Supporting Information

Strong Electro-Optically Active Ni-substituted Pb($Zr_{0.35}Ti_{0.65}$)O_3 Thin Films: Toward Integrated Active and Durable Photonic Devices

Minmin Zhu,[abc] Zehui Du,[c] Soon Siang Chng,[a] Siu Hon Tsang,[c] Edwin Hang Tong Teo*[ab]

[a] *NOVITAS, School of Electrical and Electronic Engineering, Nanyang technological University, 50 Nanyang Avenue, Singapore 639798*

[b] *CINTRA, CNRS-International-NTU-THALES Research Alliances / UMI 3288, Research Techno Plaza, 50 Nanyang Drive, Singapore 637553*

[c] *Temasek Laboratories, Research Techno Plaza, 50 Nanyang Drive, Singapore 637553*

Electro-Optic Study:

The electric field induced optical modulations of PZTN thin films were then characterized by a customized ellipsometric setup using a 632 nm He-Ne laser as the light source. In this setup, an input laser beam is focused through film at an incident angle of 45° onto the top electrode. Under the applied electric field, the laser beam propagates through the PZTN thin film. Then an oscilloscope converts this phase shift ($\Delta\Phi$) into an intensity modulation, which is measured by a photodetector locked to the frequency of the excitation voltage. Therefore, the birefringence phase shift ($\Delta\Phi$) is given by:

$$\frac{I}{I_0} = \frac{1}{4}\sin^2\left(\frac{\Delta\Phi\pi}{\lambda}\right) \tag{1}$$

where I_0 and I are the incident laser intensity and the laser intensity through EO thin films in applied electric field, respectively. The linear EO effect (Pockels effect) alters the refractive index of the EO materials, which give rise to a shift in phase and thus convert into intensity modulation. The relationship between the phase shift ($\Delta\Phi$) and the birefringence shift (Δn) is given by:

$$\Delta\Phi = \frac{2\pi}{\lambda} l \Delta n \tag{2}$$

where λ and l are the wavelength of laser and the light path, respectively.

For these electro-optic materials, the linear and quadratic EO effects are defined as:

$$\Delta n = -\frac{1}{2} n^3 r_c E \tag{3}$$

where r_c, n, and E are the linear EO coefficient (Pockels coefficient), refractive index, and applied electrical field, respectively. Based on Equation (2) and (3) above, the linear EO coefficient (r_c) can be calculated:

$$r_c = \lambda \Delta \Phi / (\pi n^3 E L) \quad (4)$$

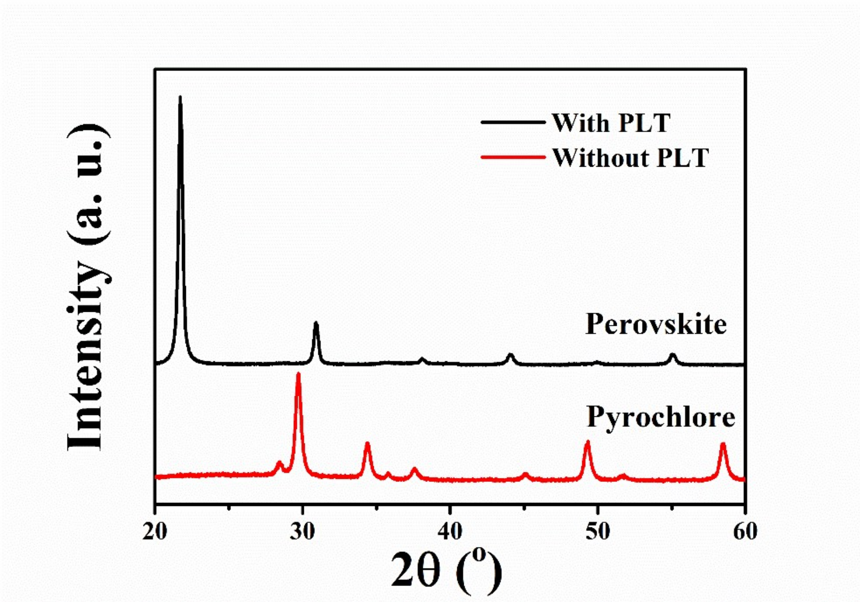

Fig. S1. XRD patterns of the as-grown PZT thin films with and without PLT seeding layer.

