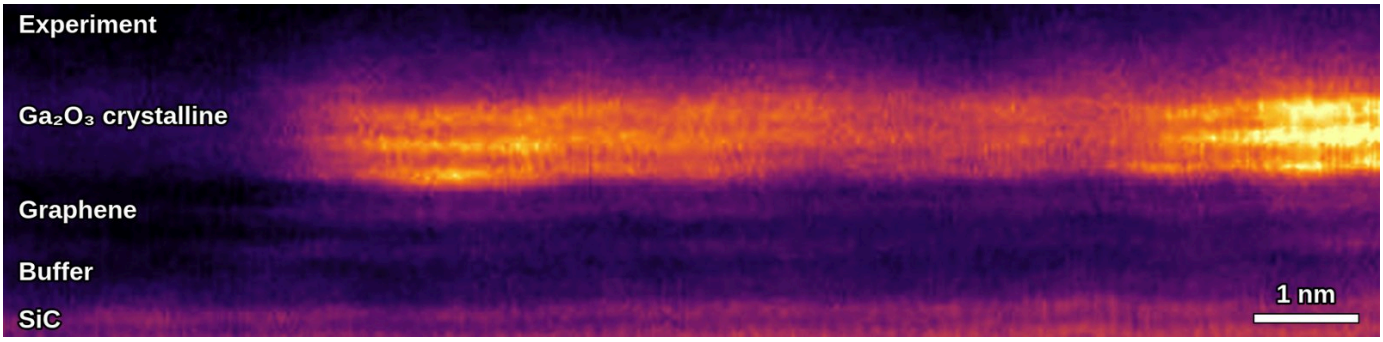

Fig. 1: Crystalline oxide on intact graphene. HAADF-STEM image of the ultrathin crystalline Ga_2O_3 layer on graphene and its carbon buffer layer on SiC. Upper part of Fig. 2b over its full width, taken from the unlabeled image in the paper and shown in false color with labels and scale bar redrawn here. Reprinted from the paper, Fig. 2d, CC BY 4.0.

Crystalline Gallium Oxide Dielectric

salve-project.de/newspress/crystalline-gallium-oxide-dielectric.html

February 02, 2026 – Researchers from the University of Nottingham (UK), Ulm University (Germany) and the Universidad de Cádiz (Spain) have turned a few layers of gallium selenide on graphene into an ultrathin film of crystalline gallium oxide (Ga_2O_3), a candidate high-κ dielectric for two-dimensional electronics. Atomic-resolution scanning transmission electron microscopy (STEM) shows a 1-nm-thick oxide whose first gallium layer sits 3.5 Å above an intact graphene sheet, and, together with EDX and EELS, how far the oxidation temperature can be raised before the graphene beneath is damaged.

As electronic components shrink further, two-dimensional semiconductors need equally thin dielectrics that isolate them electrically and allow precise gate control[1],[2]. Hexagonal boron nitride, the most widely used 2D dielectric, has a low static dielectric constant ($\kappa < 5$)[3], which limits its use in low-power devices[4],[5]. High-κ dielectrics ($\kappa > 10$) are needed, and layered gallium oxide (β-Ga_2O_3) is a thermodynamically stable candidate[4],[6]–[8] that can be made by oxidizing a two-dimensional precursor such as gallium selenide (GaSe)[9].

The team grew two to three layers of GaSe by molecular beam epitaxy on graphene on silicon carbide (SiC)[10] and oxidized them in oxygen at up to 410 °C, followed by annealing in ultrahigh vacuum at up to 800 °C. For comparison, 30-nm-thick GaSe layers were oxidized in a tube furnace at 400, 600 and 800 °C. The graphene beneath is the conducting layer that the oxide is meant to insulate, so it has to survive the process.

Fig. 1 shows what the process is meant to achieve: a band of crystalline gallium oxide lies on the graphene, which stays intact beneath it. How delicate this balance is becomes clear when two to three layers of GaSe are oxidized at 800 °C. The GaSe is then fully converted into gallium oxide, but the graphene disintegrates, and a layer of silicon oxide grows between the gallium oxide and the SiC (Supplementary Figure 5.2 of the paper, not reproduced here).

