Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.
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Supplementary Material: Isotopic study of Raman active phonon modes in $\beta - Ga_2O_3$

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AFM MICROGRAPHS

AFM micrographs evidence a morphology characterized by (i) the presence of deep trenches, almost orthogonal to the [001] in-plane direction, related to island-coalesence growth mechanism (Fig. S1a), [1] and (ii) (110) facets visible as elongated features oriented along the [001] orientation as a result of the metal-rich growth conditions of the layer (Fig. S1b).

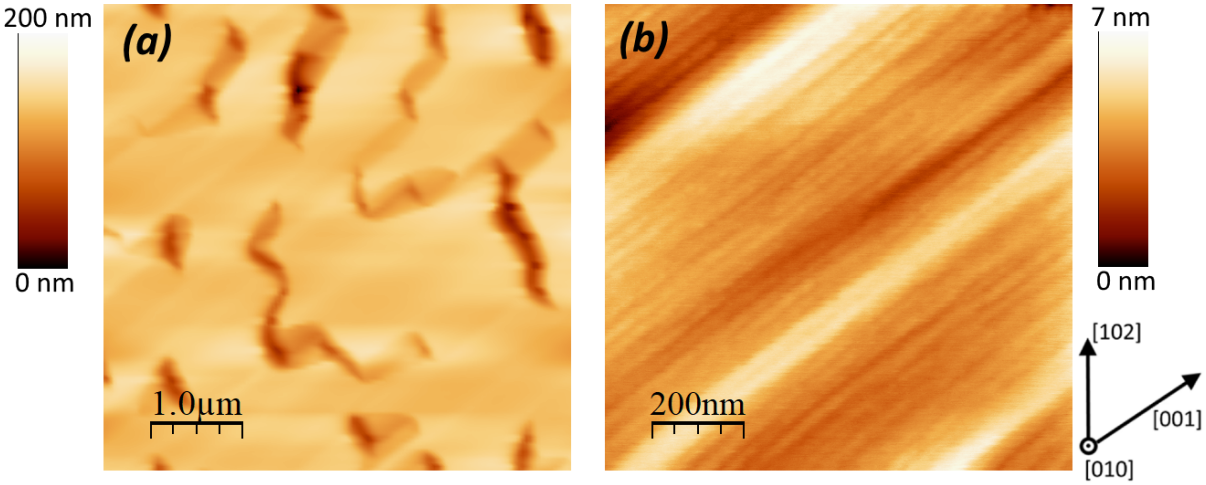

FIGURE S1. (a) 5x5 and (b) 1x1 μm AFM images of the 1.6 μm thick (010) β-Ga_2O_3 homoepitaxial layer deposited with ^{18}O isotopes.

TOF-SIMS MEASUREMENTS

Fig. S2 illustrates the ToF-SIMS depth profiles for the isotope fraction of ^{18}O. The ^{18}O isotope fraction n^* is calculated by the SIMS intensities:

$$n^* = \frac{I(^{18}O^-)}{I(^{16}O^-) + I(^{18}O^-)} \tag{S1}$$

In the film, 96.3% ^{18}O are employed (nominal isotope fraction of the gas: 97.39%). At the interface, the isotope fraction is decreased over a transient region with an extent of 300 nm to 0.3%, which is slightly above the natural isotope abundance of 0.2%.

