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Fresnoite Thin Films grown by Pulsed Laser Deposition: Photoluminescence and Laser Crystallization

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Here we provide additional data on the crystalline BTS film structure on amorphous quartz substrates in as-grown and annealed state (Figure S1), on the evolution of surface microstructure, i.e. grain size and RMS roughness, due to laser direct scribing with changing mark speed (Figures S2 and S3, corresponding to Figure 7 (a)), SEM images of crystallized BTS surfaces with dendritic structures on Si substrates, and suppressed microcracks on sapphire substrates (Figure S4), and finally film composition depth profiles measured by Secondary Neutrals Mass Spectrometry for BTS on MgO and sapphire (Figure S5).

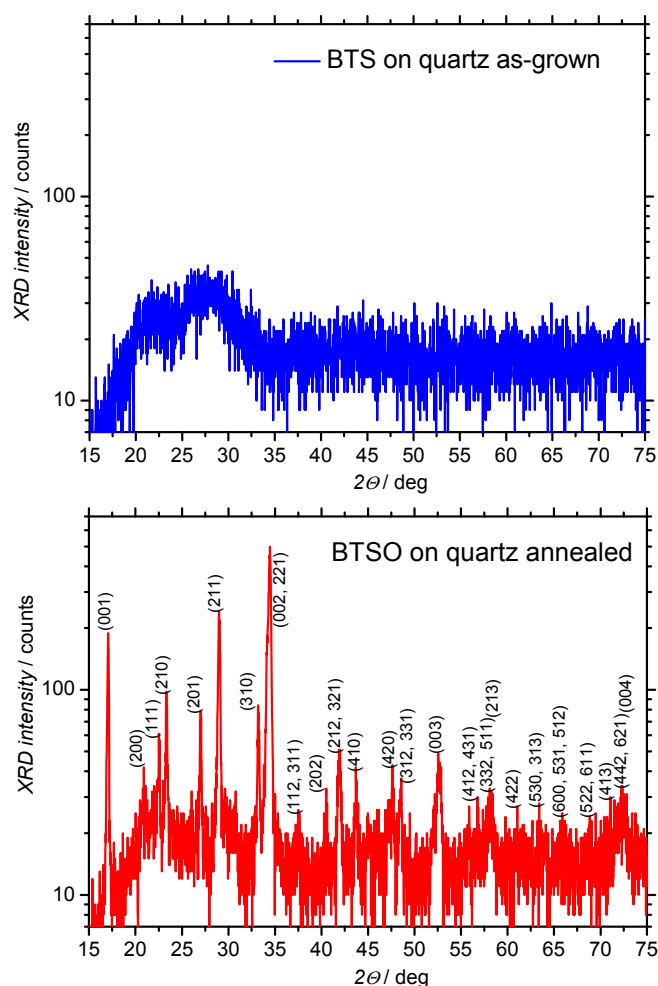


Figure S1. XRD 2θ-ω scans (Cu K α) of BTS thin films on amorphous quartz (SiO $_2$) substrate, in as-grown state (*top*), and after thermal annealing (*bottom*). In comparison to the films on single crystalline Si(100), MgO(100), and a-plane sapphire substrates (see Figure 2 (b), (c)), the evolution of (001) texture of the annealed film is weaker, see the intensity ratio of (001) and (211) peak. Note that the scattering background is clearly enhanced due to the amorphous substrate. The peak assignment is according to JCPDS (1998) No. 84-0924.

