Supporting Information
Mechanically transferred large-area Ga$_2$O$_3$ passivates graphene and suppresses interfacial phonon scattering

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S1. Device Fabrication

Graphene/h-BN/SiO$_2$/Si heterostructures were commercially purchased from Graphene Supermarket, consisting of 1 cm square monolayer graphene on monolayer h-BN, both wet transferred onto 285 nm SiO$_2$ on 525 µm Si (100) p-doped substrates. This heterostructure was etched into Hall bars using an argon-oxygen plasma, via standard photolithographic techniques and a PVA TePla Plasma Asher.

S2. Mechanical transfer of Ga$_2$O$_3$

We started with a PPC mask prepared by spin coating PPC (Polypropylene carbonate) onto Gel-Film from Gel-Pak at 3000 rpm, and baked at 100 °C. Gallium was heated to liquid phase using a hotplate at 40 °C in ambient. A drop of gallium was then placed on the PPC mask at 40 °C. The liquid metal was then “rolled” by flattening a glass coverslip on the surface, depositing the Ga$_2$O$_3$ sheets on the PPC. Followed by this, the polymer stack consisting of Gel-film, PPC, and Ga$_2$O$_3$ was cut for sizing and to remove excess PPC & gallium metal residues using a hot scalpel. Finally, PPC & Ga$_2$O$_3$ stack was transferred onto the device using a remote-controlled micro-mechanical transfer apparatus with a microscope to align to device geometry. Transfer occurred at temperatures up to 120 °C. PPC was removed by dissolving it in acetone. A schematic of this process is illustrated at Fig. S1 below.
FIG. S1: Transfer process for Ga$_2$O$_3$.
(a) Gel-Film on glass slide. (b) PPC spin-coated onto gel film. (c-d) Gallium metal rolled over PPC to deposit Ga$_2$O$_3$. (e) Gel-Film/PPC/Ga$_2$O$_3$ stack cut using high temperature razor. (f) Gel-Film/PPC/Ga$_2$O$_3$ stack flipped to transfer onto SiO$_2$/Graphene device using a transfer stage with a microscope. (g) PPC/Ga$_2$O$_3$ deposited by heating the sample to 120°C. (h) PPC washed in Acetone/IPA leaving clean.
S3. Atomic Layer Deposition (ALD) of Al₂O₃

Al₂O₃ ALD was performed following the plasma-enhanced methodology of Tang et al., using a Fiji F200 ALD instrument from Ultratech/Cambridge NanoTech Inc. The metallic precursors were released first, prior to oxidant precursor created through plasma exposure, to minimize plasma damage to the graphene surface in the first cycle. The ALD temperature differed from Tang et al., being performed at 150 °C instead of 250 °C, in order to keep annealing conditions consistent for Ga₂O₃. 55 cycles were applied, leading to a growth of 5.5 nm thick Al₂O₃-film as confirmed on Si reference samples by ellipsometry.

S4. Electrical Measurements in PPMS & Probestation

Samples were wirebonded to chip carriers and measured using a Quantum Design PPMS 6000, operating between 4 K and 400 K under vacuum (< 10 mTorr) in presence of He exchange gas. Samples were annealed first for 8 hours at 390 K in vacuum.

ALD-processed samples were measured in a Lakeshore TTPX probestation at room temperature, with vacuum ≈ 10⁻⁴ mBar, both before and after ALD. Annealing was also performed in these samples for 8 hours at 390 K.

Four-probe resistance was measured using a standard lock-in technique, using two Stanford Research SR830 lock-in amplifiers to synchronously measure the voltages across both bare and Ga₂O₃ areas of Gr. Low current (100 nA) was supplied as a current source through the devices to avoid Joule heating. The highly-doped Si-substrate was used as a backgate with voltage applied through a Keithley 2400 sourcemeter.

S5. Raman Spectroscopy

Raman spectroscopic measurements were acquired at room temperature using a Renishaw InVia microscope, using a blue 488 nm source with a 50X objective. Data was taken across at least 10 points in a sample. A quadratic baseline was subtracted from each spectrum. All curves were normalised to the G peak amplitude in order to facilitate comparison between different samples.

