Supplemental Information: Tuning the Čerenkov Second Harmonic contrast from ferroelectric domain walls via anomalous dispersion

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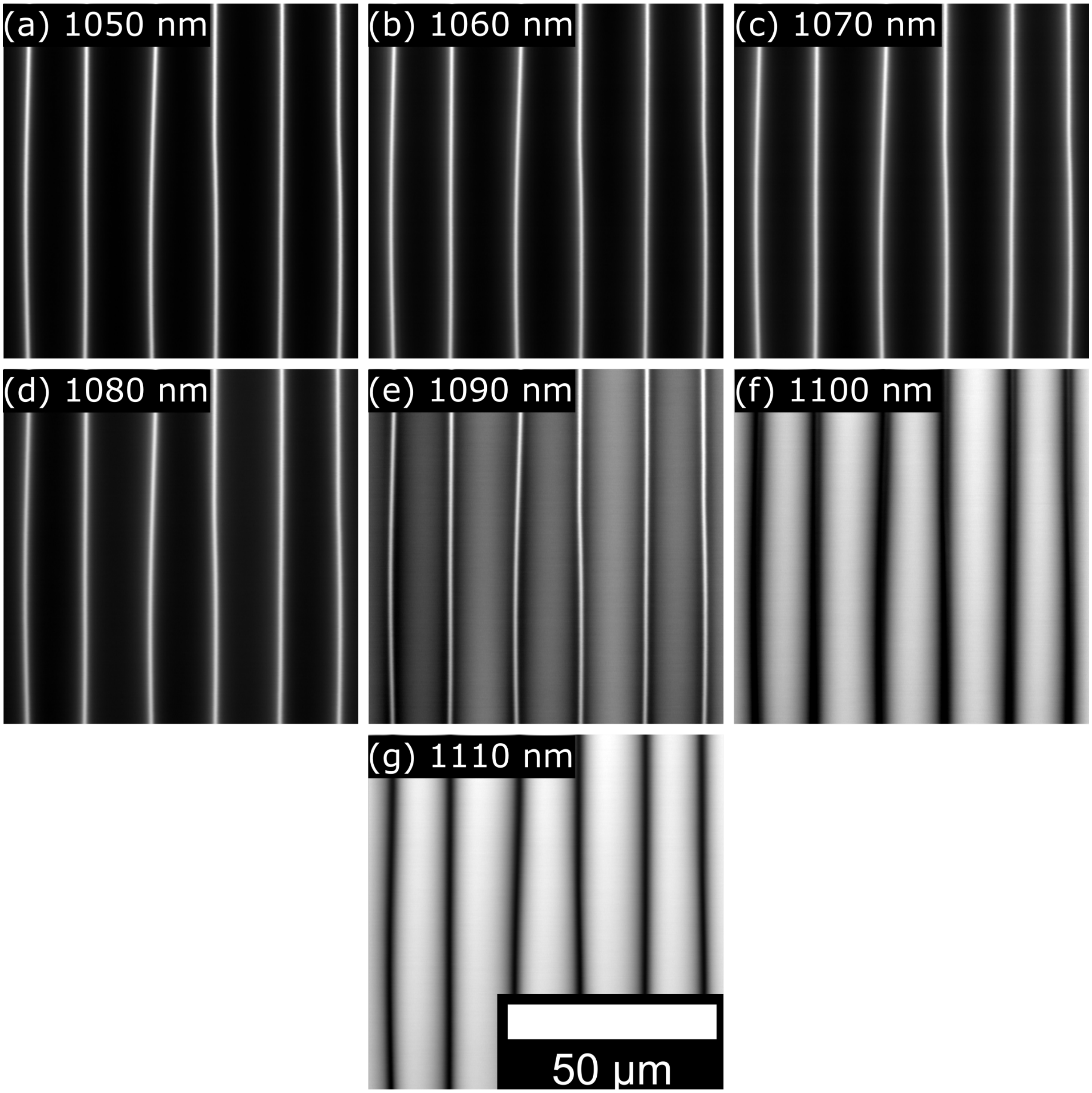

FIG. S1. Cross-section images from 500 μm within a periodically-poled congruent lithium niobate (cLN) crystal with increasing fundamental wavelength. Each image was recorded with the laser beam incident along the y axis and polarized parallel to the x axis. For the lower wavelengths, i.e. in the normal dispersion regime, the domain walls (DWs) only appear as bright lines on the dark background, as they give signal due to Čerenkov second harmonic generation (CSHG). As the incident wavelength increases, the magnitude of the DW signal decreases compared to the surrounding domain bulk as both, less of the exciting laser pulse is below the anomalous threshold, and also more signal is generated by the bulk material itself. For approximately 1090 nm, this results in the superposition of two contrasts visible in Fig. 3 of the main text. For even higher wavelengths, the CSHG contribution disappears and only bulk domain signal is detected, i.e. the DWs appear as dark lines on a bright background.

