Supplementary Information:

Atomistic Observation of Structural Evolution during Magnesium Oxide Growth

Fan Cao[†], He Zheng[†], Shuangfeng Jia[,†], Huihui Liu[†], Lei Li[†], Boyun Chen[†,‡], Xi Liu[†,§], Shujing Wu[†], Huaping Sheng[†], Ru Xing[⊥], Dongshan Zhao[†], and Jianbo Wang[*,†]*

[†] *School of Physics and Technology, Center for Electron Microscopy, MOE Key Laboratory of Artificial Micro- and Nano-structures, and Institute for Advanced Studies, Wuhan University, Wuhan 430072, China*

[‡] *Shuiguohu Senior Middle School, Wuhan 430071, China*

[§] *Middle School Attached to Huazhong University of Science and Technology, Wuhan 430074, China*

[⊥] *The Department of Physics Science and Technology, Baotou Normal College, Baotou 014030, China*

*Email: sfjia@whu.edu.cn; wang@whu.edu.cn

To whom correspondence should be addressed

Movie 1. Real-time video showing the growth of the three MgO variants. The time-code is shown in the format of mm:ss:ff (minutes:seconds:frame-intervals).

Movie 2. Atomic-scale observation of the grain rotation and GB migration process. The time-code is shown in the format of mm:ss:ff (minutes:seconds:frame-intervals).

Movie 3. The dislocation-mediated GB migration, as cut out from Movie 2. The time-code is shown in the format of mm:ss:ff (minutes:seconds:frame-intervals), and the time for beginning in Movie 3 is corresponding to 07:14:30 in Movie 2.

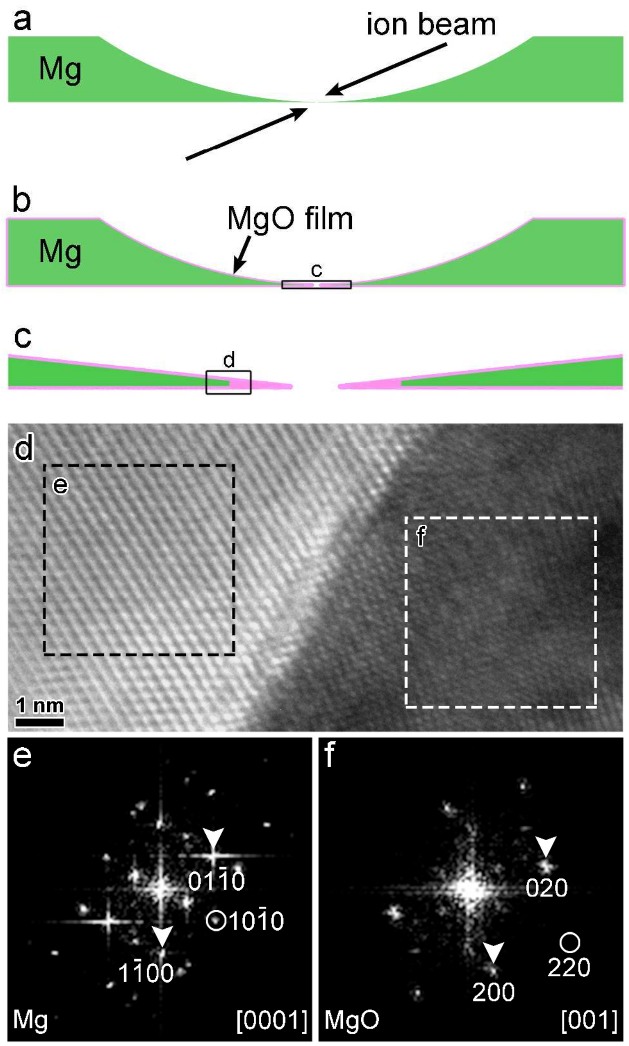

Figure S1. a) Cross-section view of obtaining a thin Mg substrate (colored green) by ion polishing. b) After annealed in air, the substrate was covered with MgO (colored pink). c) The thin area that could be penetrated by e-beam, corresponding to the black-boxed region in (b). d) HRTEM image corresponding to the black-boxed region in (c). e,f) Fast Fourier transformation (FFT) patterns corresponding to the left and right part of (d), respectively, implying the OR: $[0001]_H//[001]_C$, $(10\bar{1}0)_H//(110)_C$.