A. Electrical Analysis

To calculate the field-effect mobility, the ρ(Vg) data was first smoothed using a Savitzky-Golay filter to remove discretisation at high carrier density. This was then differentiated, followed by application of a second Savitzky-Golay filter.
S6. Self-consistent theory for graphene’s conductivity

Graphene’s conductivity on SiO$_2$ is calculated using self-consistent theory from Adam et al. At high carrier density \( n > n^* \), the residual carrier density, graphene’s conductivity \( \sigma \) is given by:

\[
\sigma = \frac{2e^2}{\hbar} \frac{n}{n_{imp}} \frac{1}{G[r_s]} \left[ \frac{\pi}{4} + 3x - \frac{3\pi x^2}{2} + \frac{x(3x^2 - 2) \arccos[1/x]}{\sqrt{x^2 - 1}} \right]
\]

where \( n_{imp} \) is the impurity density, and \( r_s \) is the characteristic interaction length (Wigner Seitz radius), calculated for a local environment dielectric of strength \( \epsilon \equiv \kappa \epsilon_0 \) where \( \epsilon_0 \) is the electric constant:

\[
r_s[\kappa] = \frac{1}{4\pi \epsilon_0} \frac{e^2}{\kappa}.
\]

\( \kappa \) is the average of relative dielectric constants of the layers above and below graphene:

\[
\kappa \sim \frac{\kappa_1 + \kappa_2}{2}.
\]

which gives the average relative dielectric constants for bare-Gr and Ga$_2$O$_3$-Gr,

\[
\kappa_{\text{SiO}_2/\text{Bare}} \sim \frac{3.9 + 1.0}{2} = 2.5,
\]

\[
\kappa_{\text{SiO}_2/\text{Ga}_2\text{O}_3} \sim \frac{4.0 + 10.0}{2} = 7.0,
\]

and \( r_s \) for bare-Gr and Ga$_2$O$_3$-Gr,

\[
r_s^{\text{SiO}_2/\text{Bare}} = 0.8,
\]

\[
r_s^{\text{SiO}_2/\text{Ga}_2\text{O}_3} = 0.3.
\]

Hence, using Eq. (S1) we can deduce \( \sigma \) for bare-Gr and Ga$_2$O$_3$-Gr,

\[
\sigma_{\text{SiO}_2/\text{Bare}} = 20 \frac{e^2}{\hbar} \frac{n}{n_{imp}^{\text{SiO}_2/\text{Bare}}},
\]

\[
\sigma_{\text{SiO}_2/\text{Ga}_2\text{O}_3} = 45 \frac{e^2}{\hbar} \frac{n}{n_{imp}^{\text{SiO}_2/\text{Ga}_2\text{O}_3}}.
\]
A. Impurity concentration

To calculate the impurity concentration for each substrate, we can use a self-consistent RPA method.  

1. Bare graphene on SiO$_2$

For bare graphene, the conversion from mobility to impurity density is already calculated by Adam et al.:

\[
\frac{\mu_0}{\mu_{\text{SiO}_2/\text{Bare}}} \sim 50 \frac{n_0}{n_{\text{imp}}^{\text{SiO}_2/\text{Bare}}},
\]

where $\mu_0 = 10,000$ cm$^2$/Vs and $n_0 = 10^{10}$ cm$^{-2}$. Using an electron mobility of 2,800 cm$^2$/Vs, the impurity density is inferred to be

\[
n_{\text{imp}}^{\text{SiO}_2/\text{Bare}} = 1.8 \times 10^{12} \text{ cm}^{-2}.
\]

2. Ga$_2$O$_3$-covered graphene on SiO$_2$

As the prefactor is different in eqn. S11 and eqn. S12 for Ga$_2$O$_3$-covered graphene, Eq. (S13) changes to become:

\[
\frac{\mu_0}{\mu_{\text{SiO}_2/\text{Ga}_2\text{O}_3}} \sim 110 \frac{n_0}{n_{\text{imp}}^{\text{SiO}_2/\text{Ga}_2\text{O}_3}},
\]

Using the mobility of the Ga$_2$O$_3$ covered area, 2900 cm$^2$/Vs,

\[
n_{\text{imp}}^{\text{SiO}_2/\text{Ga}_2\text{O}_3} = 3.8 \times 10^{12} \text{ cm}^{-2}.
\]

B. Change of $n^*$ and $\sigma_{\text{min}}$ in Ga$_2$O$_3$-covered graphene

According to the self-consistent theory by Adam et al. the minimum conductivity $\sigma_{\text{min}}$ in bare graphene on SiO$_2$ is

\[
\sigma_{\text{SiO}_2/\text{Bare}}^{\text{min}} = 20 \frac{e^2}{\hbar} \frac{n^*_{\text{SiO}_2/\text{Bare}}}{n_{\text{imp}}^{\text{SiO}_2/\text{Bare}}},
\]

where $n^*$, the residual carrier density, is proportional to the $\delta V$, the width of the minimum conductivity plateau.

