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Evolution of orientation degree, lattice dynamics and electronic band structure properties in nanocrystalline lanthanum-doped bismuth titanate ferroelectric films by chemical solution deposition

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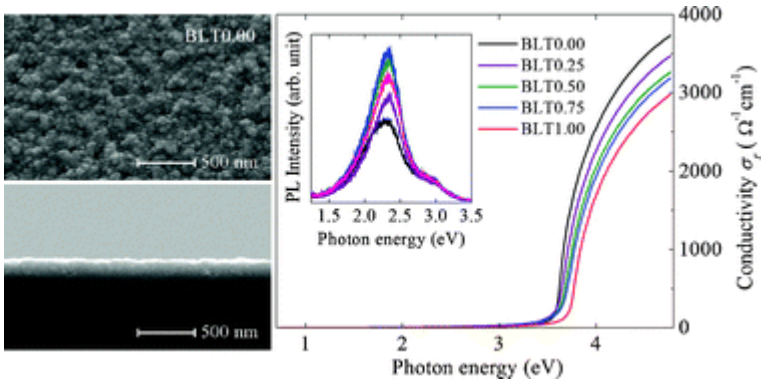
Ferroelectric lanthanum (La)-substituted bismuth titanate ($Bi_{4-x}La_xTi_3O_{12}$, BLT) nanocrystalline films with the composition range of $0 \le x \le 1$ have been directly deposited on n-type Si (100) substrates by chemical solution deposition. The La substitution effects on the preferred orientation, surface morphology, phonon modes, emission bands and electronic band structures of the BLT films have been investigated by microscopy, Raman scattering, photoluminescence and spectroscopic ellipsometry at room temperature. X-Ray diffraction analysis shows that the films are polycrystalline and exhibit the pure perovskite phase structure. With increasing La composition, the (100)-orientation degree can be enhanced and the root-mean-square roughnesses slightly increase from 6.5 to 8.3 nm. It was found that the Raman-active mode A_{1g}[Bi] at about 59 cm^{-1} is unchanged while the B_{1g} and A_{1g}[Ti] phonon modes at about 648 and 853 cm^{-1} are shifted towards higher frequency by about 36.6 and 8.4 cm^{-1}, respectively. Photoluminescence spectra show that the intensity of the peak located at about 2.3 eV increases with the La composition, except for the $Bi_3LaTi_3O_{12}$ film, due to the smallest grain size and oxygen vacancy defects. The optical constants of the BLT films have been uniquely extracted by fitting the measured ellipsometric spectra with a four-phase layered model (air/surface rough layer/BLT/Si) in the photon energy range of 0.73–4.77 eV. The Adachi dielectric function model has been successfully applied and reasonably describes the optical response behavior of the ferroelectric BLT films. Moreover, the film packing density decreases while the optical band gap linearly increases from 3.610 ± 0.066 to 3.758 ± 0.068 eV with increasing La composition. It is surmised that the phenomena are mainly ascribed to the variations of the electronic structure, especially for the conduction band, which is perturbed by the La doping.



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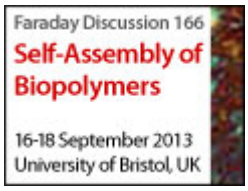

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