Cross sections of the samples were cut with a focused ion beam and examined in two laboratories. In Ulm, the surface was first covered with a protective carbon layer, a micrometre-thick slab was cut out with 30 kV gallium ions, and the lamella was polished with 5 kV ions to a final thickness of about 50 nm. In Cádiz, a few nanometres of gold against charging and a platinum layer protected the surface; after lift-out, the lamellae were thinned to 80–100 nm, cleaned with ions of 5–2 kV at 50 pA to reduce amorphization, and examined at a thickness of about 60 nm. At Ulm University, the 1-nm-thin oxides were imaged by STEM in a Thermo Fisher Talos 200X at 200 kV, which carries a windowless four-quadrant SuperX EDX detector for elemental mapping. At the Universidad de Cádiz, the thicker layers were studied in a double aberration-corrected FEI Titan Cubed Themis at 200 kV and 60 kV. Its high-angle annular dark-field (HAADF) STEM images reach a spatial resolution of 0.07–0.09 nm at 200 kV with a probe size of 0.1 nm, and integrated differential phase contrast (iDPC) STEM at 60 kV images light and heavy atoms together at atomic resolution; its enhanced contrast for light elements makes the atomic structure visible across the heterointerfaces. The microscope has a monochromated electron gun and a high-voltage tank of ultra-high stability. EELS spectrum images were recorded with a Gatan Quantum ERS energy filter in Dual-EELS mode, covering core loss and low loss. The STEM and EELS work was done by Manuel Mundszinger, Johannes Biskupek and Ute Kaiser in Ulm and by David González and Teresa Ben in Cádiz, among others.

