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Supplemental Material: Multi-Valley Electron Conduction at the Indirect-Direct Crossover Point in Highly-Tensile-Strained Germanium

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I. ϵ -Ge/ $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ Heterostructure Surface Morphology

Surface morphology characterization *via* atomic force microscopy (AFM) provides an additional means of investigating the strain relaxation dynamics of metamorphic heterostructures. In particular, the strain relief mechanisms that govern film relaxation during the growth of mismatched (001) oriented cubic materials are known to result in cross-hatch surface patterns wherein surface topography can be directly correlated with the nature and degree of strain relaxation within the film(s) [47,48]. This cross-hatch morphology originates from plastic relaxation processes as film thickness exceeds the critical thickness for the release of accumulated strain energy. Within this regime, initial misfit stress is relaxed by threading dislocation (TD) formation and glide, thereby creating slip steps at the film surface and misfit dislocations (MDs) at the film/substrate interface. For (001) oriented cubic systems, the most common slip system is $a/2 \langle 110 \rangle \{111\}$, indicating that the TDs and associated MDs that form occur within $\{111\}$ planes ($\alpha \approx 54.7^\circ$) and propagate laterally along $\langle 110 \rangle$ directions [49]. The step formation that occurs at the growth front is subsequently smoothed *via* lateral mass transport and preferential step-site growth, *i.e.*, in a typical step-flow growth process. Successive relaxation results in the formation of hillocks and valleys oriented along dislocation lines and in the vicinity of buried MDs. Hence, any anisotropy in relaxation along orthogonal $\langle 110 \rangle$ directions can be directly evidenced by an asymmetry in the ultimate cross-hatch topography. In the case of lattice-mismatched $\text{In}_x\text{Ga}_{1-x}\text{As}$ on GaAs epitaxy, an additional source of anisotropic strain relaxation is due to the disparity between α (As-terminated core) and β (Ga-terminated core) dislocation glide velocities oriented along the $[1\bar{1}0]$ and $[110]$ directions, respectively [49]. Thus, for a metamorphic $\text{In}_x\text{Ga}_{1-x}\text{As}$ film with isotropic strain relaxation, it would be expected that the surface exhibit a symmetric cross-hatch pattern with peak-to-valley amplitude and uniformity dependent on film thickness and the degree of relaxation [47–49]. A minor degree of asymmetry between the $[1\bar{1}0]$

and $[110]$ oriented surface features could arise due to the associated difference in dislocation glide velocities; however, a large asymmetry in the cross-hatch morphology would suggest a strong degree of anisotropic strain relaxation and the presence of epitaxial tilt or similar defect-related film non-idealities.

As can be seen from Fig. S1, a representative $20 \mu\text{m} \times 20 \mu\text{m}$ AFM micrograph of the ϵ -Ge/ $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ heterostructure's surface reveals a distinct, symmetric cross-hatch with a peak-to-valley amplitude of $\sim 9 \text{ nm}$ and RMS roughness (R_q) of 3.35 nm . The degree of uniformity between the orthogonal $[1\bar{1}0]$ and $[110]$ directions is indicative of a largely isotropic film relaxation. Nevertheless, the $[1\bar{1}0]$ oriented surface features exhibit a qualitatively coarser topography. Quantitative analysis of

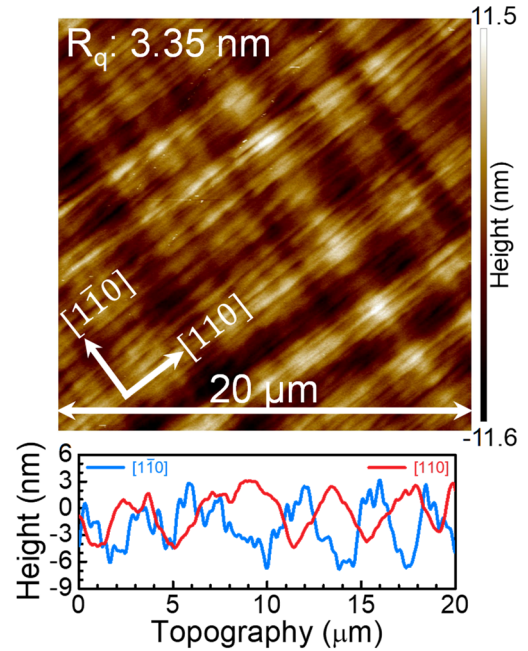


FIG. S1. Atomic force micrograph of a representative $20 \mu\text{m} \times 20 \mu\text{m}$ region of the as-grown ϵ -Ge/ $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ surface and associated line height profiles taken along the two orthogonal $\langle 110 \rangle$ symmetric directions.

line height profiles taken along the $[1\bar{1}0]$ and $[110]$ directions yield different R_q values, *i.e.*, 3.28 nm and 2.24 nm, respectively, suggesting a greater degree of relaxation in the $[110]$ direction. This is reinforced by the comparatively larger separation between hillocks and valleys in the $[110]$ line profile, which has been previously correlated to a high degree of relaxation in thick ($\sim 1 \mu\text{m}$) $\text{In}_{0.25}\text{Ga}_{0.75}\text{As}$ films grown on GaAs [48]. Moreover, the slight asymmetry observed in our AFM findings corresponds well to the observed tilt (~ 540 arcsec) measured *via* XRD, indicating a small but measurable degree of anisotropy in buffer strain relaxation during growth. We attribute this anisotropy to the previously discussed difference in dislocation velocities for α ($[1\bar{1}0]$ oriented) and β ($[110]$ oriented) dislocations.

