Epitaxial hafnia-zirconia films on buffered fluorite substrates

De Luca G[1, 2]*, Ganguly S[1], Padilla J[1], Caicedo JM[1], Santiso J[1], Catalan G[1].
*gdeluca@icmab.es
1 Catalan Institute of Nanoscience and Nanotechnology (ICN2), Spain
2 Instituto de Ciencia de Materiales de Barcelona (ICMAB-CSIC), Spain

The discovery of ferroelectricity and antiferroelectricity in $Hf_{1-x}Zr_xO_2$ (HZO) fluorite oxides triggered a lot of attention because their promising integration with current semiconductor technology would facilitate the development of low-energy consuming devices [1,2]. Despite back-end-of-line-compatible polycrystalline films are the main ingredient for potential applications, stabilizing epitaxial HZO thin films enables a deeper understanding of their non-conventional ferroic properties [3]. So far, most of the research in this area has focused on the integration of fluorite HZO on the well-known perovskite oxide architecture [4,5]. One of the drawbacks of this heteroepitaxy approach is that the HZO grows textured with the co-existence of different columnar phases (monoclinic, orthorhombic) [5], thus inhibiting a clear determination of its structure-property relationship.

To overcome this limitation, we decided to use a radically different epitaxial design by growing the HZO on bixbyite-buffered and pristine Yttria-stabilized Zirconia (YSZ) fluorite substrates. To avoid any structural change induced by the Hf and Zr mutual doping, we focused on the x = 1 composition (ZrO_2). Our epitaxial films and buffer layers are grown by both off-axis magnetron sputtering and pulsed laser deposition and are characterized by a single crystalline phase. Various parameters (film thickness, growth temperature, buffer layer, growth method) affect the resulting ZrO_2 fluorite polymorph with compressive strain, in particular, inducing a structural phase transition in the fluorite. The generality of the approach suggests that functionality tuning induced by strain-, confinement- and interface-induced effects is not limited to fluorite oxides but can be extended to other systems with a similar crystal structure (pyrochlores, bixbyites).

References

[1] Müller J, Böscke TS, Schroeder U, Mueller S, Bräuhaus D, Böttger D, Frey L and Mikolajick T. Ferroelectricity in Simple Binary ZrO 2 and HfO 2. Nano Lett. 2012;12:4318.
[2] Schroeder U, Park MH, Mikolajick T and Hwang CS. The Fundamentals and Applications of Ferroelectric HfO2, Nat. Rev. Mater. 2022:7, 653.
[3] Nukala P, Ahmadi M, Wei Y, de Graaf S, Stylianidis E, Chakrabortty T, Matzen S, Zandbergen HW, Björling A, Mannix D, Carbone D, Kooi B, Noheda B. Reversible oxygen migration and phase transitions in hafnia-based ferroelectric devices, Science 2021:372, 630.
[4] Fina I and Sánchez F. Epitaxial Ferroelectric HfO 2 Films: Growth, Properties, and Devices, ACS Appl. Electron. Mater. 2021:3, 1530.
[5] Estandía S, Dix N, Gazquez J, Fina I, Lyu J, Chisholm MF, Fontcuberta J, Sánchez F. Engineering Ferroelectric Hf0.5Zr0.5O2Thin Films by Epitaxial Stress, ACS Appl. Electron. Mater. 2019:1, 1449.