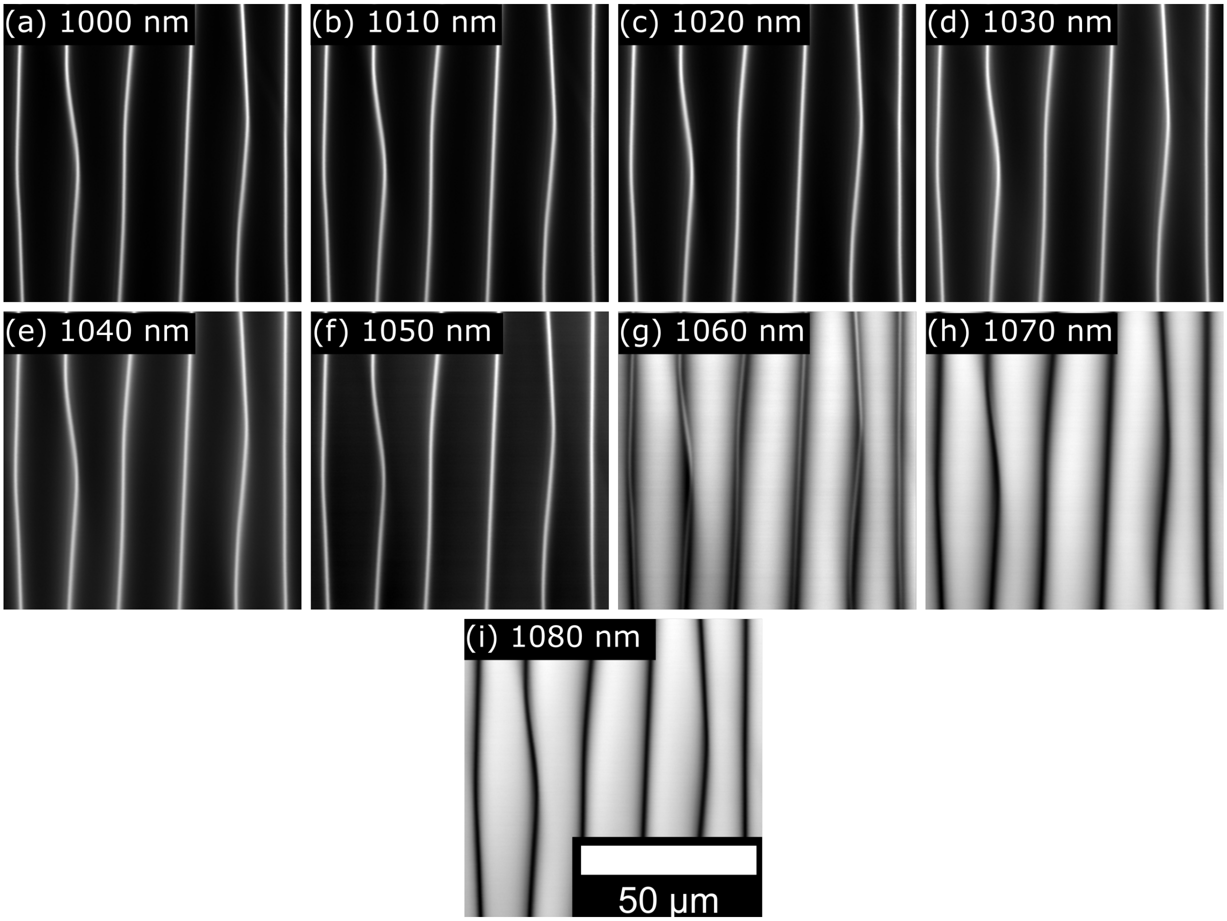

FIG. S2. Cross-section images from 500 μm within a periodically-poled (5%)-magnesium oxide-doped lithium niobate (MgO:LN) crystal with increasing fundamental wavelength. Each image was recorded with the laser beam incident along the y axis and polarized parallel to the x axis. For the lower wavelengths, i.e. in the normal dispersion regime, the domain walls (DWs) only appear as bright lines on the dark background, as they give signal due to Čerenkov second harmonic generation (CSHG). As the incident wavelength increases, the magnitude of the DW signal decreases compared to the surrounding domain bulk as both, less of the exciting laser pulse is below the anomalous threshold, and also more signal is generated by the bulk material itself, as is best visible in the image recorded at 1050 nm. For approximately 1060 nm, this results in the superposition of two contrasts visible in Fig. 3 of the main text. For even higher wavelengths, the CSHG contribution disappears and only bulk domain signal is detected, i.e. the DWs appear as dark lines on a bright background.

