Supplementary Materials for

**Layer-resolved release of epitaxial layers in III-V heterostructure via a buffer-free mechanical separation technique**

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Controlled spalling technique for layer release of single-crystalline semiconductors

Fig. S1 shows the thickness-controlled spalling technique for layer release of single-crystalline elemental semiconductors (Si) and zinc-blende compound semiconductors (GaAs and InP). In this process, a thin semiconductor layer can be mechanically released by depositing a stressor film such as Ni which has a high tensile stress (29), as shown in Fig. S1A and S1B. The crack initiation and propagation for layer release are facilitated by the strong misfit stress induced in the Ni film and substrate. The crack trajectory and mechanism of the spalling process have been explained by a fracture model developed by Suo and Hutchinson (54, 55); this model defines two major stress intensity factors: $K_I$ (mode I, opening mode) for the vertical direction and $K_{II}$ (mode II, in-plane shear mode) for the lateral direction. First, the crack is initiated near the substrate edge by an external force and propagates to the inclined direction due to the mixed-mode stresses of $K_I$ and $K_{II}$. Then, the crack seeks a specific depth at which $K_{II} = 0$ and propagates straight along the main crystal plane parallel to the film/substrate interface. In accordance with the Suo and Hutchinson model, a thin layer of Si(100) could be released with a flat surface and uniform layer thickness, as observed in the SEM images of Fig. S1A. Previous studies have shown that the spalling process enables layer release of Si and Ge with an atomically flat surface (root-mean-squared surface roughness < 5 Å) (11, 56). However, it has been reported that the spalling of zinc-blende III-V(100) compound semiconductors such as GaAs and InP cannot offer a flat surface because cracks do not straightly propagate along the (100) crystal plane (21, 25–28). Fig. S1C shows a schematic of the III-V(100) spalling process and cross-sectional SEM images of the released GaAs(100) layer and the remaining substrate. The thickness of the released layer is not uniform and periodic zig-zag corrulations are observed, unlike the result of Si spalling. To investigate the dependence of the thickness of the released layers upon Ni thickness, we released the Si(100) and III-V(100) layers using Ni films with varying thicknesses and measured the thickness of the released layers (i.e., spalling depth) from cross-sectional SEM images of these layers. In the case of the III-V(100) layers, we estimated the spalling depth from the average thickness of the released layers because layer thickness was non-uniform because of zig-zag corrugations (Fig. S1C). Fig. S1D shows the thickness of the released Si(100), GaAs(100), and InP(100) layers as a function of the Ni thickness. The released-layer thickness was controlled in the range from 2 to 15 μm for Ni thicknesses from 2.5 to 6.5 μm. This result shows that the layer thickness can be controlled by adjusting the Ni thickness, and the non-linear relationship between the Ni thickness and the released-layer thickness agrees well with the previously reported empirical model for spalling depth (11).

In this result, the large thickness deviations observed in the relatively thick layers were possibly attributed to the locally non-uniform Ni thickness and film stress resulted from the harsh sputtering environment required for the thick Ni deposition because the local variation of Ni thickness and stress causes the variation of spalling depth where $K_{II} = 0$. This indicates that the thickness uniformity of the Ni stressor film is a critical factor for layer separation with uniform thickness from a semiconductor bulk. Thus, the development of a sputtering or electroplating process that guarantees the thickness and stress uniformity of thick Ni films is essential for reliable spalling process of bulk semiconductors.

Supplementary Text
To investigate the effects of surface corrugation on a subsequent transfer process, we released the 3-µm-thick GaAs layer from bulk GaAs(100) using the spallating process and transferred the layer onto a Si substrate. Then, we characterized the quality of the transferred GaAs layer based on SEM, XRD, and PL measurements. Fig. S2A shows the cross-sectional SEM images of the GaAs layer transferred onto the Si substrate. Because of the zig-zag corrugations on the surface of the released layer, undesirable air voids were formed along the interface between the GaAs layer and the SiO$_x$ bonding layer. These voids hinder the reliable transfer process because adhesion between bonded layers is not uniform and air voids can expand or contract under temperature variation. Moreover, air voids can degrade the heat-dissipation capability and local-temperature uniformity of the transferred layer due to the low thermal conductivity of air. Although zig-zag corrugations impede the transfer process, the XRD and PL measurement results show that the crystal and material quality of the GaAs layer were maintained during the layer release/transfer processes [Fig. S2B and S2C].