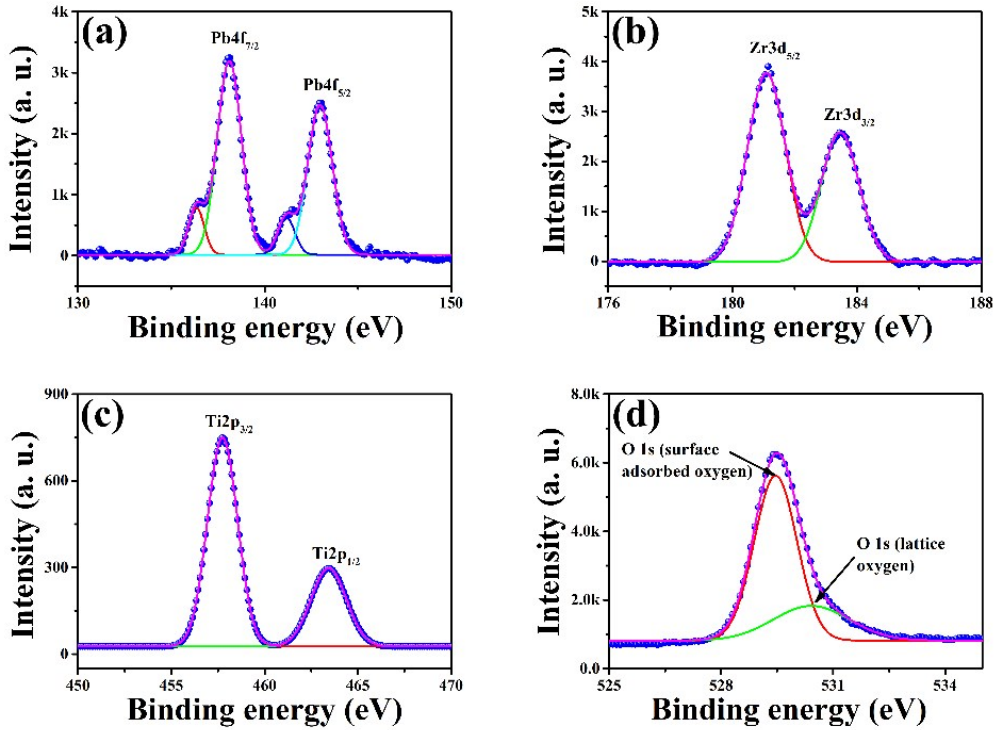

Fig. S2. High resolution spectra of (a) Pb 4f, (b) Zr 3d, (c) Ti 2p, and (d) O 1s photoelectrons for PZT thin film.

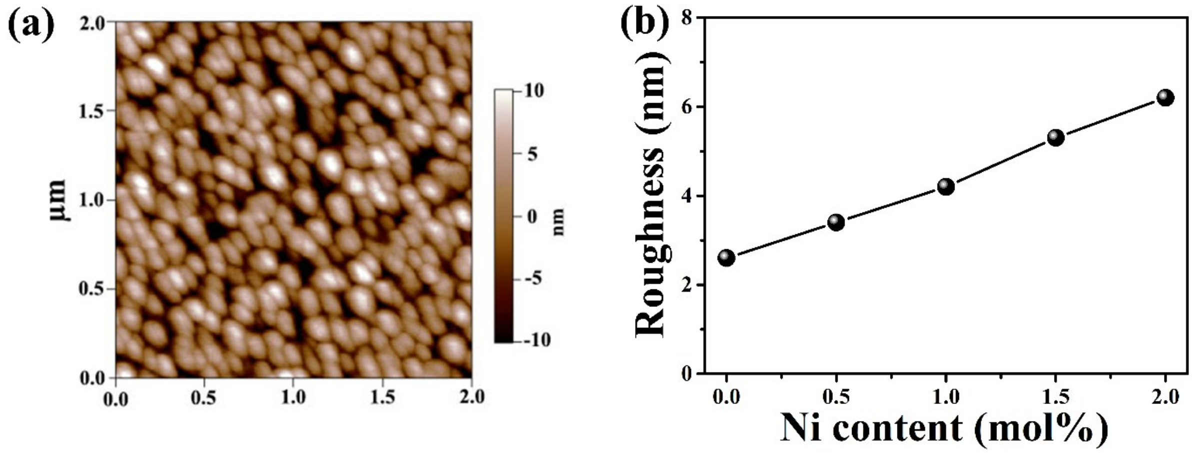

Fig. S3. (a) Roughness of PZTN20 thin film. (b) Roughness of PZTN thin films as a function of Ni content.