})$	12.13	12.51	12.89	13.27	13.65	10.06
Amorphous phase	$\{Si\}$	$\{Si_{0.75}Ge_{0.25}\}$	$\{Si_{0.50}Ge_{0.50}\}$	$\{Si_{0.25}Ge_{0.75}\}$	$\{Ge\}$	
$\gamma^s(T_0)(J\,m^{-2})$	0.87	0.81	0.75	0.68	0.62	
$T_0(K)$	1685	1567	1448	1330	1211	
V_m $(\times 10^{-6} m^3 mol^{-1})$	11.10	11.57	12.03	12.50	12.97	

$\Delta H^0_{\langle Al \rangle \text{ in } \langle Si \rangle}$	$\Delta H^0_{\langle Si \rangle \text{ in } \langle Al \rangle}$	$\Delta H^0_{Si(diam) \to Si(fcc)}$	$\Delta S_{Si(diam) \to Si(fcc)}$	$\Delta H^0_{\{Al\} \text{ in } \{Si\}}$ $\Delta H^0_{\{Si\} \text{ in } \{Al\}}$	Θ_{Si}	$\Delta H^m_{\{Al\}}$
47857 $(J\,mol^{-1})$	- 3143 $(J\,mol^{-1})$	51000 $(J\,mol^{-1})$	21.8 $(J\,mol^{-1}K^{-1})$	-11304 $(J\,mol^{-1})$	645 K	10784.4 $(J\,mol^{-1})$
$\Delta H^0_{\langle Al \rangle \text{ in } \langle Si_{0.75}Ge_{0.25} \rangle}$	$\Delta H^0_{\langle Si_{0.75}Ge_{0.25} \rangle \text{ in } \langle Al \rangle}$	$\Delta H^0_{Si_{0.75}Ge_{0.25}(diam) \to Si_{0.75}Ge_{0.25}(fcc)}$	$\Delta S_{Si_{0.75}Ge_{0.25}(diam) \to Si_{0.75}Ge_{0.25}(fcc)}$	$\Delta H^0_{\{Al\} \text{ in } \{Si_{0.75}Ge_{0.25}\}}$ $\Delta H^0_{\{Si_{0.75}Ge_{0.25}\} \text{ in } \{Al\}}$	$\Theta_{Si_{0.75}Ge_{0.25}}$	
44107 $(J\,mol^{-1})$	- 3143 $(J\,mol^{-1})$	47250 $(J\,mol^{-1})$	21.92 $(J\,mol^{-1}K^{-1})$	-11304 $(J\,mol^{-1})$	557.25 K	
$\Delta H^0_{\langle Al \rangle \text{ in } \langle Si_{0.50}Ge_{0.50} \rangle}$	$\Delta H^0_{\langle Si_{0.5}0Ge_{0.50} \rangle \text{ in } \langle Al \rangle}$	$\Delta H^0_{Si_{0.50}Ge_{0.50}(diam) \to Si_{0.50}Ge_{0.50}(fcc)}$	$\Delta S_{Si_{0.50}Ge_{0.50}(diam) \to Si_{0.50}Ge_{0.50}(fcc)}$	$\Delta H^0_{\{Al\} \text{ in } \{Si_{0.50}Ge_{0.50}\}}$ $\Delta H^0_{\{Si_{0.50}Ge_{0.50}\} \text{ in } \{Al\}}$	$\Theta_{Si_{0.50}Ge_{0.50}}$	
40357 $(J\,mol^{-1})$	- 3143 $(J\,mol^{-1})$	43500 $(J\,mol^{-1})$	22.05 $(J\,mol^{-1}K^{-1})$	-11304 $(J\,mol^{-1})$	509.5 K	
$\Delta H^0_{\langle Al \rangle \text{ in } \langle Si_{0.25}Ge_{0.75} \rangle}$	$\Delta H^0_{\langle Si_{0.25}Ge_{0.75} \rangle \text{ in } \langle Al \rangle}$	$\Delta H^0_{Si_{0.25}Ge_{0.75}(diam) \to Si_{0.25}Ge_{0.75}(fcc)}$	$\Delta S_{Si_{0.25}Ge_{0.75}(diam) \to Si_{0.25}Ge_{0.75}(fcc)}$	$\Delta H^0_{\{Al\} \text{ in } \{Si_{0.25}Ge_{0.75}\}}$ $\Delta H^0_{\{Si_{0.25}Ge_{0.75}\} \text{ in } \{Al\}}$	$\Theta_{Si_{0.25}Ge_{0.75}}$	
36607 $(J\,mol^{-1})$	- 3143 $(J\,mol^{-1})$	39750 $(J\,mol^{-1})$	22.18 $(J\,mol^{-1}K^{-1})$	-11304 $(J\,mol^{-1})$	441.75 K	
$\Delta H^0_{\langle Al \rangle \text{ in } \langle Ge \rangle}$	$\Delta H^0_{\langle Ge \rangle \text{ in } \langle Al \rangle}$	$\Delta H^0_{Ge(diam) \to Ge(fcc)}$	$\Delta S_{Ge(diam) \to Ge(fcc)}$	$\Delta H^0_{\{Al\} \text{ in } \{Ge\}}$ $\Delta H^0_{\{Ge\} \text{ in } \{Al\}}$	Θ_{Ge}	Θ_{Al}
32857 $(J\,mol^{-1})$	- 3143 $(J\,mol^{-1})$	36000 $(J\,mol^{-1})$	22.3 $(J\,mol^{-1}K^{-1})$	-11304 $(J\,mol^{-1})$	374 K	428 K

