Supporting Information

Section 1. Mechanical cracking and removing the irregular parts on sapphire wafer surface during rough polishing.

During the rough polishing process on sapphire, Al_2O_3 abrasives played two roles in this process: mechanical cracking and removing the irregular parts on the wafer surface, and detaching and removing the formed chemical products from the wafer surface. However, considered the low formation rate of these chemical products, the mechanical cracking and removing by irregular-shaped Al_2O_3 abrasives played a dominant role in fast removing the irregularities on sapphire wafer surface. **Figure S1(a)** shows the main mechanism of this process, and **Figure S1(b)** shows corresponding images and surface roughness (Ra) values characterized and measured via optical interferometry profiler (OIP).

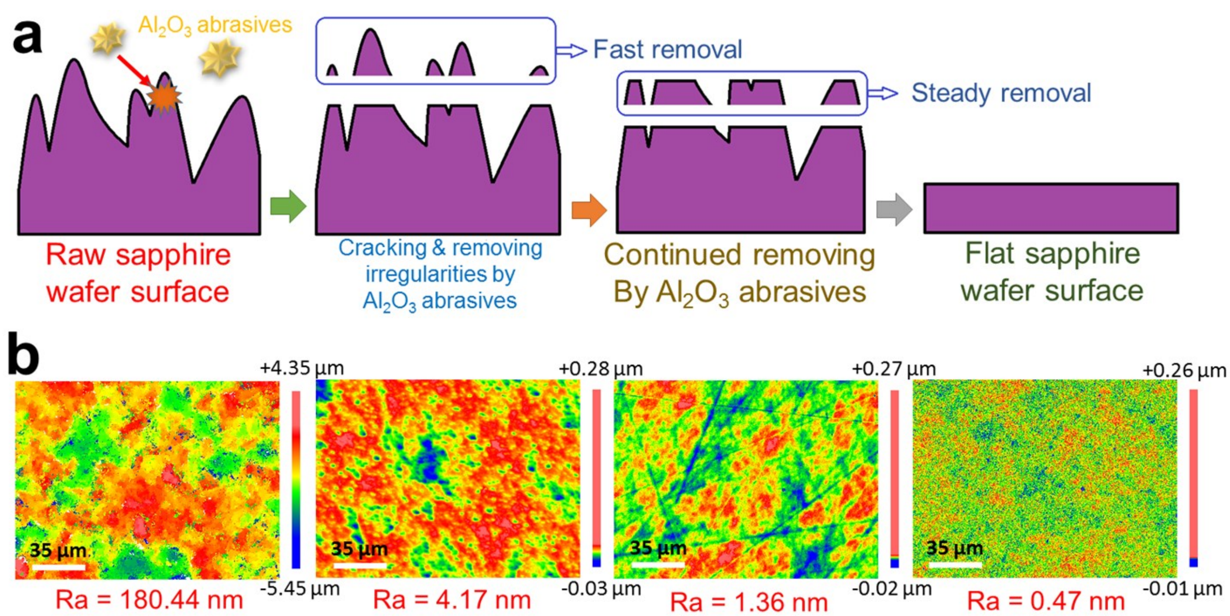
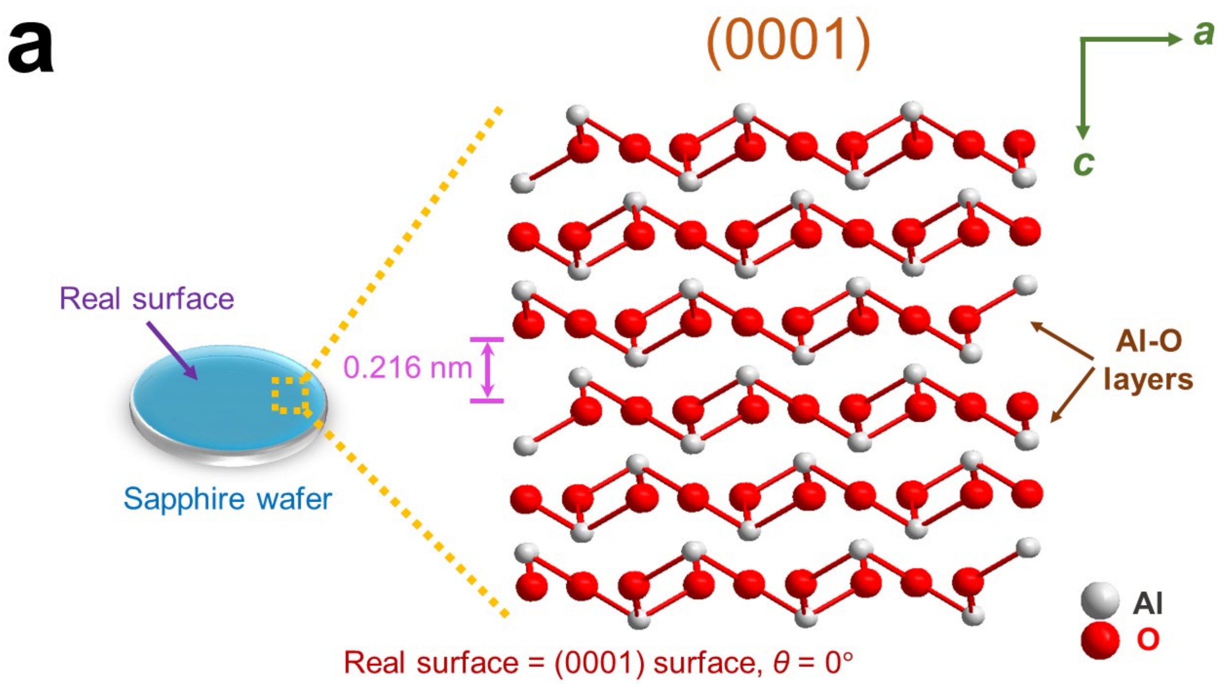


