Ultrafast Crystallization Kinetics in (Pb,La)(Zr_{0.30}Ti_{0.70})O₃ Thin Films by Pulsed Excimer Laser Annealing

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Abstract

The crystallization kinetics of laser-annealed La-modified Pb(Zr,Ti)O₃ (PLZT) thin films on LaNiO₃-coated silicon substrates were investigated for substrate temperatures below 400°C. A KrF excimer laser having a ~20 ns pulse width and an energy density ~40 mJ/cm² was used to crystallize the films. The perovskite phase developed with cumulative laser pulse exposures; it was found that ~380 to 400 nm thick films could be fully crystallized for a total exposure time of 0.1 to 1 ms. Laser-crystallized films exhibited comparable dielectric and ferroelectric properties to those prepared by rapid thermal annealing at 650°C for 1 min. The evolution of the dielectric properties as a function of the number of laser strikes suggests that once nuclei are present, they rapidly grow through the depth of the film. This is consistent with the electron microscopy results, which did not show a well-defined planar growth front that proceeds from the top to the bottom of the film. The resulting films showed comparatively large lateral grain sizes (on the order of 250 to 300 nm), with high defect concentrations. The nucleation and growth mechanisms were modeled using Avrami kinetics under rate-dependent and nonisothermal conditions. These results indicate that PLZT crystallization via laser annealing is nucleation-limited.