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**Detecting and characterizing ferroelectric domain boundaries using nonphase-
matched second harmonic generation***

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We demonstrate that tilted planar ferroelectric domain boundaries create tilted second harmonic beams in nonphase-matched second harmonic generation, and that the tilt of the boundary can be deduced from the harmonic direction.

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Detecting and characterizing ferroelectric domain boundaries using nonphase-matched second harmonic generation

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Ferroelectric crystals such as KTP and its isomorphs, KNbO_3 , LiNbO_3 , and LiTaO_3 , are important nonlinear optical crystals. The detection and control of their ferroelectric domains is important for quality control to assure single domain crystals, and in poling crystals for quasiphase matching. A number of methods have been used to characterize domains¹⁻⁵. Most are based on examination of the crystal surfaces and so are not particularly useful in characterizing crystal interiors. Only electro-optic techniques and phase-matched harmonic generation have been used to determine whether domains extend through the crystal. The phase-matched harmonic method is difficult to apply to a wide range of crystals because the fundamental wavelength must be tuned to a specific wavelength for each crystal type. The electro-optic method requires that electrodes be placed on the crystal, and it is less sensitive than required for some applications.

We have developed a new method that uses nonphase-matched second harmonic generation. A suitable Q-switched laser capable of producing about 10 mJ in pulses of a

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few nanosecond duration can be used to characterize any crystal transparent to the fundamental and second harmonic light. In a crystal with parallel input and output faces, a domain boundary anywhere in the light path that is tilted by angle γ relative to the input face will create a second harmonic beam tilted by angle $(\Delta k/2k_0 \tan \gamma)$ relative to the

forward direction, where k_0 is the vacuum wavevector of the fundamental and Δk is the phase velocity mismatch between the

fundamental and harmonic light. This tilt is typically a few mrad so it is easily distinguished from the forward harmonic light also generated by the crystal, and its irradiance is comparable to that of the forward beam. Using a setup like that diagrammed above, we have imaged the harmonic beams from several KTP crystals and found tilted domains in four of them. An example of the beam pattern is shown below. The leftmost

beam is the forward beam while the three tilted beams are due to a single domain boundary. There are three because there are three allowed nonphase-matched processes with different values of Δk . The ratio of beam deflection angles matches the

ratio of the Δk 's for the three processes as expected. We confirmed the existence of multiple domains in the four crystals using phase matched second harmonic generation.

This new technique should be useful in quality control screening of ferroelectric crystals for domain problems because it is easily implemented and versatile. A relatively low power Q-switched Nd:YAG laser could be used to screen all KTP family crystals plus KNbO_3 , and LiNbO_3 . Further, it is possible to map the location and tilt of each domain boundary so detailed studies of domain behavior could be easily implemented.

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