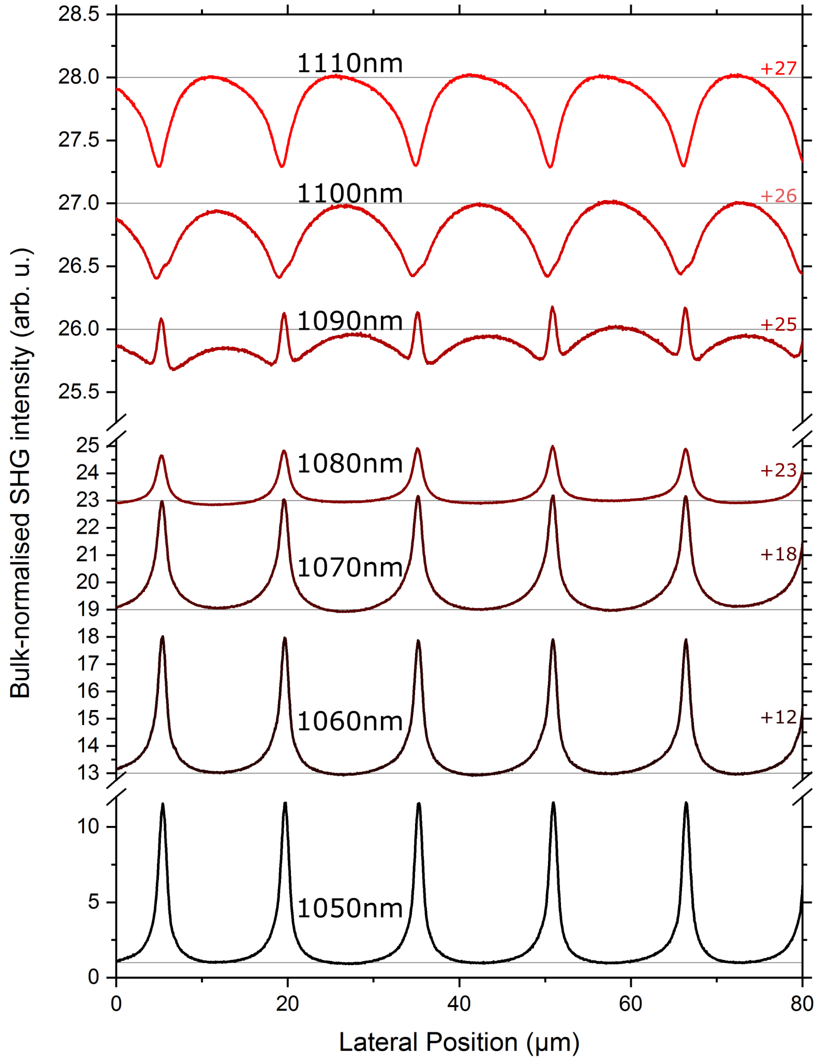

FIG. S3. Profiles extracted from a depth of 500 μm into a periodically-poled congruent lithium niobate (cLN) crystal with increasing fundamental wavelength. Each profile was recorded with the laser beam incident along the crystallographic y axis and polarized parallel to the x axis. In comparison to Fig. 3 in the main text, each profile was normalized to its respective bulk domain signal level, allowing for a better comparison of the difference in relative peak magnitudes. Additionally, the profiles were shifted vertically in order to group any profiles that have a similar magnitude, with the shift indicated by the number listed on the right of each profile. Below the expected threshold wavelength of 1078 nm, we observe the domain walls (DWs) as positive peaks upon the dark background signal from the domains (black colored profile). The cause of this DW signal is Čerenkov second harmonic generation (CSHG), as a majority of the spectrum of the incident laser pulse still lies below the threshold. As the wavelength increases, the height of the DW peaks compared to the domains decreases, as less CSHG is generated and the domains themselves begin to generate appreciable SH signal. At a fundamental wavelength of 1090 nm, this results in a superposition of both a positive as well as a negative peak at the location of the domain wall, showing the simultaneous existence of multiple contrast mechanisms within a generated SH signal, which is usually obscured due to the far larger strength of the CSHG mechanism. At the highest investigated wavelengths, we observe that the majority of the signal stems from the domains, with the DWs appearing as dips in the signal (red colored profile).