Similarly, for Ga$_2$O$_3$-covered graphene on SiO$_2$

\[
\sigma_{\text{SiO}_2/\text{Ga}_2\text{O}_3}^{\text{min}} = 45 \frac{e^2}{\hbar} \frac{n^*_{\text{SiO}_2/\text{Ga}_2\text{O}_3}}{n_{\text{imp}}^{\text{SiO}_2/\text{Ga}_2\text{O}_3}},
\]

$n^*$ can be determined from numerically solving the following self-consistent equation,

\[
\frac{n^*}{n_{\text{imp}}} = 2r_s^2 C_0^{\text{RPA}}(r_s, a = 4d\sqrt{\pi n^*}).
\]
where

\[ C_0^{\text{RPA}}(r_s, a) = -1 + \frac{4E_1(a)}{(2 + \pi r_s)^2} + \frac{2e^{-a r_s}}{1 + 2r_s} + (1 + 2r_s a) e^{2 r_s a} \left( E_1[2r_s a] - E_1[1 + 2r_s a] \right) \]

and

\[ E_1(Z) = \int_z^\infty t^{-1} e^{-t} dt \]  

We obtain the theoretically expected values for \( n^* \) and \( \sigma_{\text{min}} \) for SiO\(_2\) and Ga\(_2\)O\(_3\) using eqn. S19 and \( n_{\text{imp}} \) obtained in section S2.A. The results are shown in Table S1 below.

**TABLE S1: Theoretically expected \( n^* \) and \( \sigma \) for bare graphene and Ga\(_2\)O\(_3\)-covered graphene**

| Dielectric environment | \( n_{\text{imp}} \) (cm\(^{-2}\)) | \( n^* \) (cm\(^{-2}\)) | \( \sigma_{\text{min}} \) (e\(^2\)/h) |
|------------------------|-----------------------------|------------------|-----------------|
| Bare graphene on SiO\(_2\) | 1.8e12 | 4.4e11 | 4.9 |
| Ga\(_2\)O\(_3\)-covered graphene on SiO\(_2\) | 3.8e12 | 3.9e11 | 4.6 |
S7. Temperature-dependence of the hysteresis of $\sigma$-$V_g$ in bare graphene and Ga$_2$O$_3$-covered graphene

FIG. S2: Gate voltage-dependent conductivity $\sigma(V_g)$ at different temperatures for (a) bare graphene and (b) Ga$_2$O$_3$-covered graphene. The hysteresis becomes prevalent above 180 K. Dark colour indicates up-sweep, light indicates down-sweep.
TABLE S2: Intrinsic dielectric parameters used to compute ROP scattering.

Dielectric constants in units of $\epsilon_0$ and frequencies in units of meV. For Ga$_2$O$_3$, $\epsilon_0$ and is taken from Ref[4] and $\omega_{TO}$ is inferred from FTIR measurements in Ref[5].

|          | SiO$_2$ | Ga$_2$O$_3$ | HfO$_2$ | Al$_2$O$_3$ | h-BN |
|----------|---------|-------------|---------|-------------|------|
| $\epsilon^\infty$ | 3.9     | 10          | 17      | 12.53       | 5.09 |
| $\epsilon^{int}$   | 3.05    | -           | -       | 7.27        | 4.57 |
| $\epsilon^\infty$   | 2.5     | 3.55        | -       | 3.20        | 4.1  |
| $\omega_{TO1}$    | 55.6    | 67.7        | 40      | 48.18       | 97.4 |
| $\omega_{TO2}$    | 138.1   | -           | -       | 71.41       | 187.9|