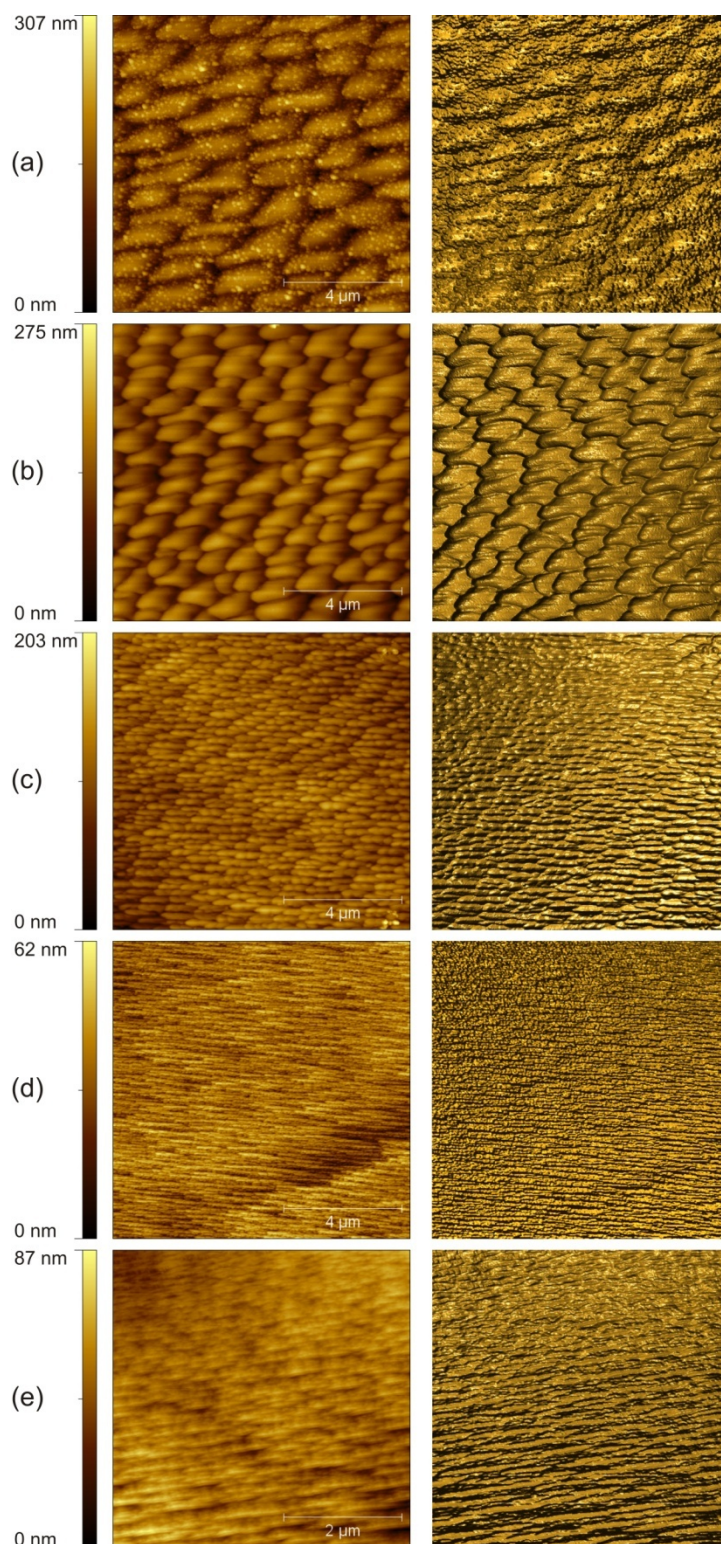


Figure S2. Surface morphology of laser-crystallized fresnoite films on three-inch diameter r-plane sapphire wafer is dependent on mark speed of the CO₂ laser writer, demonstrated by AFM images in 2D (*left column*), and 3D presentation (*right column*). For laser scribing, BTS on a large-area r-plane sapphire wafer was used because other substrates were not available with this size. However, the enhancement of luminescence is found to be similar.

The images correspond directly to the luminescence photograph of Figure 7 (a), to the line pattern of left column (10 W laser power, single pass, 2 to 128 mm s⁻¹ mark speed). The AFM images of (a), (b), (c), (d), and (e) correspond to the lines written with mark speeds of 4, 8, 16, 32, 64 mm s⁻¹, respectively. Obviously, the structure size decreases with increasing scan speed, see also the graphical evaluation in Figure S3.