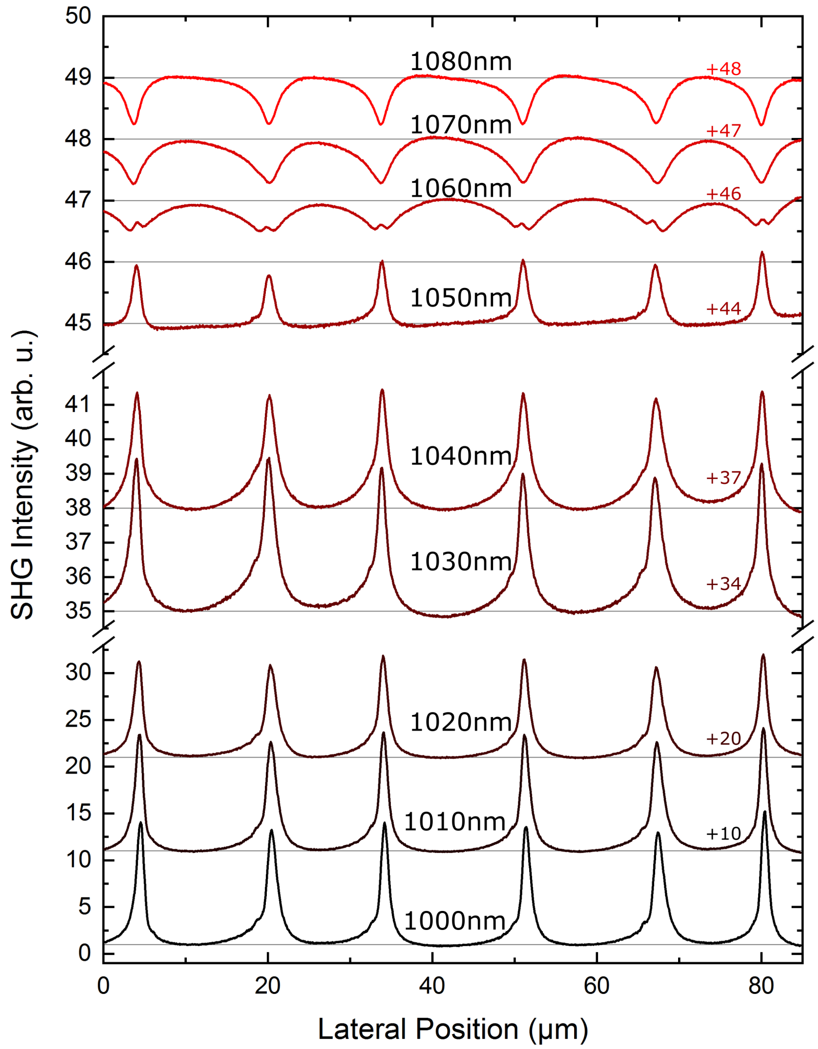

FIG. S4. Profiles extracted from a depth of 500 μm into a periodically-poled (5%)-magnesium oxide-doped lithium niobate (MgO:LN) crystal with increasing fundamental wavelength. Each profile was recorded with the laser beam incident along the crystallographic y axis and polarized parallel to the x axis. In comparison to Fig. 3 in the main text, each profile was normalized to its respective bulk domain signal level, allowing for a better comparison of the difference in relative peak magnitudes. Additionally, the profiles were shifted vertically in order to group any profiles that have a similar magnitude, with the shift indicated by the number listed on the right of each profile.Below the expected threshold wavelength of 1034 nm, we observe the domain walls (DWs) as positive peaks upon the dark background signal from the domains (black colored profile). The cause of this DW signal is Čerenkov second harmonic generation (CSHG), as a majority of the spectrum of the incident laser pulse still lies below the threshold. As the wavelength increases, the height of the DW peaks compared to the domains decreases, as less CSHG is generated and the domains themselves begin to generate appreciable SH signal. At a fundamental wavelength of 1060 nm, this results in a superposition of both a positive as well as a negative peak at the location of the domain wall, showing the simultaneous existence of multiple contrast mechanisms within a generated SH signal, which is usually obscured due to the far larger strength of the CSHG mechanism. At the highest investigated wavelengths, we observe that the majority of the signal stems from the domains, with the DWs appearing as dips in the signal (red colored profile).