S8. Remote optical phonon (ROP) Scattering Model

According to the model described by the references [6,10,11] when graphene is sandwiched between two different dielectrics, the optical phonon modes of the two dielectrics are modified to give rise to surface optical (SO) phonon modes. The electric field due to these modes may scatter carriers in an adjacent material, known as remote optical phonon scattering. The interface SO frequencies between two materials are calculated via a dielectric continuity equation

$$\sum \epsilon_{ox}(\omega) = 0$$  \hspace{1cm} (S21)

$$\epsilon_{ox}(\omega) = \epsilon^\infty + \sum_j N f_j \frac{\omega_{TO}^2}{\omega_j^2 - \omega^2}$$  \hspace{1cm} (S22)

where $f_j$ is the strength of the dielectric contribution of the $j^{th}$ transverse optical (TO) mode for oxide $i$ with $N$ many TO modes. $f_j$ corresponds to a difference between dielectric constants at different frequencies.

For example, for $N = 1$,

$$f_1 = \epsilon^0_{ox} - \epsilon^\infty_{ox}$$  \hspace{1cm} (S23)

Here $\epsilon^0_{ox}$ is the static dielectric constant and $\epsilon^\infty_{ox}$ is optical dielectric constant.

For $N = 2$,

$$f_j = \begin{cases} 
\epsilon^{int}_{ox} - \epsilon^\infty_{ox}, & j = 2 \\
\epsilon^0_{ox} - \epsilon^{int}_{ox}, & j = 1 
\end{cases}$$  \hspace{1cm} (S24)

Here, $\epsilon^{int}_{ox}$ is the intermediate dielectric constant describing the dielectric response of the insulator at some intermediate frequency $\omega_{int}$ such that $\omega_{TO1} \leq \omega_{int} \leq \omega_{TO2}$.

For bare graphene on an oxide dielectric, eqn. [S21] is modified to the following,

$$\epsilon_{ox}(\omega) + 1 = 0$$  \hspace{1cm} (S25)

The electron-ROP coupling strength for $k^{th}$ ROP mode resulting from the $i^{th}$ dielectric material is given by,
Here $k$ corresponds to the index of the $k^{th}$ SO mode resulting from $i^{th}$ material. $k$ also relates the dielectric pairs $\{\epsilon^{(k)}_{ox}, \epsilon^{(k+1)}_{ox}\}$ that correspond to the TO mode pairs seen in Eq. (S24) i.e. $\{\epsilon^{0}_{ox}, \epsilon^{int}_{ox}\}$ or $\{\epsilon^{int}_{ox}, \epsilon^{\infty}_{ox}\}$.

The various characteristic dielectric constants and relevant TO-mode frequencies for the various dielectric materials are mentioned in Table S2. For Ga$_2$O$_3$, we assume $\epsilon^{\infty} = 3.55$, similar to measurements in non-amorphous Ga$_2$O$_3$ ($\alpha$ and $\beta$ polymorphs).

Solving Eq. (S21) we find the resulting SO modes of various dielectric environments. Their corresponding electron-phonon coupling strengths are obtained from eqn. S26. The resultant ROP frequencies and the coupling strengths for various dielectric environment for graphene, e.g. bare graphene on SiO$_2$ Table S3, bare graphene on hBN Table S4, HfO$_2$-covered graphene on SiO$_2$ Table S5, and Ga$_2$O$_3$-covered graphene on SiO$_2$ Table S6.

### TABLE S3: Calculated SO mode frequencies and coupling strengths for bare graphene on SiO$_2$

| $\hbar\omega_1$ (meV) | $g_1$ (meV) |
|-----------------|---------|
| 61              | 2.6     |
| 149             | 5.8     |

### TABLE S4: Calculated SO mode frequencies and coupling strengths for bare graphene on hBN

| $\hbar\omega_1$ (meV) | $g_1$ (meV) |
|-----------------|---------|
| 102             | 1.6     |
| 197             | 3.3     |

### TABLE S5: Calculated SO mode frequencies and coupling strengths for HfO$_2$-covered graphene on SiO$_2$

| $\hbar\omega_1$ (meV) | $g_1$ (meV) |
|-----------------|---------|
| 53              | 5.3     |
| 71              | 1.1     |
| 145             | 1.7     |