To investigate the difference in binding energy of a heteroepitaxial interface and bulk crystal planes, we conducted DFT calculations to estimate the crystal binding energy of a heteroepitaxial interface and compared the result with those of main crystal planes in the III-V bulk. The DFT calculations were performed using the plane-wave pseudopotential code as implemented in Quantum Espresso (57), and the pseudopotential was described by the projector augmented wave (PAW) method (58) and Generalized Gradient Approximation (GGA) function (59). The kinetic-energy cutoff of plane waves was set to 450 eV and Γ-centered Monkhorst–Pack grids of 1 × 1 × 1 k-points were utilized for the Brillouin-zone integration. The force and energy convergence criterion were set to 0.02 eV Å$^{-1}$ and 10$^{-5}$ eV, respectively.

In these calculations, we built three different atomic structures to estimate the crystal binding energy of the epitaxial interface between In$_{0.53}$Ga$_{0.47}$As(100) and InP(100) layers and those of (100) and (110) planes in the InP bulk. The atomic structures used for the calculation are illustrated in Fig. S3A. The binding energy was calculated by $E_b = E_{tot} - E_{slab1} - E_{slab2}$ (60), where $E_{tot}$ is the total energy of the atomic structure system, and $E_{slab1}$ and $E_{slab2}$ are the total energy of one half of the structure without considering the other half. For instance, as shown in the atomic structure of InP(100)/In$_{0.53}$Ga$_{0.47}$As(100) in Fig. S3A, $E_{slab1}$ and $E_{slab2}$ represent the total energy of In$_{0.53}$Ga$_{0.47}$As(100) slab and InP(100) slab, respectively. The estimated binding energy of the (100) interface between InGaAs and InP was −44.78 eV, and those of the (100) and (110) planes in the InP bulk were −472.05 and −165.94 eV, respectively. The DFT calculation results indicate that (i) the crystal binding energy of the bulk (100) plane is larger than that of the bulk (110) plane, as expected and (ii) the interfacial bonding between the InP(100)/InGaAs(100) layers is weaker than the crystal bonding of the (100) and (110) planes in the InP bulk. This result implies that a heteroepitaxial interface between InP and InGaAs layers can become more preferable to the bulk crystal planes for the layer release in spalling processes. In addition, it is notable that the typical formation of lattice-mismatched one or few monolayers at an epitaxial interface were not
accounted for in this calculation. If the effect of the lattice-mismatched monolayers at the interface are introduced, we think that the difference in binding energies of a heteroepitaxial interface and bulk crystal planes can be even larger because of the localized misfit strain energy induced by the lattice-mismatched intermixing layers.

Furthermore, to estimate whether the layer-release process at an epitaxial interface can also be applied to other III-V heterostructures, we investigated the binding-energy contrast in GaAs/In_{0.49}Ga_{0.51}P heterostructure (one of the representative lattice-matched systems) based on the additional DFT calculation. The atomic structures used for the calculation are illustrated in Fig. S3B. The crystal binding energy was calculated by the same method (\(\text{i.e., } E_{\text{tot}} - E_{\text{slab1}} - E_{\text{slab2}}\)) used for the InP/InGaAs heterostructure. The estimated binding energy of the (100) interface between In_{0.49}Ga_{0.51}P and GaAs was \(-51.61\) eV, and those of the (100) and (110) planes in the GaAs bulk were \(-419.88\) and \(-130.43\) eV, respectively. This DFT calculation results also indicate that (i) the crystal binding energy of the bulk (100) plane is larger than that of the bulk (110) plane, and (ii) the interfacial bonding between the GaAs(100)/InGaP(100) layers is weaker than the crystal bonding of the (100) and (110) planes in the GaAs bulk. As with the result in the InP/InGaAs heterostructure (Fig. S3A), this result implies that a heteroepitaxial interface between GaAs and InGaP layers can become more preferable to the bulk crystal planes for the layer release in spalling processes. In addition to this binding energy contrast, it is also known an abruptly inverted heteroepitaxial interface between GaAs and InGaP layers can contain one or few monolayers of an undesirable lattice-mismatched III-V alloy which is induced by As–P phase intermixing (61, 62). Considering that two contributing factors (binding-energy contrast and intermixing layer) for layer separation at an epitaxial interface are also valid in the lattice-matched GaAs/In_{0.49}Ga_{0.51}P heterostructure, we think that the proposed technique can be extended to GaAs/InGaP heterostructures and probably generalized to a wide range of III-V heterostructures.