II. Supplementary Methods 1: Raman Spectroscopy

For (001) oriented diamond-cubic crystals, the effect of biaxial strain is to split the threefold degenerate zone-center optical phonon modes into a doublet and singlet having eigenvectors perpendicular and parallel to the plane, respectively [51,52]. Thus, following the selection rules provided in Ref. [51,52], only the longitudinal optical (LO) mode corresponding to the singlet is observable under the (001) backscattering geometry utilized in this work. Moreover, the incorporation of lattice strain results in a hydrostatic shift of the phonon frequency (Ω_s) and thus a relative shift in the measured Raman wavenumber ($\Delta\omega$) with respect to its unstrained value (ω_0). The magnitude and sign of the resultant wavenumber shift is indicative of the material's strain-state and type of strain present within the system, *i.e.*, a positive or negative $\Delta\omega$ corresponds to compressive or tensile strain, respectively. In our previous work [53], we have used the relation $\Delta\omega = -b\varepsilon_{\parallel} \text{ cm}^{-1}$ to analyze the Raman shift as a function of strain ($\varepsilon_{\parallel}$) for moderately strained $\varepsilon\text{-Ge}$ epilayers grown on $\text{In}_x\text{Ga}_{1-x}\text{As}$ metamorphic stressors heterogeneously integrated onto Si substrates. In the preceding expression, $\Delta\omega$ is the measured wavenumber shift (in cm^{-1}) whereas b can be expressed as [51,52]:

$$b = \omega_0 \left(\tilde{K}_{11} \frac{C_{12}}{C_{11}} - \tilde{K}_{12} \right), \quad (\text{S1})$$

where $\tilde{K}_{11}$ and $\tilde{K}_{12}$ are the phonon deformation potentials normalized to ω_0^2 , ω_0 is the unstrained phonon frequency ($\sim 300 \text{ cm}^{-1}$ for Ge), and C_{11} and C_{12} are material elastic constants. We note that the expression for $\Delta\omega$ assumes an isotropic, tetragonal strain of the form:

$$\varepsilon = \begin{pmatrix} \varepsilon_{\parallel} & 0 & 0 \\ 0 & \varepsilon_{\parallel} & 0 \\ 0 & 0 & \varepsilon_{\perp} \end{pmatrix}, \quad (\text{S2})$$

in order to simplify the phonon secular equation and derive the expression for the strain-modified phonon frequency and hence the relative shift, $\Delta\omega$, in phonon frequency as a function of strain. Using the above expression and the

material parameters found in Ref. [51,54], b was calculated to be -415 cm^{-1} [34,55].

III. Supplementary Methods 2: TEM Dislocation Identification

As previously discussed with respect to the investigated AFM surface morphology, dislocation formation occurs during the heteroepitaxy of metamorphic thin films in order to relieve accumulated lattice mismatch-induced film stress. Due to the packing density of $\{111\}$ atomic planes in (001)-oriented diamond cubic and zinc-blende crystals, such planes preferentially exhibit lattice distortions due to relaxation processes during growth [57]. The nucleation of dislocation half-loops at the film surface and their subsequent glide, both toward the lattice mismatched heterointerface (following $\{111\}$ slip planes) and the substrate edge (along $\langle 110 \rangle$ directions) [49], is known to result in several dislocation structures, including, but not limited to: (i) threading dislocations (TDs) propagating along the growth direction; (ii) misfit dislocations (MDs) observed at the lattice-mismatched heterointerface; and (iii), stacking faults (SFs), originating from the dissociation of perfect 60° MDs into 90° and 30° Shockley partial dislocations [56–59]. These dislocations represent localized lattice disorder, and as such, also introduce local strain gradients surrounding the dislocation core, which can be utilized in practice in order to identify, *e.g.*, TDs at a film's surface. Figure S2(a) presents a representative plan-view TEM (PV-TEM) micrograph of the $\varepsilon\text{-Ge}/\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ heterostructure, wherein the top $\sim 30 \text{ nm}$ of $\varepsilon\text{-Ge}$ has been etched in order to expose the underlying TDs propagating through the $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ stressor. The diffraction contrast induced by the interaction between the diffracted electrons and the strain gradient surrounding the TDs is evident, as highlighted in Fig. S2(a). Using the diffraction contrast in order to identify TDs at the $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ surface (*i.e.*, the $\varepsilon\text{-Ge}/\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ interface of the as-grown heterostructure), a conservative estimate for the TD areal density, ρ_{TD} , was found to be $\leq 2 \times 10^7 \text{ cm}^{-2}$.

