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Multitarget reactive magnetron sputtering of Ba(Zr,Ti)O3 thin films for electrocaloric applications

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Relaxor ferroelectric $Ba(Zr,Ti)O_3$ thin films are a promising active material for lead-free electrocaloric refrigerators.
In this work, in-situ crystallized $Ba(Zr,Ti)O_3$ thin films with fractions of 12, 20 and 35% $BaZrO_3$ and a thickness of 200 to 300 nm were fabricated on 6-inch Pt(111)-coated Si wafers by multi-target reactive sputtering (MTRS) using three metallic targets (Ba, Zr, and Ti) for the first time. The advantage of MTRS is the possibility to control the $BaZrO_3$ fraction of deposited thin films by process parameters like power and temperature. The base of the Ba target was directly connected with the RF (13.56 MHz) generator. A unit for pulsed DC (100 kHz, duty cycle ~ 50 %) and arc suppression was connected between power supply and Ti target. High-power-pulse sputtering (50 kHz, duty cycle 50 %) was performed by connecting a switching unit between the DC power supply and the Zr-target. Composition analysis was performed by means of XPS and EDX.
SEM images show a dense textured film microstructure. XRD analysis evidenced a single cubic perovskite phase with a dominant (111)/[001] texture. Higher order (00l) peaks indicate a weak peak splitting. The optical properties of the film were investigated by VUV variable-angle spectroscopic ellipsometry. The optical band gap of the films amounted to 3.4 eV. Absorption maxima at about 5.2 and 7.6 eV were attributed to the O 2p valence band to Ti 3d conducting band transition and O 2p to Zr 4d transitions, respectively. Due to the short-range disorder, a pronounced Urbach-tail occurred with a tail width of about 0.3 eV. Films with higher $BaZrO_3$ fractions exhibit the characteristic for relaxor ferroelectrics: a large and frequency-dependent peak in the temperature dependence of dielectric permittivity. Thus, they provide the large and reversible polarization change required for a large electrocaloric response.

Keywords
multi-target reactive sputtering
electrocaloric BZT thin films