[Note S4] Estimation of the spalling depth using an analytical model

In this study, the spalling depth was calculated by an analytical model based on delamination theory (11, 45, 46). In this model, the spalling depth can be calculated based on the thermodynamic equilibrium condition of the film/substrate structure in which the total strain energy accumulated in the Ni film and the released layer is balanced with the crystal binding energy (11, 46, 47, 63). A detailed calculation procedure is described with the schematic in Fig. S4 indicating the magnitude and direction of the stresses induced in the film/substrate structure.

To calculate this analytical model, we first need to know the elastic strain of the film and the substrate, which can be calculated from the measured film stress. The relationship between elastic strain and stress is expressed as follows (45):

\[
\varepsilon = \frac{(1-v)}{Y} \sigma
\]

where \(\varepsilon\), \(\sigma\), \(Y\), and \(v\) are the elastic strain, biaxial stress, Young’s modulus, and Poisson ratio of a material, respectively. Then, the elastic strain energy accumulated in the stressor film (\(U_e\)) can be calculated as follows:
Because of the strong tensile stress of the stressor film, the compressive stress ($\sigma_{\text{compression}}$) and bending stress ($\sigma_{\text{bending}}$) are induced in the substrate (4S). The total stress induced in the substrate can be estimated based on the film stress ($\sigma_f$) as follows:

$$\sigma_s = \sigma_{\text{compression}} + \sigma_{\text{bending}} = -\left(\frac{t_f}{t_s} \sigma_f + \frac{6t_f \sigma_f}{t_s^2} y\right)$$  \hspace{1cm} (3)

The elastic strain energy in the substrate ($U_S$) accumulated from the substrate surface ($y = t_s/2$) to a specific release point ($y = y_s$) can be calculated as follows:

$$U_S = \frac{1}{2} \int_{t_s/2}^{t_f} \varepsilon_s \sigma_s dy = \frac{1}{2} \int_{t_s/2}^{t_f} \sigma_{\text{compression}} + \sigma_{\text{bending}}) dy = \frac{(1-v_s)}{2Y_s} \int_{t_s/2}^{t_s} \left(\frac{t_f}{t_s} \sigma_f + \frac{6t_f \sigma_f}{t_s^2} y\right)^2 \sigma_f dy$$

$$= \frac{(1-v_s)}{2Y_s} \frac{t_f^2}{t_s} \sigma_f^2 \left[\frac{7}{2} - 12 \left(\frac{Y_s}{Y_f}\right)^3 - 6 \left(\frac{Y_s}{Y_f}\right)^2 - \left(\frac{Y_s}{Y_f}\right)\right]$$  \hspace{1cm} (4)

In this model based on the thermodynamic equilibrium condition, the layer is released at the point where the strain energy accumulated in the film/substrate ($U_f + U_s$) is equal to the crystal binding energy of the substrate ($\gamma_s$) (11). Thus, we could calculate the spalling depth that satisfies the thermodynamic equilibrium condition ($\gamma_s = U_f + U_s$) for the layer release at $y_s$:

$$y_s = \frac{(1-v_f)}{2Y_f} t_f \sigma_f^2 + \frac{(1-v_s)}{2Y_s} \frac{t_f^2}{t_s} \sigma_f^2 \left[\frac{7}{2} - 12 \left(\frac{Y_s}{Y_f}\right)^3 - 6 \left(\frac{Y_s}{Y_f}\right)^2 - \left(\frac{Y_s}{Y_f}\right)\right]$$  \hspace{1cm} (5)

The positive real solution of this cubic equation corresponds to the release point ($y_s$). Therefore, if we know the physical parameters of materials ($Y_i$, $v_i$, $Y_s$, $v_s$, and $\gamma_s$), the film stress ($\sigma_f$), and the thickness of the film and the substrate ($t_f$ and $t_s$), the spalling depth can be simply calculated from $y_s$ (i.e., $d_{\text{spall}} = t_s/2 - y_s$).

