Supplementary Information

Endotaxial Growth of Fe\textsubscript{x}Ge Single-Crystals on Ge(001) Substrates

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Additional experiments and discussions

\textbf{Figure S1.} SEM images of deposited Fe\textsubscript{x}Ge islands on Ge (001) substrate at various temperatures.
**Figure S2.** AFM of deposited Fe₃Ge islands on Ge (001) substrate at various temperatures.

**Figure S3.** Simulated diffraction pattern from three zone axes of monoclinic FeGe.
**Figure S4.** Standard XRD pattern of Fe$_{1.5}$Ge, Fe$_{1.67}$Ge and FeGe.
Figure S5. FFT of the monoclinic FeGe / Ge interface generated from HRTEM in Figure 3(a).
Figure S6. Schematic drawing of growth orientation of Fe$_3$Ge$_2$ island on Ge (001) substrate. (a) Top view of the substrate showing two major growth directions of Fe$_{13}$Ge$_8$ island, along Ge $<110>$, perpendicular to each other. The four interface facets of Fe$_3$Ge$_2$ are (002), (201), (2-2-1) and (02-1) respectively; (b) side view of the FeGe island after FIB cut, displaying the interface relationship between Fe$_3$Ge$_2$ and Ge (111); (c) 3D illustration of the island and substrate endotaxial relationship.
Figure S7. Schematic drawing of growth orientation of Fe$_{13}$Ge$_{8}$ island on Ge (001) substrate. (a) Top view of the substrate showing two major growth directions of Fe$_{13}$Ge$_{8}$ island, along Ge $<110>$, perpendicular to each other. Red top facet is Fe$_{13}$Ge$_{8}$ (0-42), the four interface facets of Fe$_{13}$Ge$_{8}$ are (4-41), (002), (-401) and (041) respectively; (b) side view of the FeGe island after FIB cut, displaying the interface relationship between Fe$_{13}$Ge$_{8}$ and Ge (111); (c) 3D illustration of the island and substrate endotaxial relationship.
Figure S8. Similar electron diffraction pattern of monoclinic and hexagonal phases. SAED of (a) monoclinic FeGe, (b) hexagonal Fe$_3$Ge$_2$; Simulated reciprocal lattice of (c) monoclinic FeGe, (d) hexagonal Fe$_3$Ge$_2$. 
Figure S9. Computer simulated atomic arrangement of interface planes of cubic Ge and hexagonal Fe$_3$Ge$_2$ respectively. (a, b) Ge (111) plane, while (c-f) Fe$_3$Ge$_2$ (02-1), (201) (2-2-1) and (002) respectively.
Figure S10. Computer simulated atomic arrangement of interface planes of cubic Ge and hexagonal \( \text{Fe}_{13}\text{Ge}_8 \) respectively. (a, b) Ge (111) plane, while (c-f) \( \text{Fe}_{13}\text{Ge}_8 \) (-401), (002) (-441) and (041) respectively.
Figure S11. The magnetization curves of Fe$_x$Ge at 5 K for samples deposited at (a) 500°C and (c) 550°C. The zero-field-cooled (ZFC) and field-cooled (FC) magnetizations at 100 Oe from 5 to 300 K for samples at (b) 500°C and (d) 550°C.