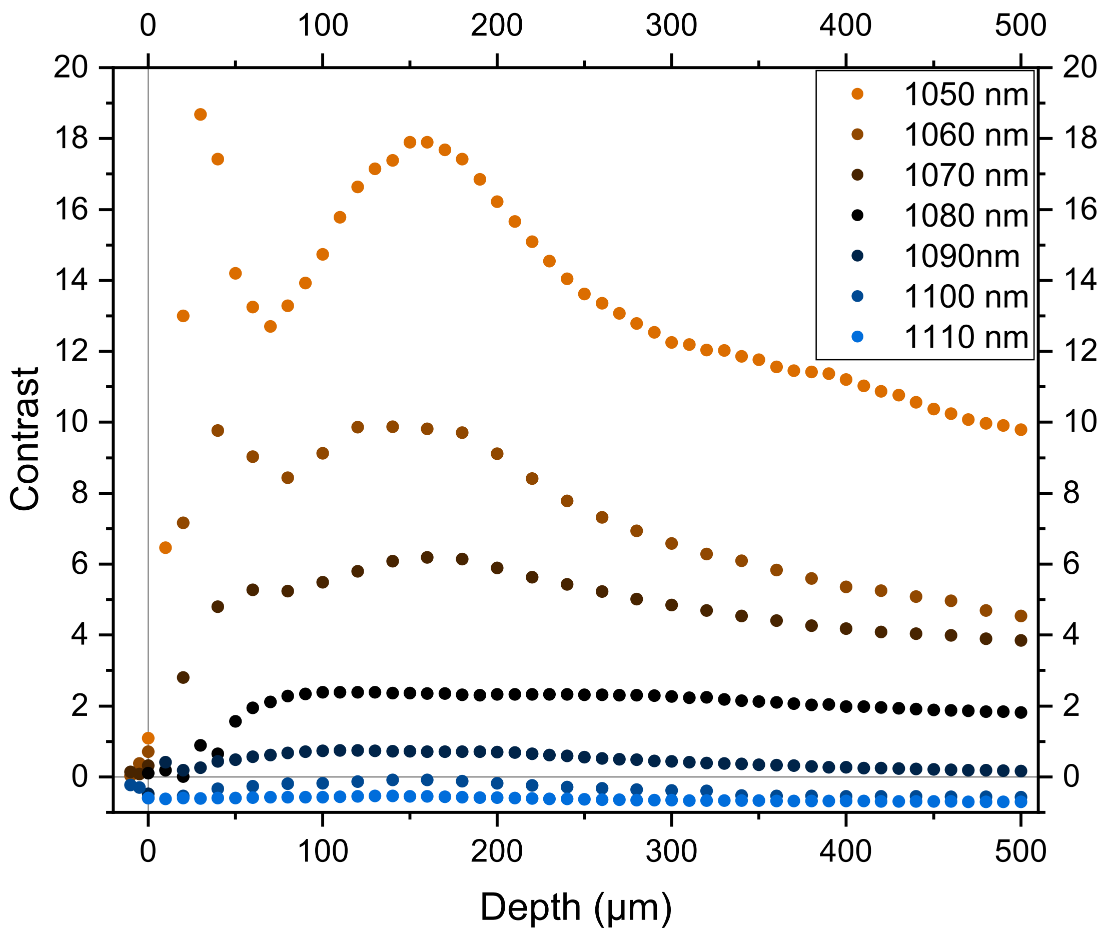

FIG. S5. Contrast values for all measured wavelengths plotted as a function of depth into the crystal. For values above, as well as below the threshold of approximately 1090 nm one contrast, either positive or negative, respectively, can be observed. In contrast, the values for 1090 nm and 1100 nm display a slight superposition of both.