[Note S5] Estimation of the layer-release yield of InP/InGaAs heterostructures

As shown in Fig. S7A, the zig-zag corrugations on the surface of the remaining substrate after the spalling process in the III-V(100) bulk can be observed in the optical microscopy (OM) image. In case the periodic stripes corresponding to the corrugations were not observed in the OM image of a spalled sample, that sample’s surface was flat when imaged by SEM; this indicates that layer release occurred at an epitaxial interface. Thus, we could estimate the layer-release yield of the InP/InGaAs heterostructure using OM images taken from the entire region of the remaining substrate after the layer-resolved mechanical separation process. The estimated yield is shown in Figs. S7B and S7C. After spalling process III, we took 441 OM images from the entire spalled region (sample size of 15 × 15 mm²). To evaluate the yield, we divided each OM image into 50 areas of 100 × 100 µm², counted the number of flat and defect-free areas and defined the yield of one region covered by the OM image by the percentage of counted areas. We repeated this process
for all OM images. The yield-mapping images of all regions corresponding to the 441 OM images are shown in Fig. S7B. Here, the color of each pixel indicates the yield of each region. Figure ‘a’ shows a representative OM and top-view SEM images for a high yield (> 95%) and Figure ‘b’ shows ones for a relatively low yield due to zig-zag corrugations or cracks. The histogram of the yield distribution is plotted in Fig. S7C. The overall layer-release yield estimated from this method was about 78%. To compare the quality of the layer-resolved III-V mechanical separation with that of conventional Si(100) spalling, we released a thin Si layer through the same process and evaluated the yield of Si spalling using the same method. The sample size of the Si layer was 22 × 22 mm². Fig. S7D shows the mapping image of the spalling yields estimated from 961 OM images and Fig. S7E shows the histogram plot of the yield distribution. The overall process yield for the entire region was about 80%, which is comparable with that of the III-V result. This implies that the process yield in this study was limited by the non-ideal manual process in a laboratory environment rather than by the process scheme of the proposed layer-resolved mechanical technique for III-V heterostructures. As suggested in several previous reports, we expect that the process yield can be further increased if the mechanical separation process is performed by using force-controlled spalling equipment. After crack initiation by an external force, the layer-release process relies on a quasi-spontaneous exfoliation (i.e., quasi-self-propagation), by which the crack propagates with a uniform layer thickness under the accumulated strain energy balanced with the crystal binding energy. Thus, after the crack initiation, an external force should be maintained with a uniform magnitude and sustained direction and an excess external force should be minimized. Furthermore, it is known that the layer-release yield can be further increased in a wafer-scale process because large-scale mechanical separation process requires a smaller external force for quasi-spontaneous exfoliation (17, 19, 21–23).
Fig. S1.

**Thickness-controlled spalling process for the layer release of single-crystalline elemental and zinc-blende III-V compound semiconductors.** (A) A schematic of the spalling process for Si(100) and cross-sectional SEM images of the released layer and remaining substrate, showing that the surfaces of the released Si layers are flat and uniform. The scale bar is 3 µm. (B) A schematic of the crack trajectory and the mechanism of the spalling process. Under strong misfit stress in the stressor film and substrate, a crack is initiated near the substrate edge with two major stress intensity factors ($K_I$ and $K_{II}$); it propagates straightly parallel to the film/substrate interface at a specific depth at which $K_{II} = 0$. (C) A schematic of the spalling process for III-V(100) semiconductors with a zinc-blende structure and cross-sectional SEM images of the released GaAs(100) layer and the remaining substrate. In contrast to Si, the surface of the released III-V layers is not flat; periodic zig-zag corrugations are observed. The scale bar is 5 µm. (D) The thickness of the released Si(100), GaAs(100), and InP(100) layers depends on the thickness of the Ni stressor film.
Fig. S2.
Characterizations of a GaAs layer released from bulk GaAs(100) and transferred onto a Si substrate. (A) Cross-sectional SEM image of the transferred GaAs(100) layer on a Si substrate after the spalling process. (B) Wide-range XRD $\omega$–$2\theta$ scans and (C) steady-state room temperature PL spectra of the bulk GaAs(100) wafer and the GaAs(100) layer after the layer release/transfer processes.
Fig. S3.

DFT calculation for estimation of crystal binding energies. (A) The atomic structures of InP(100)/In$_{0.53}$Ga$_{0.47}$As(100), bulk InP(100), bulk InP(110) and (B) those of GaAs(100)/In$_{0.49}$Ga$_{0.51}$P(100), bulk GaAs(100), bulk GaAs(110), which were used for the DFT calculations. The calculation results showed that the interfacial bonding between the heteroepitaxial layers is weaker than the crystal bonding of the (100) and (110) planes in the bulk.
Fig. S4.

**Schematic of the magnitude and direction of stresses induced in the film/substrate structure.**

Assuming that the stressor film is under tension, the film-induced stresses in the substrate are composed of the compressive stress and the bending stress (11, 43). The black-dashed line indicates a specific release point ($y = y_s$).
Fig. S5.
Cross-sectional SEM images of the remaining substrates with various magnifications after process I, II, and III. The third SEM image for each process is identical to the image shown in Fig. 2D of the main text. The color and brightness of the 200-nm-thick InGaAs layers in the images, except for the fourth images in process II and III, have been adjusted to make them more visible.
Fig. S6.