### TABLE S6: Calculated SO mode frequencies and coupling strengths for Ga$_2$O$_3$-covered graphene SiO$_2$. Here $\omega'_1$, $\omega'_2$ are the perturbed SiO$_2$ modes, while $\omega_3$ originates in Ga$_2$O$_3$.

| $\hbar\omega'_1$ (meV) | $g_1$ (meV) |
|-----------------|---------|
| 56              | 1.0     |
| 95              | 8.1     |
| 147             | 2.0     |
### S9. Ga₂O₃ Atomic-force & Darkfield Microscopy

| Sample   | Microscopy | AFM |
|----------|------------|-----|
| Sample 2 P01 | ![Sample 2 P01 Image](image) | ![Sample 2 P01 AFM](image) |
| Sample 2 P12 | ![Sample 2 P12 Image](image) | ![Sample 2 P12 AFM](image) |
| Sample 3 P3  | ![Sample 3 P3 Image](image) | ![Sample 3 P3 AFM](image) |
| Sample | Microscopy | Darkfield |
|--------|------------|-----------|
| Dev4_03 | ![Microscopy Image] | ![Darkfield Image] |
| Dev4_04 | ![Microscopy Image] | ![Darkfield Image] |
S10. Extracting $\rho_{ROP}$ as a function of temperature

To analyse the phonon contributions to the resistivity of our devices, we wish to measure the temperature dependence of the resistivity at specific voltages away from the gate voltage of minimum conductivity $V_{g,min}$. However $V_{g,min}$ may drift with temperature giving rise to a spurious temperature dependence if it is not correctly determined. $V_{g,min}$ can be determined in a variety of ways, such as extrapolation of the linear slopes of $\sigma(V_g)$, or fitting the gate-voltage-dependent conductivity to a quadratic near the minimum. These may result in slightly different values which can change the result of the temperature dependent analysis.

We found the minimum conductivity $\sigma_{min}$ using four different methods: (1) we extrapolated the linear behaviour of $\sigma(V_g)$ for $V_g < V_{g,min}$ and $V_g > V_{g,min}$ to their crossing point, and (2-4) fit $\sigma(V_g)$ to quadratics applied to data (2) $\sigma < 1.1\sigma_{min}$, (3) $\sigma < 1.5\sigma_{min}$, and (4) $\sigma < 2.0\sigma_{min}$.

We then compared the temperature dependent resistivities for positive and negative $V_g - V_{g,min}$ of the same magnitude. We found that method (4) gave the best agreement of the temperature-dependent resistivity for positive and negative $V_g - V_{g,min}$ which are expected to be similar.

Quad 1.1 fits very nicely to local minimum, however fails to capture the curvature between linear slopes like Quad 2.0. Lin gives a similar estimate for Bare, but can deviate from Quad fitting for Ga$_2$O$_3$.

To fit the intrinsic longitudinal acoustic phonon contributions to graphene’s resistivity [Eq. (S27)] is used to apply a fit to both our Ga$_2$O$_3$-covered and bare samples. A subset of temperatures are used to fit the acoustic phonons, such that influences from localisation and ROP scattering (high temperature) and quantum corrections to the conductivity (low temperature) do not affect the fitting.

$$\rho_{LA} = \left(\frac{\hbar}{e^2}\right) \frac{\pi^2 D_A^2 k_B^2}{2\hbar^2 \rho_s^2 v_s^2 v_F^2} T$$  \hspace{1cm} \text{(S27)}$$

The fits are shown in Fig. S3.

Here we used a temperature range of 70 K to 100 K to avoid the quantum corrections and ROP.
contributions. Given that the remote phonon contributions to graphene’s resistivity are expected to be smaller in the Ga$_2$O$_3$-covered graphene, we take the lower value of $D_A = 20.1 \pm 1.47$ eV found in the Ga$_2$O$_3$-covered graphene as the value for both systems when determining the $\rho_{LA}$ to subtract from both bare and Ga$_2$O$_3$-covered Gr when analysing for ROP contributions. The value of $D_A = 20.1 \pm 1.47$ eV is consistent with other experiments in the literature.