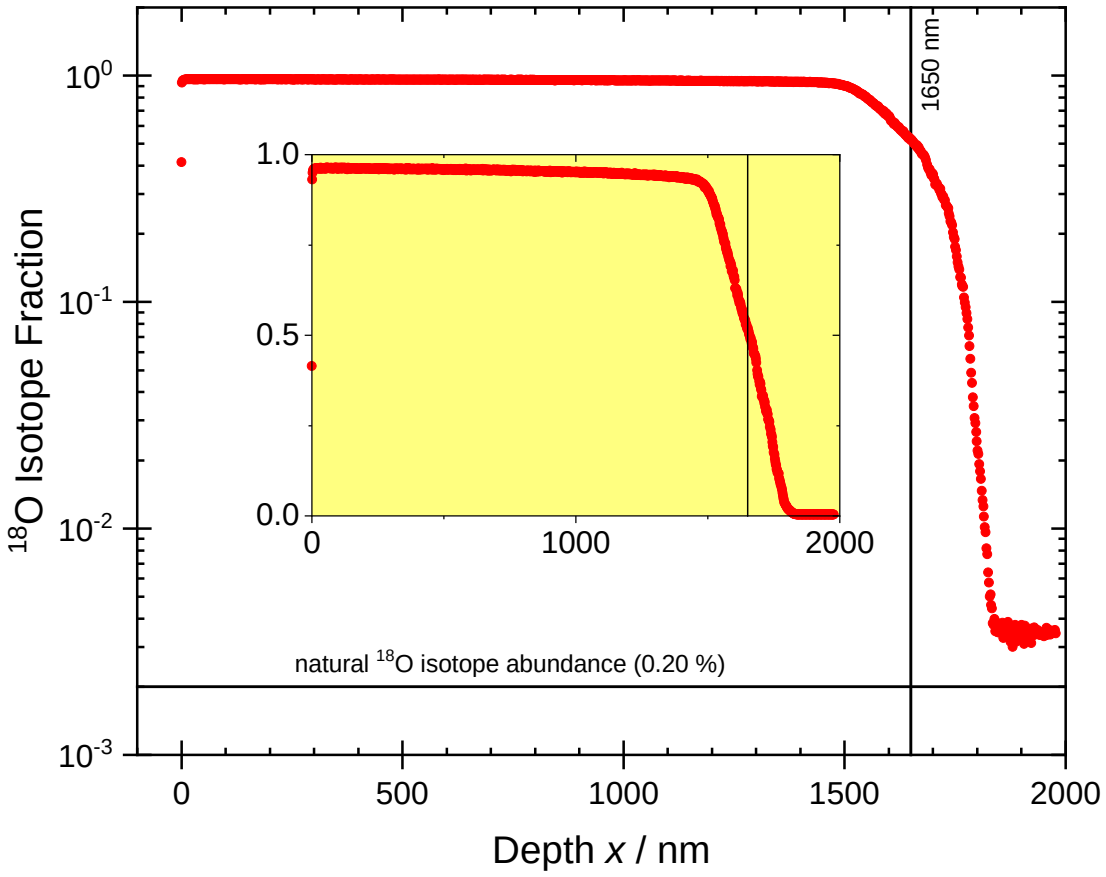

FIGURE S2. ToF-SIMS analysis of the thin film. ^{18}O isotope fraction analyzed with 25 keV Ga^+ analysis beam and 2 kV Cs^+ sputter beam. The interface (1650 nm) is obtained from the depth of 50% of the film's maximum isotope fraction. The inset shows the same graph with linear scale.

SCHEME OF RAMAN-ACTIVE PHONON MODES

For the monoclinic crystal structure of β-Ga_2O_3 there are 15 Raman-active phonon modes (10 with A_g and 5 with B_g symmetry), the schemes of which are illustrated in Fig. S3. A_g-modes are presented in projection on the b-plane. Arrows indicate the displacements of the corresponding atoms, with the length of the arrows representing each atom's

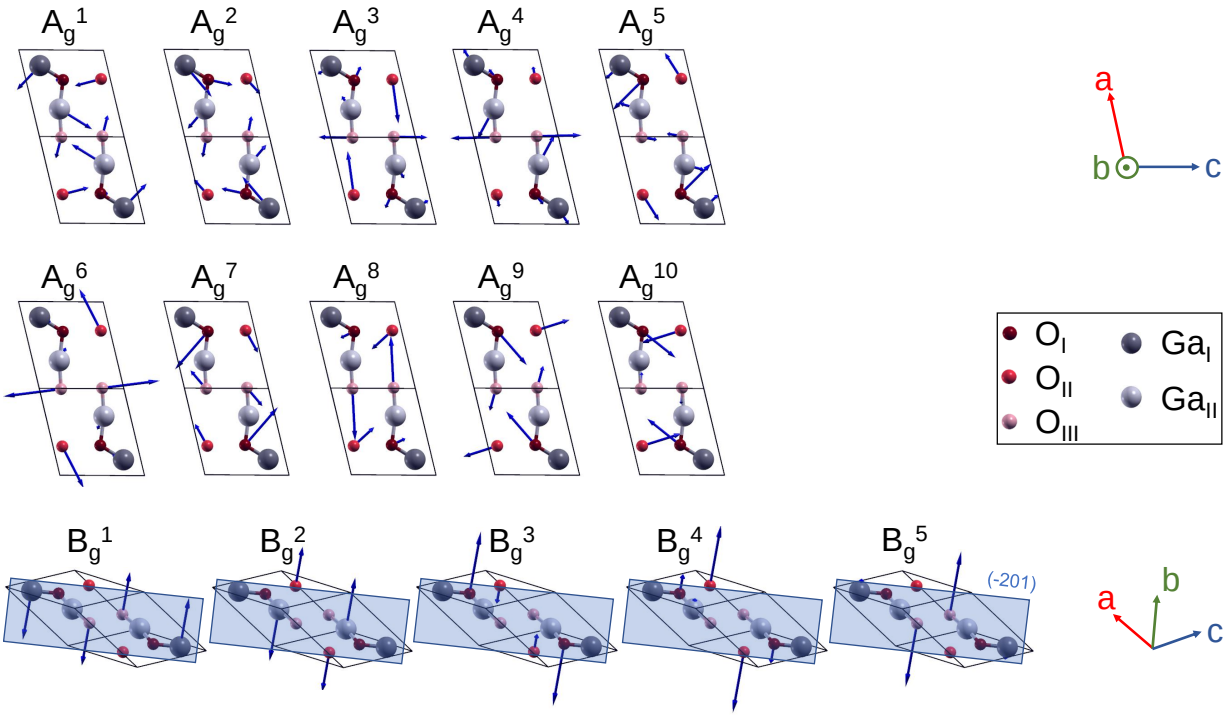

FIGURE S3. Scheme of Raman-active modes within the primitive unit cell of β-Ga_2O_3. A_g-modes are shown in projection on the b-plane. The $(\bar{2}01)$ plane (blue) is indicated for the illustration of B_g-modes. Arrows indicate the displacements of basis atoms, with lengths denoting the amplitude of vibration. Different red and grey colours indicate individual O and Ga lattice site atoms.

amplitude of vibration. A_g-modes are seen to oscillate within the b-plane, whereas modes of B_g-symmetry vibrate perpendicular to the same.

REFERENCES

1. P. Mazzolini and O. Bierwagen, "Towards smooth (010) $\beta - Ga_2O_3$ films homoepitaxially grown by plasma assisted molecular beam epitaxy: The impact of substrate offcut and metal-to-oxygen flux ratio," J. Phys. D. Appl. Phys. **53**, 354003 (2020).