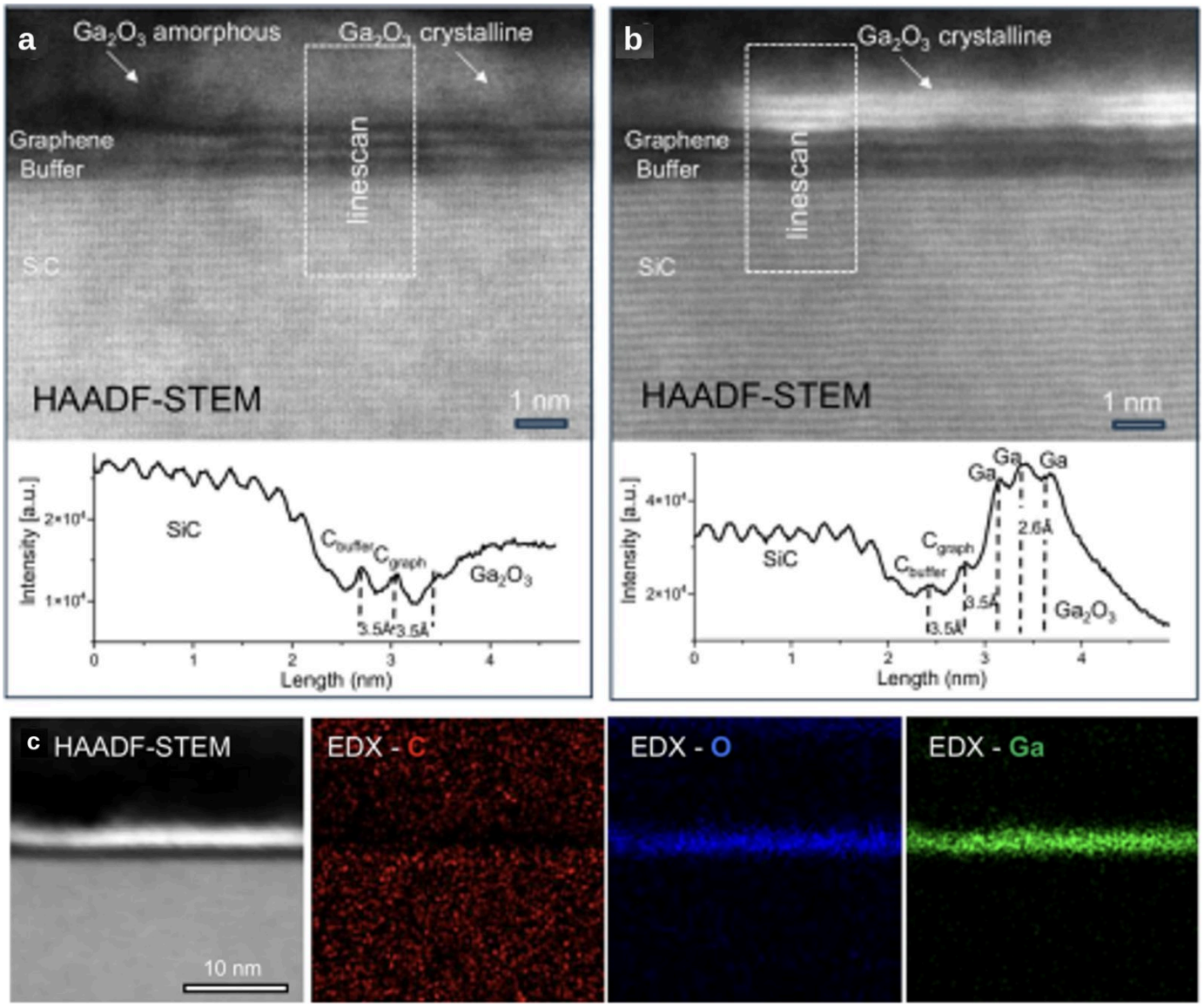

Fig. 2: Ultrathin Ga_2O_3 on graphene/SiC. (a, b) HAADF-STEM images of the oxide/graphene/SiC interface with (a) a partly crystalline oxide layer with crystalline and amorphous regions and (b) a crystalline oxide layer. Underneath: intensity line profiles of the boxes marked "linescan", resolving SiC, the carbon buffer layer (C_{buffer}), the graphene (C_{graph}) and the gallium layers of the oxide. (c) HAADF-STEM image and EDX maps of C, O and Ga of an additional oxide/graphene/SiC sample. Panels c, d and f of the paper, relettered (a)–(c) here; the other panels are not shown. Reprinted from the paper, Fig. 2, CC BY 4.0.

HAADF-STEM shows an oxide only about 1 nm thick on top of the graphene and its carbon buffer layer, with regions that are partly (Fig. 2a) or fully (Fig. 2b) crystalline. Intensity line scans across the interface resolve the stacking sequence: the SiC planes, the carbon atoms of the buffer layer and of the graphene, and the gallium atoms of the oxide. The first gallium layer lies about 3.5 Å above the graphene, and the three gallium layers of the oxide are about 2.6 Å apart. These values agree with those predicted for 2D β-Ga_2O_3 along the [100] direction[6] and with van der Waals oxides along their c-axis[4],[7]. EDX maps of a second sample show gallium and oxygen in the same thin band on top of the carbon-containing substrate (Fig. 2c).

The spacing of 3.5 Å matters for the graphene beneath. Calculations predict that hybridization with an oxide opens a gap in the Dirac cone of graphene only at distances below about 3 Å[11], so the graphene should keep its electronic properties under the oxide. Even where the oxide lacks long-range order, the STEM images show that it keeps a short-range order with well-defined bonds.

The thick layers show step by step how the oxidation proceeds (Fig. 3); each was annealed for 30 min in a flow of oxygen (0.5 sL/min) and argon (2.0 sL/min), and the inset in Fig. 3bi shows the full heterostructure. At 400 °C, the 30-nm-thick GaSe is only partly oxidized and converts into cubic α-Ga_2Se_3 and amorphous gallium oxide (a-Ga_2O_3), unlike the two to three layers, which are fully oxidized at that temperature. The iDPC images, recorded at 60 kV, show the light carbon atoms next to the heavier gallium and selenium, and they show that graphene and buffer layer stay intact up to 400 °C. At 600 °C, the

