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Impact Of Substrate And Post-Annealing Temperatures On The Refractive Index And Crystallinity Of Lead-Free BCTZ Thin Films

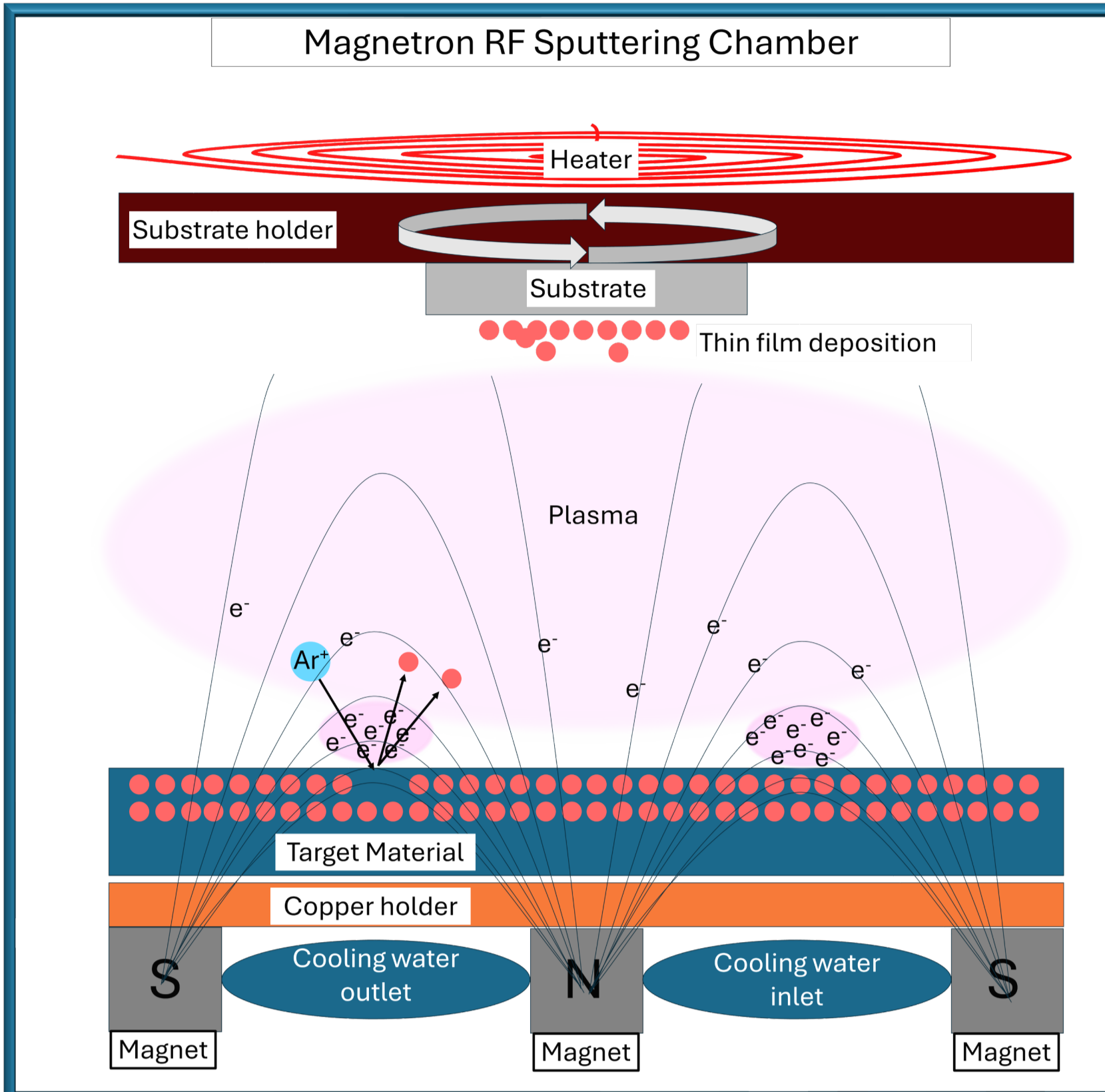
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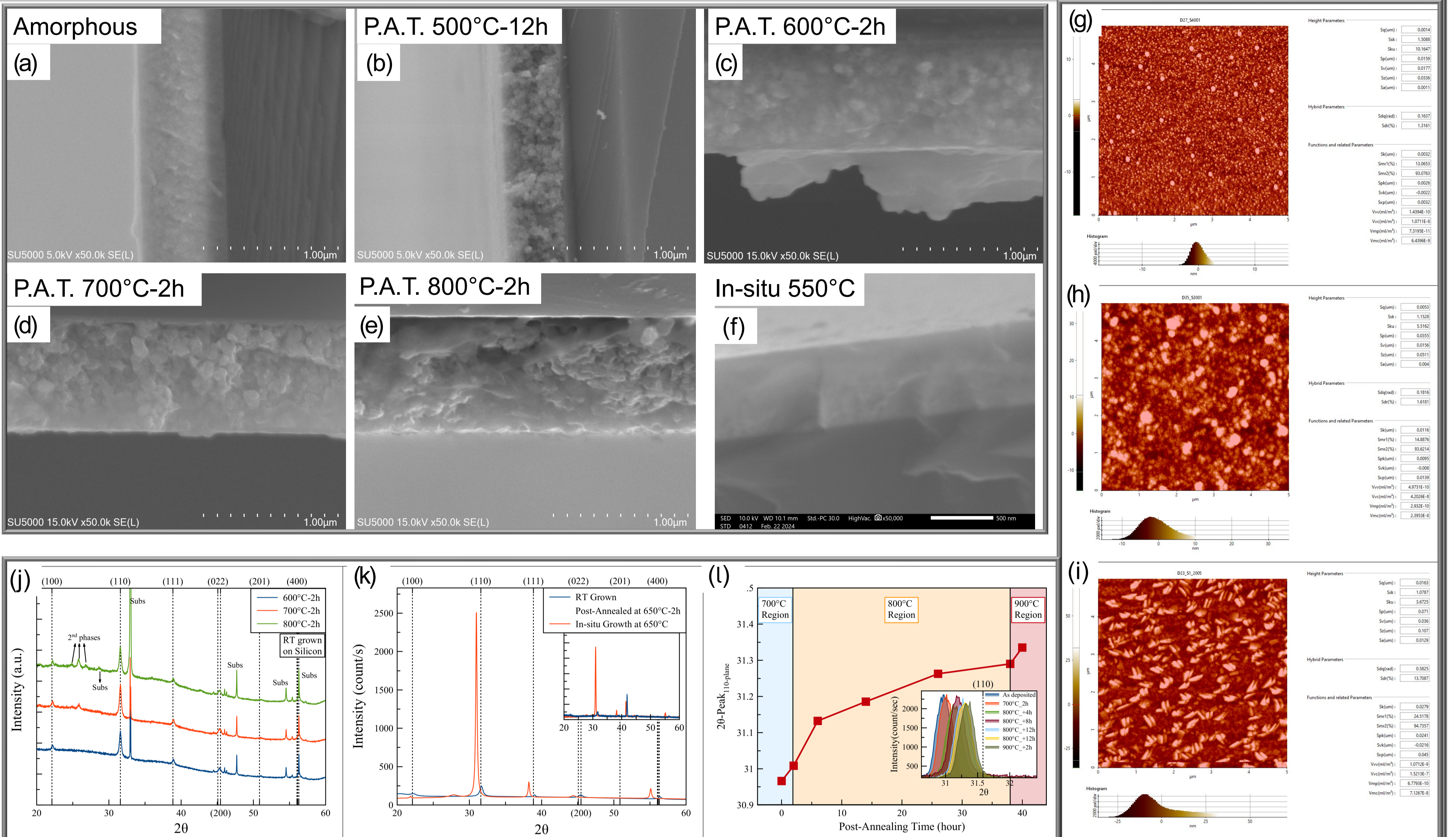
Abstract