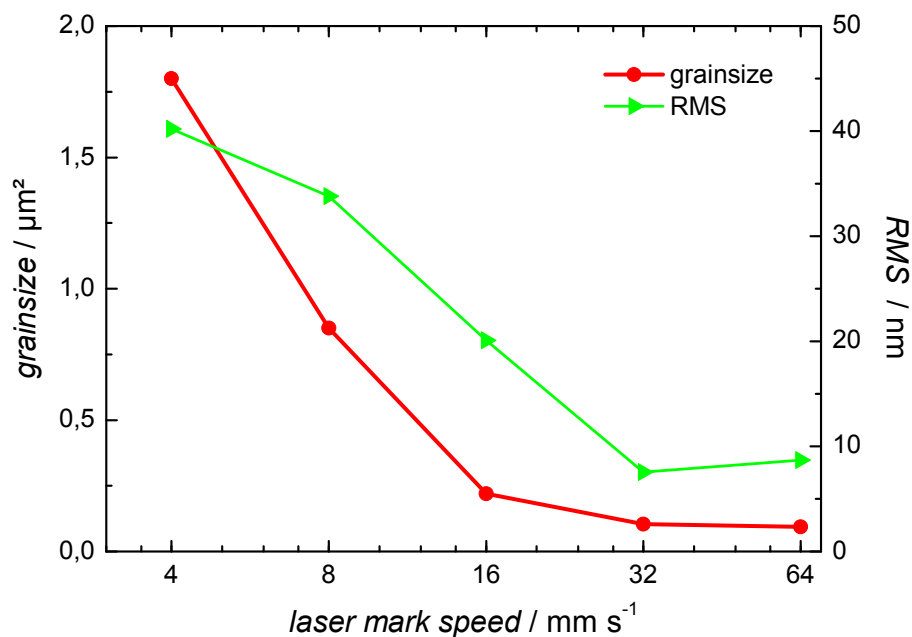


Figure S3. Grain size (*left scale*) and RMS roughness (*right*) of the crystallized fresnoite films on r-plane sapphire, as determined from the AFM images of Figure S2, and corresponding to Fig. 7 (a), the laser written line series of the left column (P = 10 W).

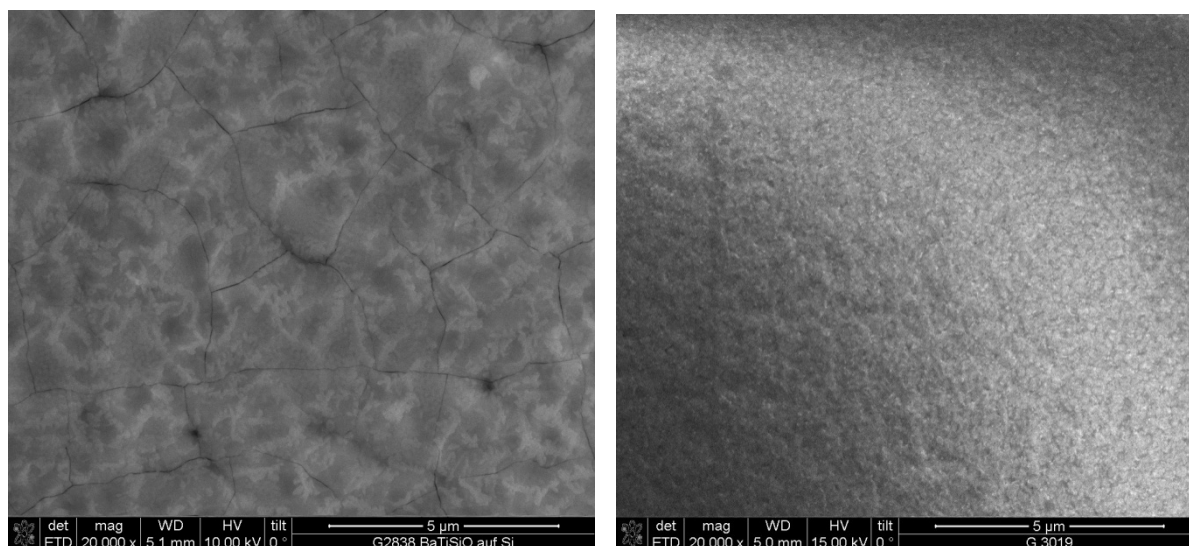


Figure S4. FE-SEM images of typical, crystallized thin BTS films on Si(100) (*left*), and on a-sapphire (*right*). The BTS films on Si exhibit a typical dendritic surface structure. The microcacks in the films visible in the left image can be avoided using sapphire substrates.