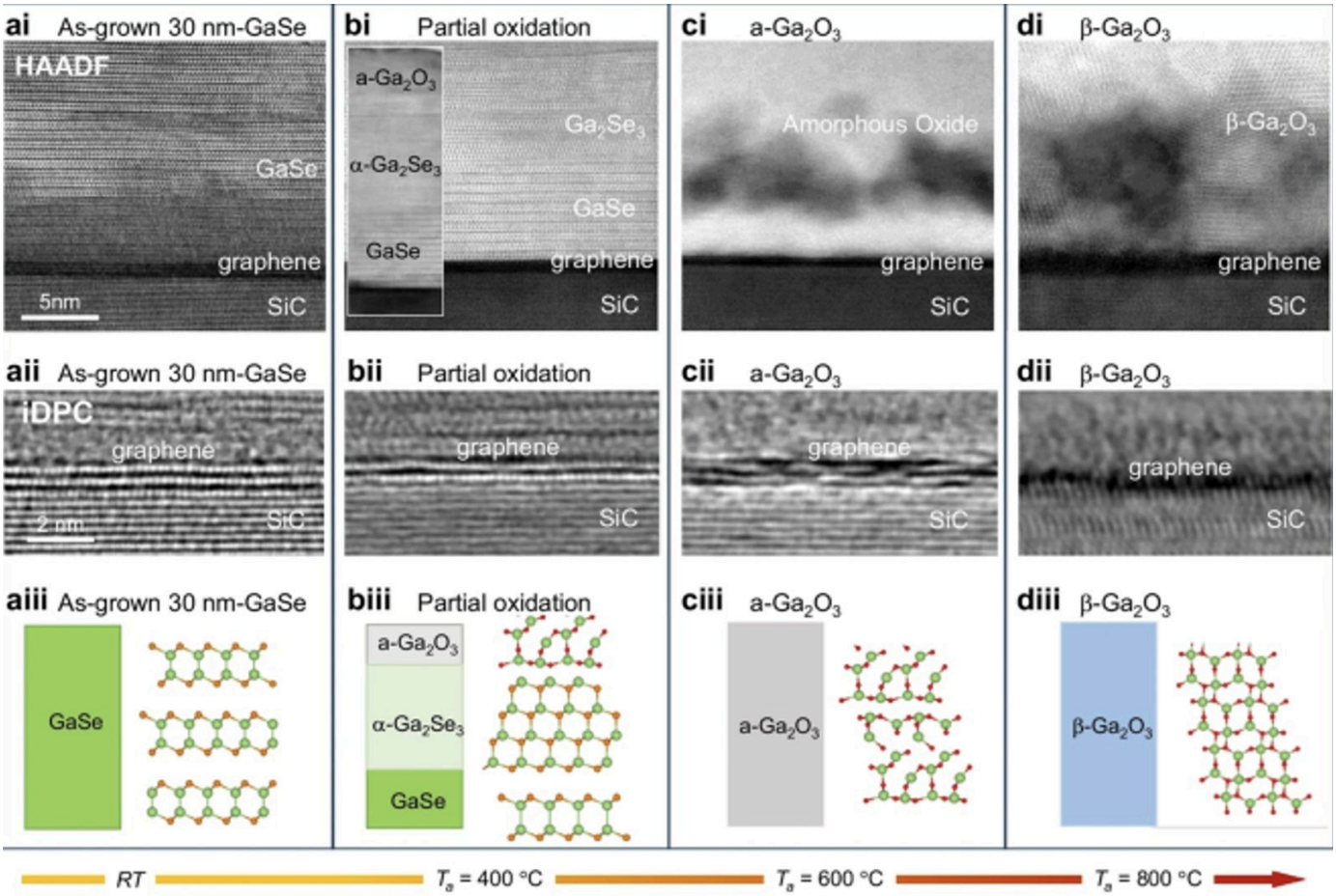

Fig. 3: Oxidation of GaSe/graphene heterostructures. (a–d) HAADF-STEM (ai–di) and iDPC (aii–dii) images of 30-nm-thick pristine GaSe/graphene/SiC (a) and of the same sample after thermal oxidation in a furnace at 400 °C (b), 600 °C (c) and 800 °C (d). The inset in panel (bi) illustrates the full heterostructure. (aiii–diii) Schematic of the evolution of the sample with increasing temperature of the oxidation, leading to the chemical conversion of centro-symmetric GaSe (aiii) into GaSe/Ga_2Se_3/a-Ga_2O_3 (biii), amorphous a-Ga_2O_3 (ciii) and β-Ga_2O_3 (diii). Reprinted from the paper, Fig. 3, CC BY 4.0.

GaSe is fully oxidized, with crystalline regions near the graphene; the graphene is still there but slightly distorted. At 800 °C, the GaSe is completely converted into β-Ga_2O_3, but the graphene has become strongly rough. The schematics below the images (Fig. 3aiii–diii) sum up the sequence: centrosymmetric GaSe turns into a stack of GaSe, α-Ga_2Se_3 and amorphous Ga_2O_3 at 400 °C, into amorphous Ga_2O_3 at 600 °C and into β-Ga_2O_3 at 800 °C.

EELS of the carbon K edge confirms this (Fig. 4). Spectrum images across the interface, which record a full spectrum at every point of the scan, give separate spectra for the SiC, the graphene and the layer above it, taken from the boxes marked in the HAADF images. Before oxidation and after 400 °C, the graphene spectrum shows the two well-defined peaks of graphene, the π peak at 285 eV and the σ peak at 293 eV, a fingerprint of the sp^2-bonded carbon of the intact layer; the same holds after 600 °C. The weak carbon signal from the layer above stems from organic contamination. After 800 °C, the spectra of the graphene and of the layer above can no longer be told apart, showing that the graphene has deteriorated. In the thin samples of two to three GaSe layers, the same temperature destroys the graphene altogether, as described above: a layer of silicon oxide takes its place.