Lastly we propagate uncertainty in $D_A$ through to the parameter obtained from the fitting to the ROP contribution to the resistivity. This is done by calculating the $\rho_{ROP}$ contribution for each of the four methods to determine $V_{g,\min}$, and for three values of $D_A$ (18.6, 20.1, 21.6) eV. We use the standard deviation of these results to calculate the propagated uncertainty contributed from uncertainty in determining $D_A$ and $V_{g,\min}$, and combine it in quadrature with the fit uncertainty in $\rho_{ROP}$ to provide the values listed. The propagated uncertainties are similar to in magnitude to the $\rho_{ROP}$ fit uncertainties.
S11. Additional devices to assess reproducibility of changes in transport properties after Ga$_2$O$_3$ passivation and ALD

A. Device fabrication and general properties

We have fabricated and measured an additional 60 devices (30 with Ga$_2$O$_3$ passivation, and 30 undergoing identical processing but without Ga$_2$O$_3$) to demonstrate the consistent effect of the addition of Ga$_2$O$_3$. These devices use the same fabrication processing as described in the main text, with the additional etching gap between device channels as described below, and larger sheets of Ga$_2$O$_3$. The devices are macroscopic (each channel is 100 µm x 120 µm and are passivated using Ga$_2$O$_3$ sheets 1 mm long. To compare devices undergoing passivation with bare graphene, we have designed the devices in matched pairs in close proximity (40 µm away) to each other.

FIG. S4: Additional devices. Each group of devices consists of upper and lower graphene channels spaced 40 µm apart, with current electrodes at the ends and seven voltage probes allowing six channel sections (“devices” with dimensions 100 µm by 120 µm) to be probed independently on each channel. Ga$_2$O$_3$ was transferred on the lower channel of each device group, and the Al$_2$O$_3$ deposited by ALD over the entire structure. The micrographs above show the devices after Ga$_2$O$_3$ deposition but before Al$_2$O$_3$. 
FIG. S5: Conductivity (a,b) and field-effect mobility (c,d) of a pair of representative devices, measured at room temperature in a vacuum probestation. (a,c) show the device which was not passivated by Ga$_2$O$_3$, while (c,d) show the Ga$_2$O$_3$ passivated device. The legend indicates the measurements performed before Ga$_2$O$_3$ passivation, after passivation and after ALD.
B. Statistical analysis

1. Ga$_2$O$_3$ transfer

|                      | Bare Gr | Ga$_2$O$_3$/Gr Side |
|----------------------|---------|----------------------|
| $\mu_e^{After}/\mu_e^{Before}$ | 0.96±0.12 | 1.45±0.15          |
| $\mu_h^{After}/\mu_h^{Before}$ | 1.18±0.08 | 0.48±0.04          |

TABLE S7: Ratio of mobilities (after:before) Ga$_2$O$_3$ deposition. Electron ($\mu_e$) and hole ($\mu_h$) mobilities are shown for bare (control) and Ga$_2$O$_3$ passivated sides of device.

FIG. S6: Changes to (a) $\mu_e$ and (b) $\mu_h$ mobility measured at room temperature after Ga$_2$O$_3$ transfer step on bare and Ga$_2$O$_3$ passivated areas. Filled (open) circles correspond to voltage sweep direction in the -ve to +ve (+ve to -ve) direction. Colour indicates devices that are (i) physically connected to each other or (ii) in local proximity as a pair of bare-Ga$_2$O$_3$ devices as also indicated by device number.
2. ALD Al₂O₃ deposition

|                      | Bare Gr | Ga₂O₃ Gr |
|----------------------|---------|----------|
| μₑₐₜₜ / μₑₜₜ Before  | 1.17±0.28 | 1.12±0.11 |
| μₜₜₜ / μₜₜₜ Before  | 0.57±0.08 | 1.06±0.14 |

TABLE S8: Ratio of mobilities (after:before) ALD Al₂O₃ deposition. Electron (μₑ) and hole (μₜₜ) mobilities are shown for bare (control) and Ga₂O₃ passivated sides of device.

FIG. S7: Changes to (a) μₑ and (b) μₜₜ mobility measured at room temperature after the step of ALD deposition of Al₂O₃ on bare and Ga₂O₃ passivated areas. Filled (open) circles correspond to voltage sweep direction in the -ve to +ve (+ve to -ve) direction. Colour indicates devices that are (i) physically connected to each other or (ii) in local proximity as a pair of bare-Ga₂O₃ devices. (ii) is also indicated by device number in all cases with the exception of μₜₜ where two devices groups were too doped to properly measure hole mobility.
S12. References

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