In this study, we investigated the effects of substrate temperature and post-annealing treatment (P.A.T.) on the refractive index of lead-free $\text{Ba}_{0.85}\text{Ca}_{0.15}\text{Ti}_{0.9}\text{Zr}_{0.1}\text{O}_3$ (BCTZ) thin films synthesized by magnetron RF sputtering. Our results indicate that both the in-situ substrate temperature and the post-annealing treatment temperature influence the crystallinity of the films. Crystallinity was characterized using X-ray diffraction (XRD), while the refractive index was measured using the prism coupling method. Additionally, we utilized Atomic Force Microscopy (AFM) and cross-sectional Scanning Electron Microscopy (SEM) to analyze the film's roughness and in-plane morphology, respectively. We found that the refractive index of the films increased from 1.957 to 2.380, directly proportional to both the in-situ substrate temperature and the post-annealing temperature. However, significant porosity developed in the room-temperature-growth (R.T.G.) films which are subsequently subjected to post-annealing treatment at various temperatures. These findings highlight the critical role of temperature control in optimizing the optical properties and structural integrity of BCTZ thin films.

Structural Analysis

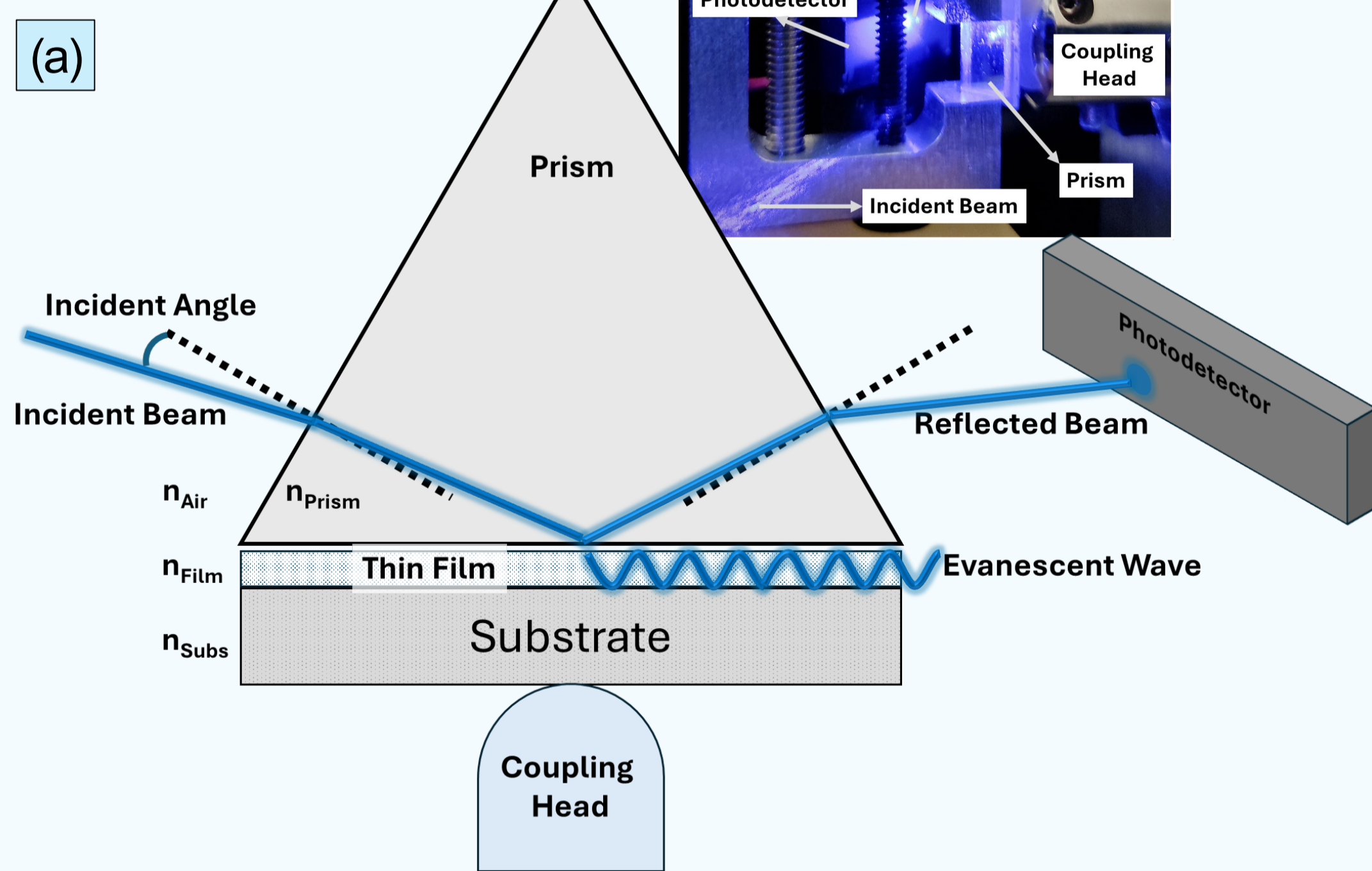
(a-f) SEM images of (a) amorphous, (b)(c)(d)(e) R.T.G BCTZ thin films with P.A.T. at 500°C for 12h, 600°C for 2h, 700°C for 2h and 800°C for 2h, respectively. Crystallization seems to start locally, and microstructure changes via increasing temperature. (f) In-situ depositions remove the porosity effect. (g-i) AFM images of in-situ deposited BCTZ films on (g) STO(110), (h) Pt/Ti/Si and (i) Sapphire (110) substrates. Sample on Sapphire has been annealed for 36h before taking this AFM measurement. Roughness is measured as (g) 1.4nm, (h) 5.3nm, (i) 16.3nm. (j) XRD comparison of the sample in figure (c-d-e), 2nd phases start to form at 700°C. These XRD results indicate the existence of perovskite structure. (k) 18 times enhanced XRD intensity for in-situ deposition at 650°C with respect to R.T.G. (l) The primary peak of BCTZ(110) on Sapphire evolves after undergoing P.A.T. at different temperatures and durations, indicating stress relaxation.