Oxidation conditions can thus be chosen to create different structures, including crystalline oxide regions with a clean interface to graphene, which is well preserved up to about 600 °C. To keep the graphene intact, the oxidation therefore has to stay well below 800 °C. The oxidation also distorts the lattice and contracts its bond lengths, and its porosity and defects can let oxygen accumulate on the graphene surface, an effect that plays a critical role in the manufacture of high-quality graphene[12],[13]. The ultrathin oxide has a band gap of about 4.5 eV, band offsets of more than 1.5 eV to graphene and breakdown fields of 5–30 MV/cm. The STEM images also show what limits the oxide as a dielectric: the layers contain both crystalline and amorphous regions, and the authors infer that imperfections such as local cracks may change the breakdown field compared with an ideal oxide. Even so, the measured breakdown fields are the highest reported for β-Ga_2O_3 films, which makes the oxide a promising building block for scalable, miniaturized electronics.

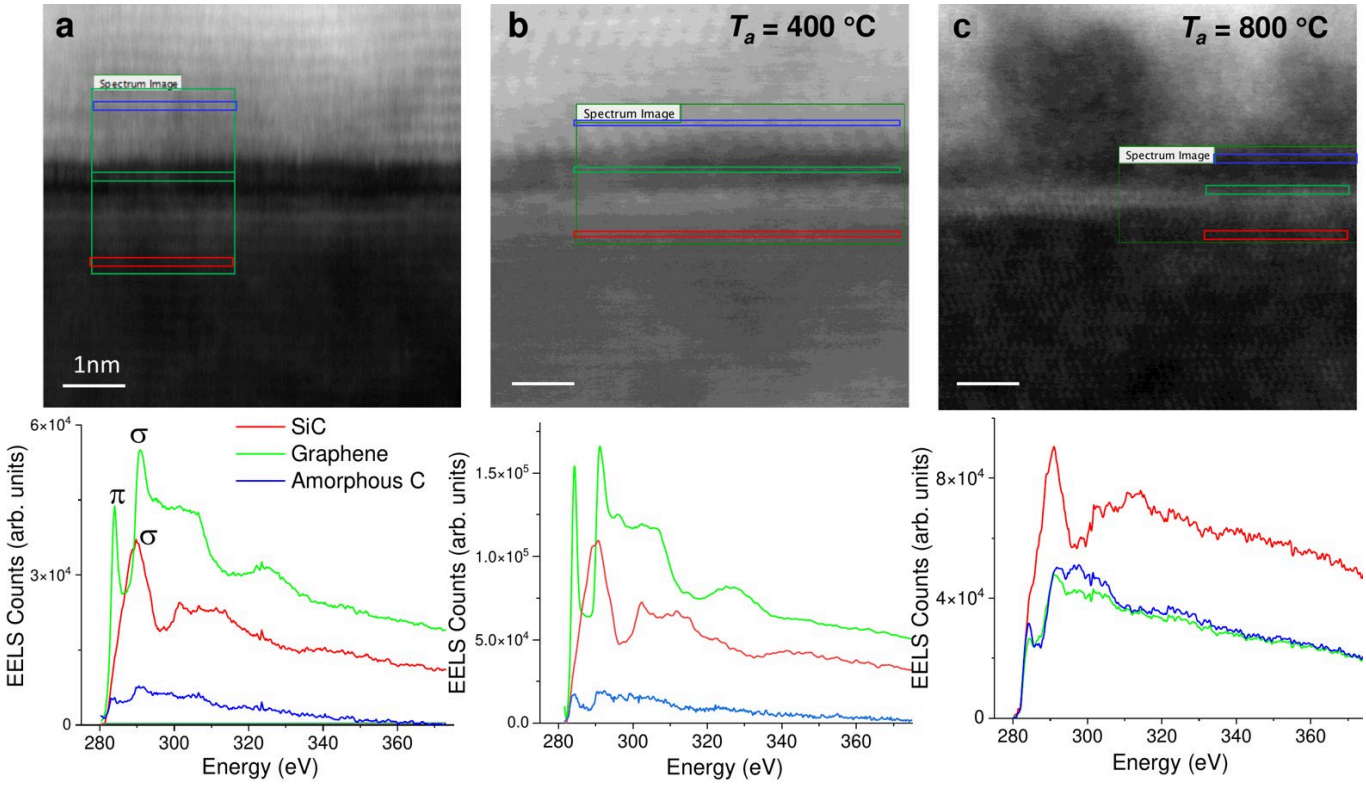

Fig. 4: EELS of the carbon layers. HAADF-STEM images (top) and EELS spectra of the carbon K edge (bottom) of 30-nm-thick GaSe/graphene/SiC before (a) and after thermal oxidation at 400 °C (b) and 800 °C (c). The spectra come from the boxes marked in the images: SiC (red), graphene (green) and the layer above it (blue). Scale bars 1 nm. Panel d of the Supplementary Information relettered (c) here. Reprinted from the paper, Supplementary Fig. 5.1, CC BY 4.0.

Resource

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