The thermodynamic formulas used in this study[1]:

1. **Crystallization energy**

$$\Delta G^{Crystal}_{<Si_{1-x}Ge_x>-\{Si_{1-x}Ge_x\}}(T) = \Delta H^{Crystal}_{<Si_{1-x}Ge_x>-\{Si_{1-x}Ge_x\}}(T) - T\bullet\Delta S^{Crystal}_{<Si_{1-x}Ge_x>-\{Si_{1-x}Ge_x\}}(T)$$

$$\Delta H^{Crystal}_{\langle Si_{1-x}Ge_x\rangle-\{Si_{1-x}Ge_x\}}(T) = \Delta H^{Crystal}_{\langle Si_{1-x}Ge_x\rangle-\{Si_{1-x}Ge_x\}}(T_c) + \int_{T_c}^{T}\Delta C_p(T)dT$$

$$\Delta S^{Crystal}_{\langle Si_{1-x}Ge_x\rangle-\{Si_{1-x}Ge_x\}}(T) = -S^{0}_{\{Si_{1-x}Ge_x\}} + \int_{0}^{T}\frac{\Delta C_p}{T}dT$$

where, $\Delta H^{Crystal}_{\langle Si_{1-x}Ge_x\rangle-\{Si_{1-x}Ge_x\}}$ and $\Delta S^{Crystal}_{\langle Si_{1-x}Ge_x\rangle-\{Si_{1-x}Ge_x\}}$ are the crystallization enthalpy and crystallization entropy of $Si_{1-x}Ge_x$, respectively; the symbol of < > means crystalline phase; the symbol of { } represents amorphous phase; Tc stands for the crystallization temperature. ΔCp(T) is the specific heat difference between the crystalline and amorphous phases; S^0 are the residual entropies of amorphous phase at 0K.

2. **Surface energy**

$$\gamma^{S}_{\langle Si_{1-x}Ge_x\rangle} = \frac{\left(\gamma^{S}_{\langle Si_{1-x}Ge_x\rangle}(T_0)\times 0.35\times C_0\times V^{2/3}_{\langle Si_{1-x}Ge_x\rangle} + T_0\times S^{S}_{\langle Si_{1-x}Ge_x\rangle}\right) - T\times S^{S}_{\langle Si_{1-x}Ge_x\rangle}}{0.35\times C_0 V^{2/3}_{Si_{1-x}Ge_x}}$$

$$\gamma^{S}_{\{Si_{1-x}Ge_x\}} = \frac{\left(\gamma^{S}_{\{Si_{1-x}Ge_x\}}(T_0)\times 0.33\times C_0\times V^{2/3}_{\{Si_{1-x}Ge_x\}} + T_0\times S^{S}_{\{Si_{1-x}Ge_x\}}\right) - T\times S^{S}_{\{Si_{1-x}Ge_x\}}}{0.33\times C_0 V^{2/3}_{\{Si_{1-x}Ge_x\}}}$$

