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Optoelectronic properties and polar nano-domain behavior of sol-gel derived $\text{K}_{0.5}\text{Na}_{0.5}\text{Nb}_{1-x}\text{Mn}_x\text{O}_{3-\delta}$ nanocrystalline films with enhanced ferroelectricity

[Qinglin Deng](#)^a, [Jinzhong Zhang](#)^a, [Ting Huang](#)^a, [Liping Xu](#)^a, [Kai Jiang](#)^a, [Yawei Li](#)^a, [Zhigao Hu](#)^{*a} and [Junhao Chu](#)^a

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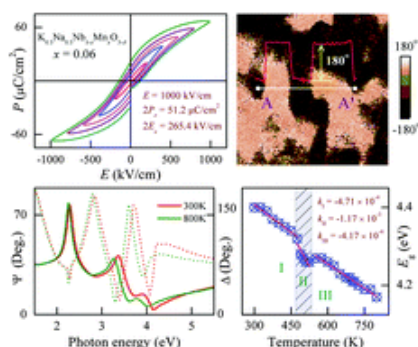
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High-quality lead-free piezoelectric $K_{0.5}Na_{0.5}Nb_{1-x}Mn_xO_{3-x}$ (KNNM x , $0 \leq x \leq 0.10$) films have been successfully deposited on Pt(111)/Ti/SiO₂/Si(100) substrates by a modified sol-gel method. The effects of Mn substitution on the microstructure, morphology, lattice vibrations, and optical and ferroelectric properties of the KNNM x films have been investigated in detail. All films are polycrystalline, crack-free and show a pseudo-cubic (pc) structure with a thickness of about 215 nm. Raman analysis indicates that the characteristic frequency of ν_1 , ν_5 and $\nu_1 + \nu_5$ modes shifts towards lower wavenumbers with increasing Mn concentration. The optimal ferroelectric properties were obtained in the film doped with $x = 0.06$, whose remnant polarization ($2P_r$) and coercive field ($2E_c$) values at the applied electric field of 1000 kV cm^{-1} are $51 \mu\text{C cm}^{-2}$ and 265 kV cm^{-1} , respectively. The increased valence of Mn^{2+} , which is substituted at the Nb^{5+} site as Mn^{3+} , plays an important role in reducing the amount of both oxygen vacancies and holes. In addition, the dielectric functions of the KNNM x films have been uniquely extracted by fitting ellipsometric spectra with the **Adachi dielectric function model** and a four-phase layered model (air/surface rough layer/KNNM x /Pt) in the photon energy range of 1.5–5.5 eV. The optical band gap (E_g) slightly decreases, while the high-frequency dielectric constant (ϵ_∞) linearly increases with increasing Mn concentration. Moreover, temperature dependent optical dispersion behavior of the KNNM0.06 film has been investigated from 300 K to 800 K. The analysis of E_g and the extinction coefficient (κ) reveals the correlation between optical properties and structural phase transition. Furthermore, a distinct in-plane (180°) polar nano-domain pattern with a well-defined rectangular phase hysteresis loop has been observed in the KNNM0.06 film from piezoresponse force microscopy (PFM) experiments. The present results could be crucial for potential multifunctional KNN-based device applications.



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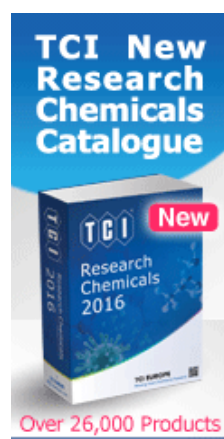
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