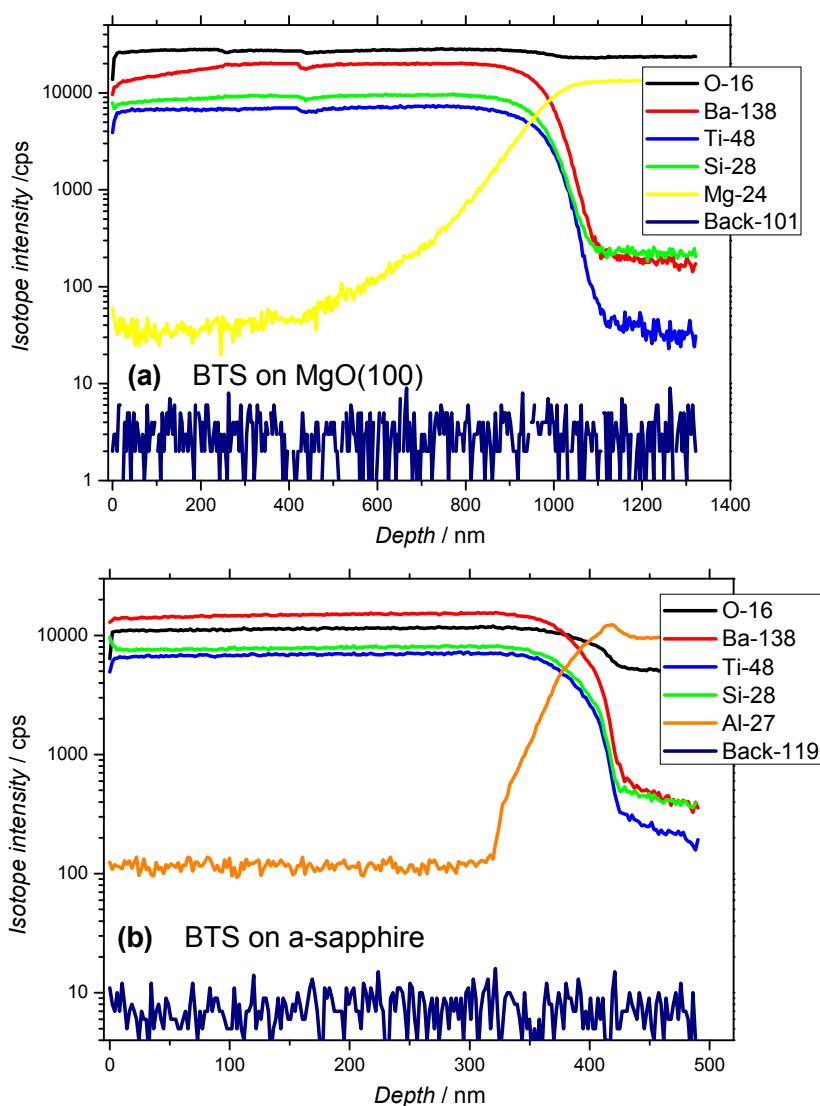


Figure S5. Profiles of isotope intensities vs. sputtered depth of annealed BTS films on (a) MgO(100), and (b) a-plane sapphire, as measured by Secondary Neutrals Mass Spectrometry (SNMS Leybold INA 3) with 750 eV Ar^+ ions. The sputter time scales were converted into depth scales using the final depth of the sputtering crater as determined by a surface profiler DEKTAK 3030, and with the assumption of a linear sputtering rate for BTS and substrate. The interface abruptness is superior for BTS on sapphire. For BTS on MgO, the Ba profile seems to show a weak concentration gradient near the film surface.