Where the symbol of < > means crystalline phase; the symbol of { } represents amorphous phase; $\gamma^{S}_{\langle Si_{1-x}Ge_x\rangle}(T_0)$ and $\gamma^{S}_{\{Si_{1-x}Ge_x\}}(T_0)$ acquired from experiment data; C_0 is a constant relating the surface area to the corresponding bulk volume, with an average value of 4.5×10^8 $mol^{-1/3}$; $V^{2/3}_{\langle Si_{1-x}Ge_x\rangle}$ and $V^{2/3}_{\{Si_{1-x}Ge_x\}}$ stand for the molar volumes;;The surface entropies $S^{S}_{\langle Si_{1-x}Ge_x\rangle}$and $S^{S}_{\{Si_{1-x}Ge_x\}}$ are approximately equal to 7.72 and 7.34 J $mol^{-1}K^{-1}$.

3. **Interface energy**

   a. Two crystalline phase:

$$\gamma^{interface}_{<A>/<B>} = \frac{G^{interface}_{<A>/<B>}}{0.35 \times C_0 \times \bar{V}^{2/3}_{<A>/<B>}}$$

b. One crystalline phase and one amorphous phase:

$$\gamma^{interface}_{<A>/\{B\}} = \frac{G^{intf}_{<A>/\{B\}}}{0.33 \times C_0 \times \bar{V}^{2/3}_{<A>/\{B\}}}$$

Where, < > stands for the crystalline phase; { } stands for the amorphous phase; A and B could be $Si_{1-x}Ge_x$, Al, crystalline Si; $G^{interface}_{<A>/<B>}$ and $G^{interface}_{<A>/\{B\}}$ mean interfacial Gibbs free energies between two phases; C_0 is a constant relating the surface area to the corresponding bulk volume, with an average value of 4.5×10^8 $mol^{-1/3}$; $\bar{V}^{2/3}_{<A>/<B>}$ or $\bar{V}^{2/3}_{<A>/\{B\}}$ stand for the average molar volume of A and B.

4. Critical thickness

Position 1:

$$h^{critical}_{\langle Al\rangle GBs} = \frac{2 \times (\gamma^{interface}_{\langle Al\rangle/\langle Si_{1-x}Ge_x\rangle} - \gamma^{interface}_{\langle Al\rangle/\{Si_{1-x}Ge_x\}})}{\Delta G^{Crystal}_{\langle Si_{1-x}Ge_x\rangle - \{Si_{1-x}Ge_x\}}}$$

Position 2:

$$h^{critical}_{\langle Al\rangle/\{Si_{1-x}Ge_x\}} = \frac{\gamma^{interface}_{\langle Al\rangle/\langle Si_{1-x}Ge_x\rangle} + \gamma^{interface}_{\langle Si_{1-x}Ge_x\rangle/\{Si_{1-x}Ge_x\}} - \gamma^{interface}_{\langle Al\rangle/\{Si_{1-x}Ge_x\}}}{\Delta G^{Crystal}_{\langle Si_{1-x}Ge_x\rangle - \{Si_{1-x}Ge_x\}}}$$

Position 3:

$$h^{critical}_{\langle Al\rangle/\langle sc-Si\rangle} = \frac{\gamma^{interface}_{\langle Al\rangle/\langle Si_{1-x}Ge_x\rangle} - \gamma^{interface}_{\langle Al\rangle/\{Si_{1-x}Ge_x\}} - \gamma^{interface}_{\langle sc-Si\rangle/\{Si_{1-x}Ge_x\}}}{\Delta G^{Crystal}_{\langle Si_{1-x}Ge_x\rangle - \{Si_{1-x}Ge_x\}}}$$

Reference:

1. Z. M. Wang, J. Y. Wang, L. P. H. Jeurgens and E. J. Mittemeijer, *Physical Review B*, 2008, **77**, 045424.
2. T.-W. Zhang, F. Ma, W.-L. Zhang, D.-Y. Ma, K.-W. Xu and P. K. Chu, *Applied*

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