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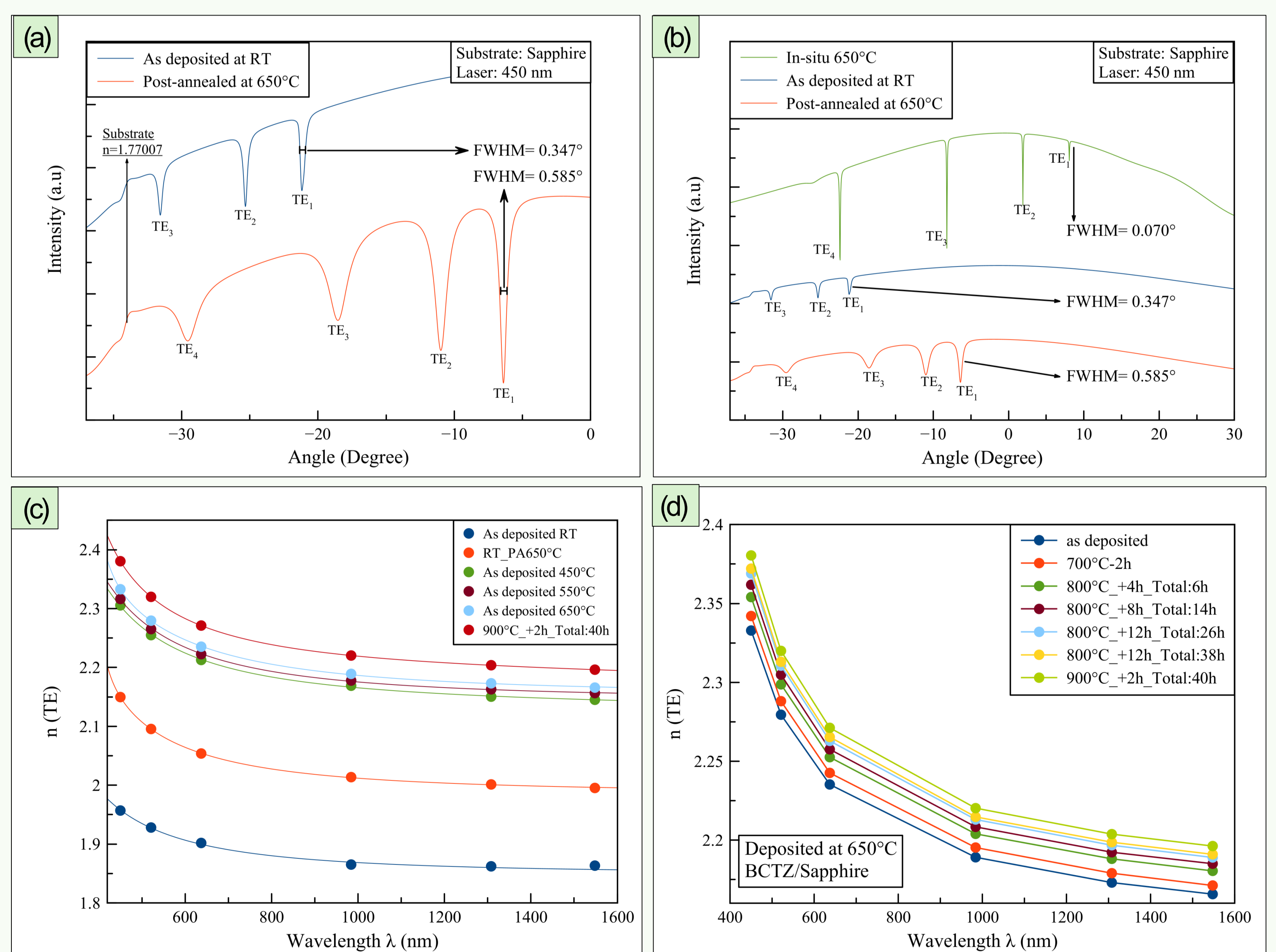