Original versions of the false-colored SEM images. (A) False-colored cross-sectional SEM images of the released layers and the remaining substrate after process II and III (shown in Fig. 2D) and (B) their original SEM images. The color and brightness of the relatively thin epitaxial layers (denoted as $a$, $b$, $c$, $d$, and $e$ regions) have been adjusted to make them more visible.
Fig. S7.

**Estimation of the layer-release yield at the InP/In$_{0.53}$Ga$_{0.47}$As interface.** (A) OM image of the surface of the remaining substrate after the layer release in the InP bulk and the cross-sectional SEM image of the substrate defined by the dashed box in the OM image. It can be confirmed that the periodic stripes in the OM image correspond to zig-zag corrugations. (B) The yield-mapping image for the layer-resolved mechanical separation (process III) and representative OM and top-view SEM images for a high yield (denoted by ‘a’, yield ~98%) and a relatively low yield (denoted by ‘b’, yield ~40%). The InP/InGaAs epitaxial layer was released from ‘b’ to ‘a’. (C) Histogram of the yield distribution for the layer-resolved mechanical separation. (D) The yield-mapping image for the conventional Si(100) spalling and representative OM and top-view SEM images for a high yield (denoted by ‘a’, yield ~98%) and a relatively low yield (denoted by ‘b’, yield ~40%). The Si layer was released from ‘b’ to ‘a’. (E) Histogram of the yield distribution for Si spalling. Scale bars in all OM and top-view SEM images are 100 µm.
Uniformity evaluation of the released points after process II. (A) The plot for the released points at 50 positions covering the entire region of the spalled sample. The released points were evaluated by measuring the thickness of the top epitaxial layer in the remaining substrate after obtaining the cross-sectional SEM images from the 50 positions. The released point means the thickness of the released layer estimated from the thickness of the top epitaxial layer in the remaining substrate. The sample size was $15 \times 15$ mm$^2$ and the spacing between the measured positions was approximately 0.3 mm. The release points corresponding to the heteroepitaxial interfaces are denoted by dashed lines among which the designed local symmetry plane for process II is highlighted by the red dashed line. (B) Representative cross-sectional SEM images of the remaining substrate at the positions denoted by circles in the plot (A).
**Fig. S9.**

**Uniformity evaluation of the released points after process III.** (A) The plot for the released points at 50 positions covering the entire region of the spalled sample. The released points were evaluated by measuring the thickness of the top epitaxial layer in the remaining substrate after obtaining the cross-sectional SEM images from the 50 positions. The released point means the thickness of the released layer estimated from the thickness of the top epitaxial layer in the remaining substrate. The sample size was $15 \times 15 \text{ mm}^2$ and the spacing between the measured positions was approximately 0.3 mm. The release points corresponding to the heteroepitaxial interfaces are denoted by dashed lines among which the designed local symmetry plane for process III is highlighted by the red dashed line. (B) Representative cross-sectional SEM images of the remaining substrate at the positions denoted by circles in the plot (A).
Fig. S10.

Confirmation of undesirable intermixing layers formed at the epitaxial interface of the lattice-matched InP/In_{0.53}Ga_{0.47}As heterostructure. (A) High-resolution TEM images at the heteroepitaxial interface between InP and In_{0.53}Ga_{0.47}As layers, showing the presence of an intermixing layer with a two- or three-monolayer thickness. (B) The Raman spectrum measured on the surface of the remaining substrate after process III (adopted from Fig. 3D of the main text), where non-negligible sub-peaks are observed in the range of 350–410 cm\(^{-1}\). (C) The Raman spectrum measured in the sub-peak region (350–410 cm\(^{-1}\)) denoted by the blue dashed box in (B). The spectrum with the GaP-like LO- and TO-mode peaks (64) indicates that the epitaxial interface contains an InGaP-like undesirable intermixing layer induced by phase intermixing of As–P atoms. In the proposed layer separation technique, this intermixing layer can facilitate the layer release at the epitaxial interfaces, and further, the few-monolayer thickness of the intermixing layer can suppress the probability of the crack propagation taking the detour path to the bulk crystal planes, leading to make the straight crack propagation at the epitaxial interface more stable in the layer-release process.
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