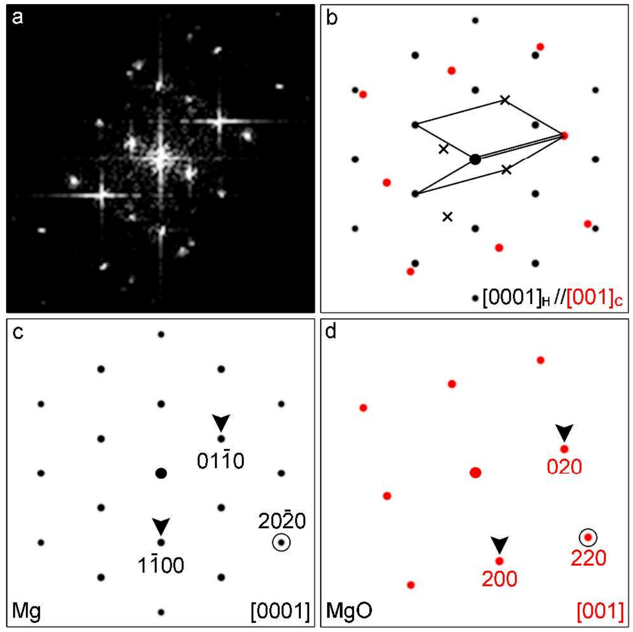

Figure S2. Further analysis of Figure S1e. a) The same FFT pattern as shown in S1e. b) Corresponding simulated diffraction pattern, comprised of (c) and (d), which indicates the OR: $[0001]_H//[001]_C$, $(10\bar{1}0)_H//(110)_C$ between Mg and MgO. The symbol "×" represents double diffraction spots and the routes are pointed out by black lines. c,d) Simulated diffraction patterns of Mg along [0001] and MgO along [001] directions, respectively.

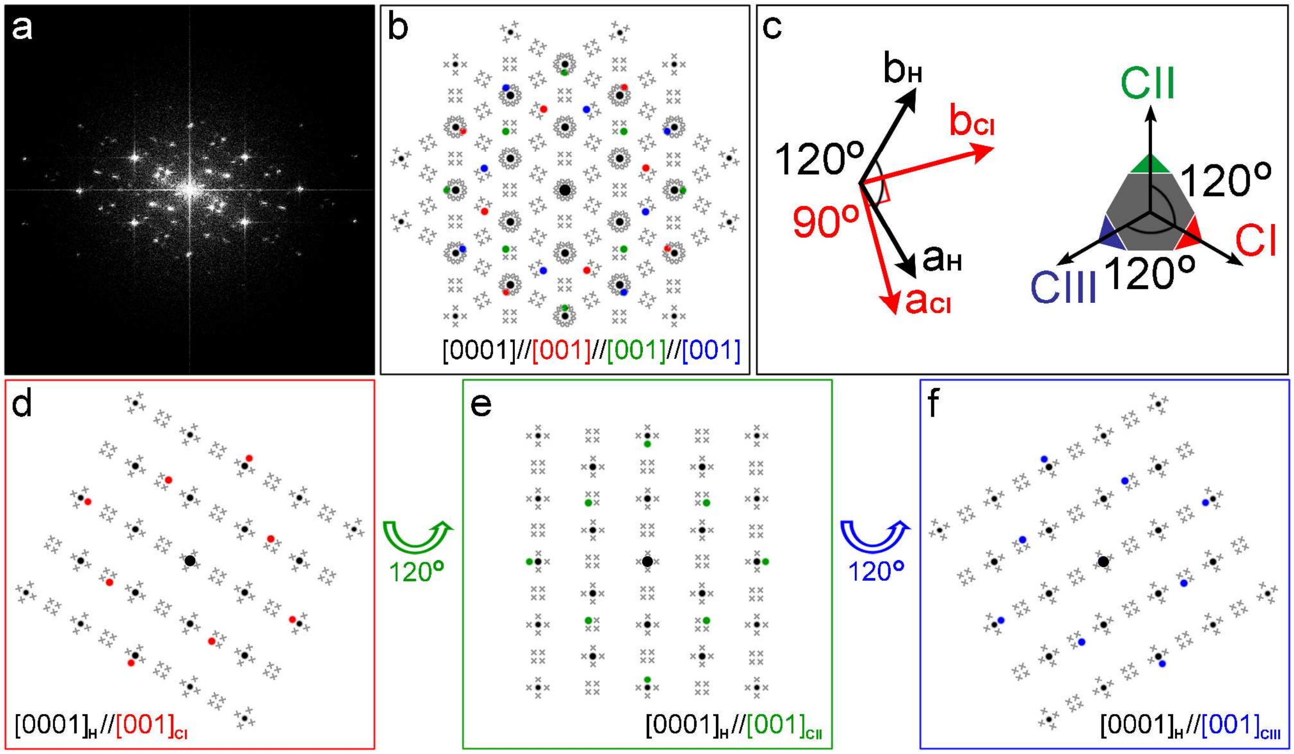

Figure S3. a,b) Fast Fourier-transformed (FFT) pattern of Mg substrate covered by MgO and the corresponding simulated diffraction pattern based on d-f): Mg matrix and the three MgO variants ("×" represents double diffraction spots), indicating the existence of three MgO variants. c) Schematic illustration of the OR between Mg matrix and the three MgO variants.