Figure S1. (a) The main mechanism of rough polishing for commercial sapphire wafer by Al_2O_3 abrasives. (b) Corresponding images and Ra values characterized and measured via optical interferometry profiler.

Section 2. A detailed discussion for the formation mechanism of atomic step-terrace morphology on sapphire (0001).

Generally speaking, the commercial sapphire wafers are cut from the single crystal ingots, and the cutting directions are mainly parallel to the (0001) surfaces (perpendicular to the *c*-axis) to ensure that the real sapphire wafer surfaces can fit well with ideal (0001) surfaces. **Figure S2(a)** shows a typical balls-and-sticks model of sapphire with ideal (0001) surface as its real surface. In this situation, the off-angle deflection $\vartheta = 0°$. However, due to the limitation of cutting precision, such cutting process always causes a slight miss-cut error, which results in a slight deflection of the real surfaces from ideal (0001) surface. In this situation, the off-angle deflection $\vartheta \neq 0°$ (within ±0.5°), as shown in **Figure S2(b)**. Thus, when the sapphire wafer surface reaches to an atomically smooth level, the atomic step-terrace morphology can be observed, which reflects a real surface of the sapphire wafer, as shown in **Figure S2(c)**. The section line taken from such step-terrace morphology shows the measured values of step heights (h) and widths (d), which can be used to calculated the average off-angle deflection degree (ϑ values) and the average distance of two neighboring Al-O layers (w, which is approximated to h when $\vartheta \approx 0$), as shown in **Figure S2(d)**. The average height of steps ($w \approx h =$ ~0.218 nm) fits well with the theoretical value of the distance between neighboring Al-O layers (0.216 nm),[1-4] indicating the successful achievement of atomically flat surface via CMP.