Similarly, MDs in the vicinity of the lattice-mismatched heterointerface are identifiable as disruptions to the otherwise ordered atomic planes when examined under cross-section using high magnification. Fig. S2(b) presents the origin of such MDs for the $\{111\}$ slip system, wherein $\mathbf{u}$ represents the dislocation line, $\boldsymbol{\tau}$ the resolved shear stress on the planes, and $\mathbf{b}$, $\mathbf{b}_1$, and $\mathbf{b}_2$ are the Burgers vectors for the 60° perfect, 90° partial, and 30° partial dislocations, respectively [56]. For tensile interfaces, it is energetically favorable for the 60° perfect dislocation to dissociate, first nucleating the 90° partial dislocation and giving rise to a stacking fault [56,58,59]. Assuming a dislocation line parallel to the $[1\bar{1}0]$ direction, the Burgers vector for the 90° and 30° partial dislocations could be given by $a/6$ [112] and $a/6$ [211], respectively. Should dislocation glide continue,

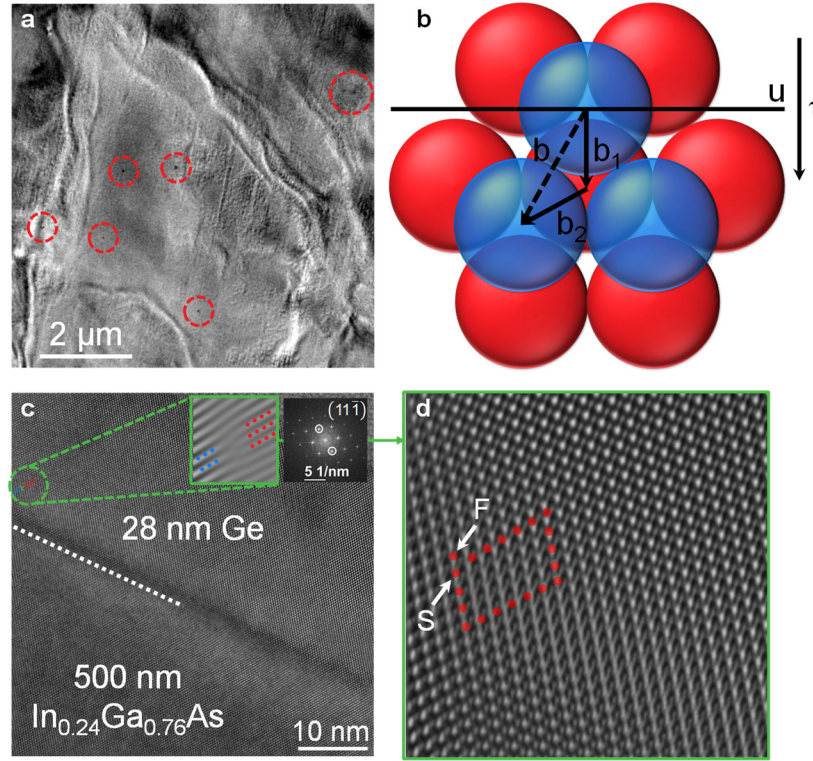


FIG. S2. (a) Plan-view TEM micrograph of the ϵ -Ge/ $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ heterostructure wherein the top 30 nm ϵ -Ge epilayer has been etched in order to identify threading dislocations in the underlying $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ stressor. (b) Schematic representation of the Burgers vectors for leading 90° partial, trailing 30° partial, and perfect 60° dislocations – b , b_2 , and b_1 , respectively – originating in $\{111\}$ planes within a semiconductor having a diamond cubic (or zinc-blende) crystal structure. Adapted from Petruzzello *et al.* [56]. (c) Representative misfit dislocation in the ϵ -Ge epilayer, identified as originating in the $(11\bar{1})$ plane, and (d) its associated Burgers circuit.

the 30° partial dislocation will subsequently nucleate, reacting with the 90° partial dislocation following [57]:

$$\frac{a}{6}[112] + \frac{a}{6}[211] \rightarrow \frac{a}{2}[101]. \quad (\text{S3})$$

The result of this reaction is the annihilation of the stacking fault and the formation of a perfect 60° dislocation. Thus, the type of lattice disorder introduced *via* MD formation can be used to identify the corresponding MD. Figs. S2(c) and S2(d) present a representative high-magnification cross-sectional TEM micrograph, from which the $(11\bar{1})$ plane was used (Fig. S2(c), insets) to identify MDs in the vicinity of the ϵ -Ge/ $\text{In}_{0.24}\text{Ga}_{0.76}\text{As}$ heterointerface, and the corresponding

Burgers circuit tracing the lattice distortion surrounding a representative MD, respectively. As can be seen from Fig. S2(c), the MD was associated with the insertion of an additional atomic plane (*i.e.*, an edge dislocation having a dislocation line within the plane of the page). Moreover, examination of the Burgers circuit encompassing the MD (Fig. S2(d)) reveals SF formation, indicating the nucleation of a leading 90° partial dislocation having a projected Burgers vector (S to F) of $a/6\langle 112 \rangle$. Additionally, the persistence of the SF indicates a lack of trailing 30° partial dislocation formation following nucleation of the leading 90° partial dislocation, thereby preventing dislocation reaction and annihilation of the SF according to Eq. (S3).