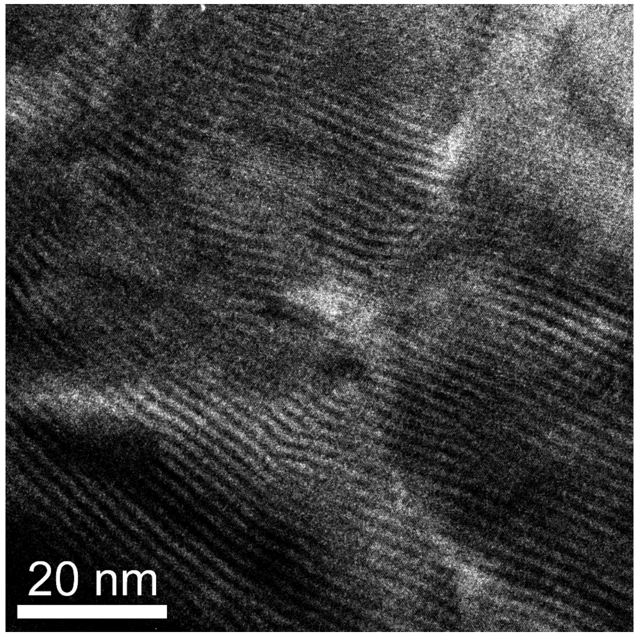

Figure S4. A lower magnification TEM image showing the Moiré fringes mentioned in Figure 1b.

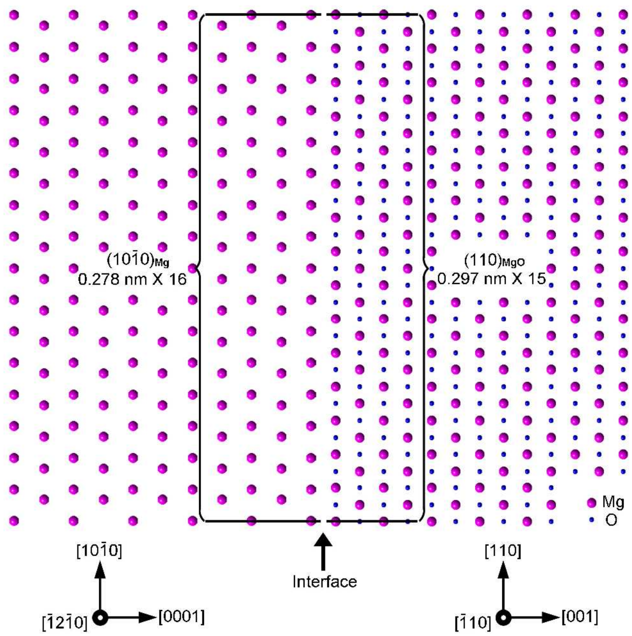

Figure S5. Side view of the possible atomic arrangements at the interface of Mg and MgO, based on a near coincidence site lattice (CSL) model. The mismatch is estimated to be $(0.297\times 15-0.278\times 16)/(0.278\times 16)=0.16\%$.

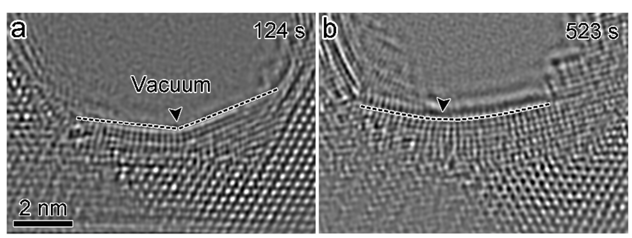

Figure S6. a,b) Same images shown in Figure 3d and e in the manuscript, respectively, depicting the GB between MgO grains and the vacuum with dashed black lines.

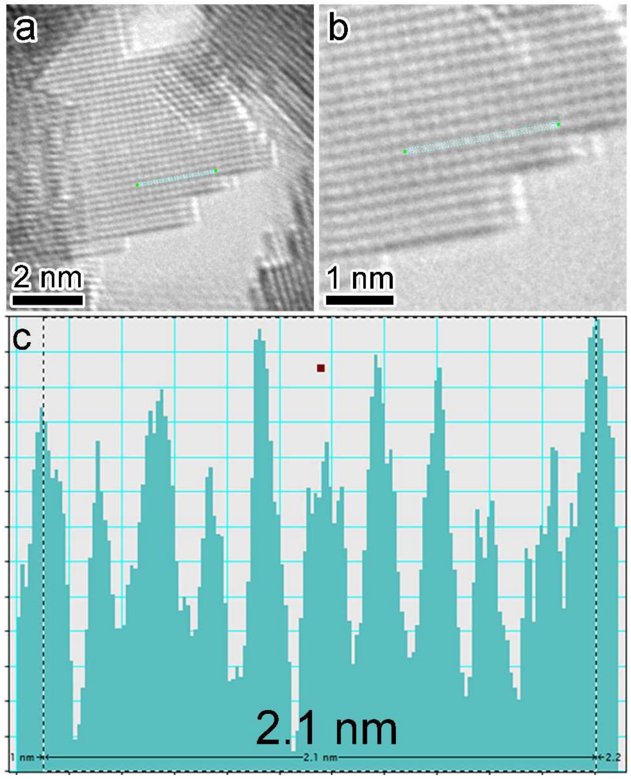

Figure S7. a) A dislocation-free MgO lattice image is chosen as the reference for the measurement of lattice strain. b-c) The spacing of $\{200\}_C$ planes is measured to be 0.21 nm, which matches well with the theoretical value (0.21 nm). As a result, the reference of sextuple lattice distance is $D = 0.21\,\text{nm}$.