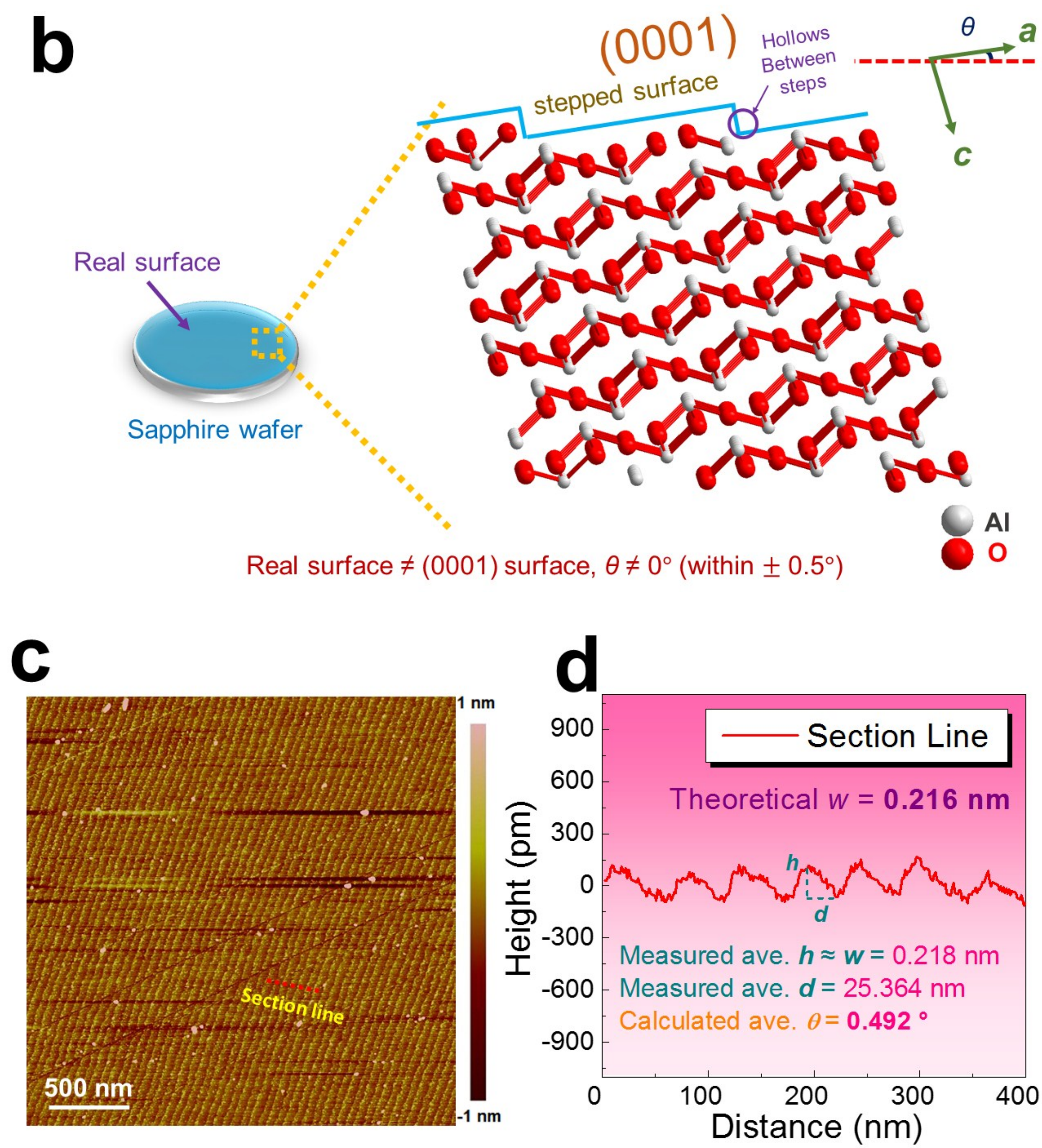


Figure S2. (a) An illustration of the case when the real sapphire wafer surface is (0001) surface. (b) An illustration of the case when the real sapphire wafer surface deflects from (0001) surface with an off-angle ϑ. (c) A typical AFM image of atomic step-terrace morphology which reflects the real sapphire wafer surface. (d) The section line taken from (c) to show the calculations of the off-angle ϑ and the distance between two neighboring Al-O layers w.

Section 3. An illustration of scan image when the set-point value is too high.

In our *in situ* study, if the set-point value is set as too high (such as 500 mv), the AFM tip cannot image correctly due to the poor vibration status of the cantilever, as shown in **Figure S3**. This is why we choose an appropriate set-point value of 440 mv in our in situ study.

