Epitaxial growth of Piezoelectric $KNbO_3$ thin films by the PI-MOCVD process

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Among the large family of ferroelectrics, the perovskite-type $KNbO_3$ (KN) has attracted considerable interest in the field of agile dielectric devices for its electro-optic and acoustic properties. However, the synthesis of $KNbO_3$ single crystals is very challenging, and only expensive single crystals with small sizes are available at present. To bring this material towards industrial applications, the only remaining possibility is to use the grown $KNbO_3$ thin films. However, the growth of $KNbO_3$ films is far from being a routine process due to the high volatility and reactivity of K_2O. The chemical deposition methods, enabling better control of volatile element composition, face difficulty in finding a reliable K precursor. The industrially available K precursors present low volatility or high instability at ambient conditions.

KN films on C-sapphire, R-sapphire, and 36°Y-$LiNbO_3$ substrates (supplied by Roditi) were deposited by pulsed-injection metalorganic chemical vapor deposition (MOCVD) - a method providing digital control of film deposition. Mixtures of K_4(hfa)$_4$tetraglyme-Nb(thd)$_4$, dissolved in 1,2-dimethoxyethane, were used for the growth of $KNbO_3$ films, where thd - 2,2,6,6-tetramethyl-3,5- heptanedionate and hfa - hexafluoroacetylacetonate. Micro-doses of the solution were injected into a hot evaporator with a frequency of 0.5 Hz, and the vapor was transported to a hot substrate by a mixture of Ar and O_2 (33%) gases. The deposition temperature was 700°C. The phase composition and the texture have been analyzed utilizing X-ray diffraction (XRD). The stereographic projections have been visualized by using Winwulf software. The surface morphology and the elemental composition were studied employing surface scanning microscopy (SEM) and energy dispersive X-ray analysis (EDX). The $KNbO_3$ films, grown by using advanced K_4(hfa)$_4$tetraglyme and standard Nb(thd)$_4$ precursors presented a K/Nb ratio close to 1, as indicated by EDX analysis. The film texture and morphology were highly dependent on the substrate. In the case of R-sapphire substrates, a mixture of (011) & (100) with the presence of (111) orientation has been obtained. The morphology of these films consisted mainly of squares rotated by 90° and 45° in the substrate plane with respect to each other. In the case of C-sapphire, the dominating orientation was (120), which presents triangular growth symmetry, which can be identified in its stereographic projection as well. The purest growth texture was obtained on 36°Y-LN substrates, which is identified from the homogenous oriented squares in the morphology and XRD pattern.

Better control of K composition in $KNbO_3$ films has been demonstrated by using K4(hfa)4tetraglyme precursors with respect to the industrially available K(thd) precursor. This enabled the growth of pure KNbO3 films and their epitaxial growth on different substrates. This has enabled the growth of high-quality KN on substrates with bottom electrodes, and ferroelectric measurements are conducted. The preliminary results of P-E measurements will also be presented.