Prism-Coupling Measurements

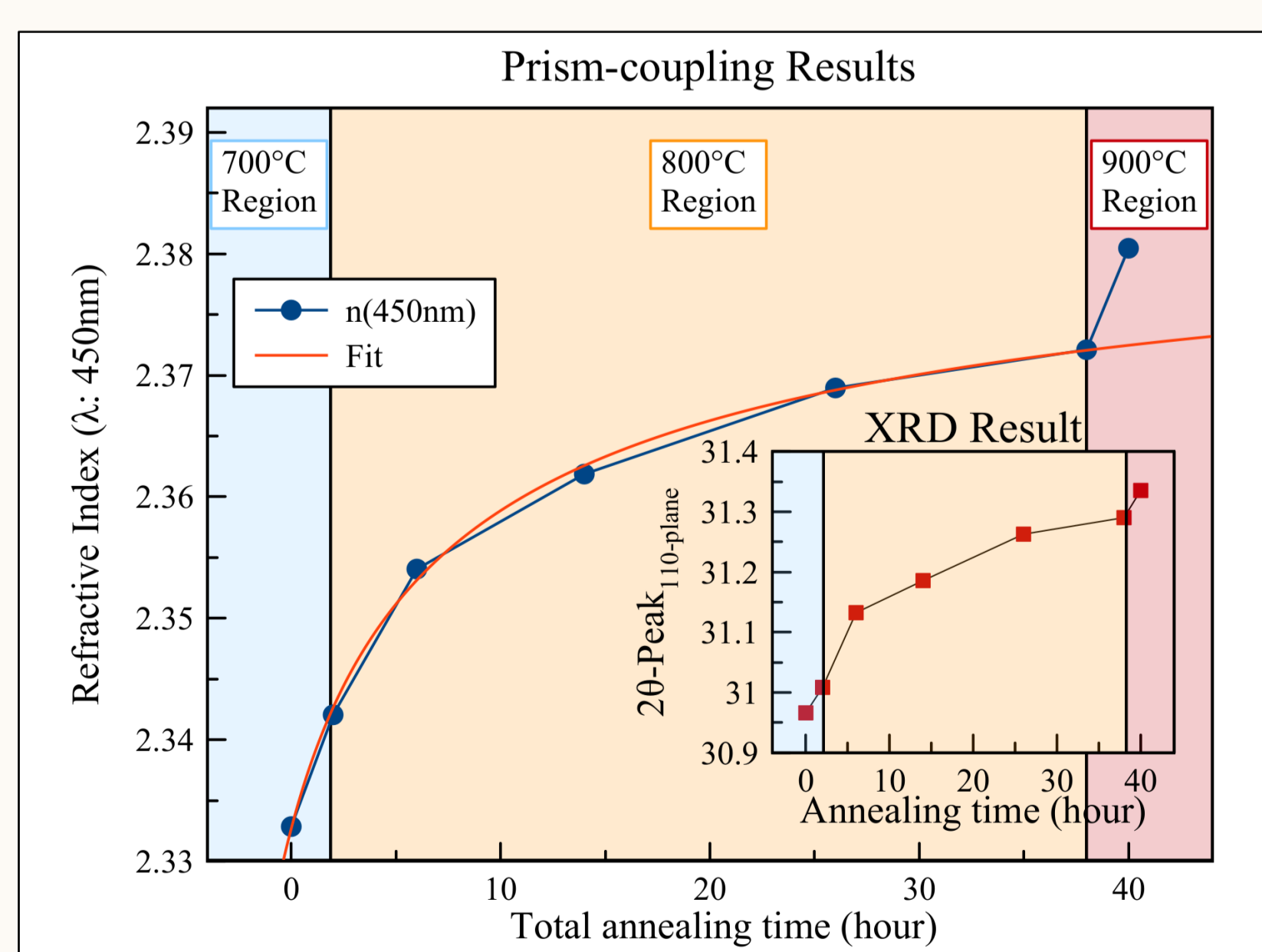
(a) Schematic diagram and (b) set-up image of prism-coupling system



(a) TE refraction modes of R.T.G. BCTZ film; $n(450\text{nm})$ increased from 1.957 to 2.150.
 (b) Comparison of TE modes between R.T.G. film and in-situ-growth (I.S.G) film; FWHM of I.S.G. film is significantly lower than R.T.G. samples. After post annealing treatment, $n(450\text{nm})$ of I.S.G. is enhanced up to 2.380.
 (c) Evolution of refractive index of BCTZ thin film via growth temperature along with one post-annealed I.S.G. sample.
 (d) Progressive effect of successive post annealing treatment on the refractive index under different wavelengths.



The progressive effects of successive post-annealing treatments on the refractive index and (inset) the 2θ-peak of BCTZ/Sapphire thin film deposited at 650°C.



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