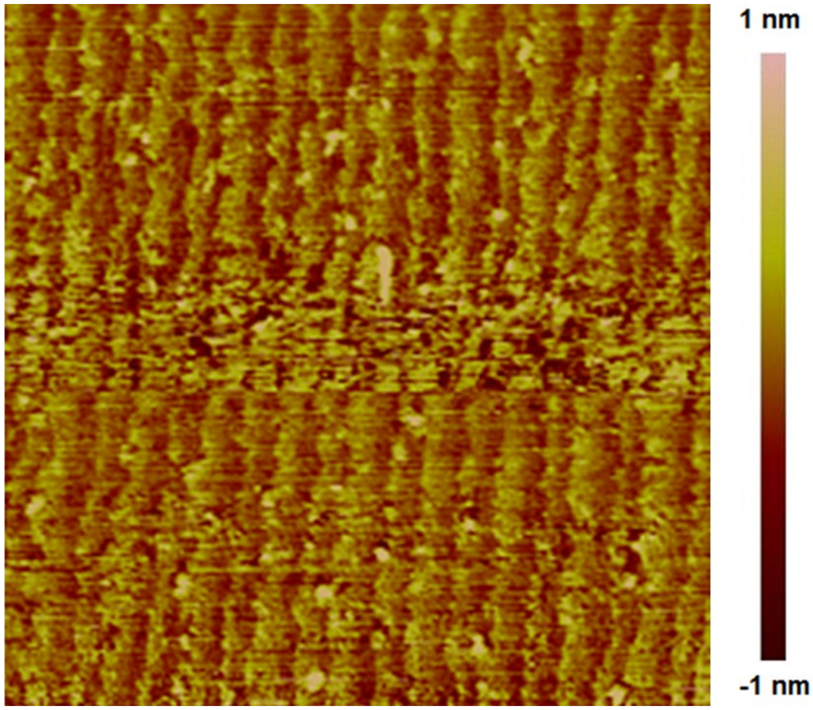


Figure S3. Blur image caused by over-set set-point value of 500 mv.

Section 4. Experimental details

Samples preparation. Several two-inch commercial sapphire wafers with 0°-off (< 0.5°, on-axis oriented) (0001) face were used. The wafers have been preliminarily polished by double sides lapping with a high degree of planar and parallel on the two sides.

The wafers were polished by CETR CP-4 machine (Bruker Corporation), and several polyurethane (PU) polishing pads were used for CMP. The rotating speeds of upper dynamic polishing head and polishing stage were 120 and 160 r min^{-1}, respectively. The process was carried out with slurry recycling flow. The slurries contained alumina (α-Al_2O_3) abrasives and silica abrasives (SiO_2) were prepared. The average size of alumina and silica abrasives was ~1.9 µm and ~70 nm, respectively. The slurry supplying rate was 70 ml min^{-1}, After CMP, the wafers were cleaned by liquid cleaner and deionized water, and then dried off by air spray gun for further characterizations.

Characterizations. The processed wafers were observed by Leica DM2500 optical microscope (Leica Microsystems Pty Ltd). The surface morphologies and surface roughness (Ra) values were evaluated by AFM (Bruker Dimension Icon) and OIP (Zygo New View 7200). For AFM, Si-based tapping mode probes (NSG11 series, NT-MDT) were applied for *in situ* study of simultaneous imaging and material manipulation. The tips from these AFM probes possess a typical tip radius of 10 nm, an aspect ratio of 3:1, a tip height of 10~15 µm, and a tip cone angle of < 22°. The reflective side of cantilever for laser reflection is covered by gold (Au). The typical resonant frequency is 255 kHz, and the force constant is from 5.5 to 22.5 N m^{-1}. For the tapping mode taken under liquid environment, a special fluid probe holder and fluid meniscus were used. The scan rate of AFM was 0.8 Hz, the set-point values for imaging and for *in situ* study were 150 mv and 440 mv, respectively, and the scan size was 768×768 points. For force-distance curve measurement, Si-based contact mode probes (CSG11 series, NT-MDT) were applied. The tips from these AFM probes possess a typical tip radius of 10 nm, an aspect ratio of 3:1, a tip height of 10~15 µm, and a tip cone angle of < 22°. The reflective side of cantilever for laser reflection is covered by Au. The typical resonant frequency is 20 kHz, and the typical force constant is 0.1 N m^{-1}. All probes were kept in vacuum environment before using to avoid potential oxidization. The working temperature was kept at 25 °C.

Reference

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4. R. Wang, D. Guo, G. Xie and G. Pan, *Sci Rep*, 2016, **6**, 29964.