

# Quasi-phase-matched second-harmonic generation of 532 nm radiation in 25°-rotated, x-cut, near-stoichiometric, lithium tantalate fabricated by vapor transport equilibration

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Quasi-phase-matched second-harmonic generation of 532 nm radiation in 25°-rotated, x-cut, near-stoichiometric lithium tantalate has been performed. Using a face-normal topology for frequency conversion applications allows scalable surface area to avoid surface and volume damage in high-power interactions. First-order, quasi-phase-matched second-harmonic generation was achieved using near-stoichiometric lithium tantalate fabricated by vapor transport equilibration. These crystals supported 1 J of 1064 nm radiation and generated 21 mJ of 532 nm radiation from a 7 ns, Q-switched Nd:YAG laser within a factor of 4.2 of expectation. © 2007 Optical Society of America  
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Quasi-phase-matched (QPM) nonlinear optical devices are advantageous for many frequency conversion applications, since they provide increased engineerability and efficiency compared with their birefringently phase-matched counterparts. Ferroelectrics can be used as QPM nonlinear media by periodic poling (PP). Periodically poled lithium tantalate is a common, engineerable, and efficient periodically poled ferroelectric for frequency conversion in the near-infrared and the visible. However, commercially available, congruent-composition lithium tantalate (CLT) suffers from photorefractive damage (PRD) and green-induced infrared absorption (GRIIRA).<sup>1,2</sup> Recently, the sensitivity of lithium tantalate to PRD and GRIIRA has been correlated with the native defects formed due to the nonstoichiometry of congruently melting crystals. Near-stoichiometric lithium tantalate (SLT) has been produced by various means<sup>3,4</sup> and is two orders of magnitude less sensitive to PRD and GRIIRA than CLT. By avoiding PRD and GRIIRA using appropriate composition control, practical QPM devices are possible.

The thickness of congruently melting periodically poled lithium tantalate crystals is limited to approximately 1 mm because of the high applied electric field required for the PP process. Without elliptical focusing, the thickness limits the available aperture to an area of approximately 1 mm<sup>2</sup>. Previous efforts to increase the useful aperture of periodically poled ferroelectrics included PP thick substrates (~5 mm) of lower coercive field ferroelectrics such as magnesium-oxide-doped lithium niobate<sup>5</sup> and diffusion bonding unpoled lithium niobate to periodically poled lithium niobate.<sup>6</sup> Previously, we proposed a new topology that allows for scaling the useful aperture up to the diameter of available substrates (typically 7.62 to 10.16 cm).<sup>7</sup> In the remainder of this Letter, we discuss this topology and its realization in periodically poled SLT.

In Ref. 7, we proposed a topology for scalable crystal aperture based on periodic poling of a rotated-cut ferroelectric. This topology is depicted in Fig. 1a. Light incident on the large wafer surface can be polarized with a component along the extraordinary axis to use the largest nonlinear coefficient and make use of a geometrical projection of the propagation vector onto the QPM grating  $k$ -vector. Previous results of PP of rotated-cut substrates include those for surface acoustic wave applications<sup>8</sup> and our previously published result.<sup>7</sup> For nonlinear optics purposes, we have chosen to develop PP of 25°-rotated, x-cut, near-stoichiometric lithium tantalate fabricated by vapor transport equilibration (VTE). We shall discuss the choice for each characteristic briefly.

The 25° angle of rotation allows light incident near Brewster's angle to propagate normal to domain walls containing the  $z$ -axis and hence allows collinear interactions taking advantage of the largest component of the nonlinear susceptibility tensor. A second-harmonic generation (SHG) interaction is shown in Fig. 1b.

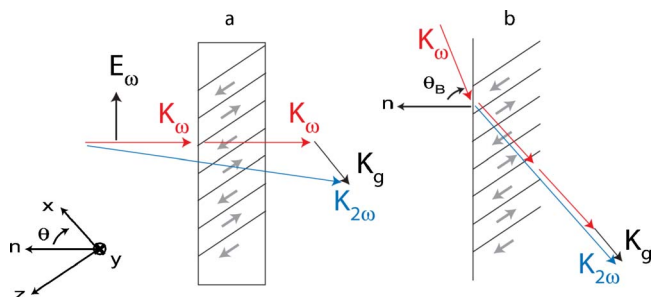


Fig. 1. (Color online) a, Topology of large aperture periodically poled crystals. Light propagates through the large wafer face and has a component of the electric field polarized along the  $z$ -axis, enabling efficient SHG among other nonlinear interactions. b, For 25°-rotated ( $\theta=25^\circ$ ), x-cut substrates, light incident at near Brewster's angle,  $\theta_B$ , allows a collinear SHG interaction.

The cut of the crystal indicates that the alternate ordinary axis is in the plane of the wafer. We have chosen to force the  $y$ -axis to be in-plane, since  $+y$  and  $-y$  domains have a differential etch rate. This differential etch rate permits us to visualize domains from the side, after PP.

SLT avoids the aforementioned PRD and GRIIRA. Most commercially available lithium tantalate is congruently melting and contains 48.39 mol.% lithium oxide ( $\text{Li}_2\text{O}$ ), whereas crystals of near-stoichiometric composition have compositions close to 49.95 mol.%  $\text{Li}_2\text{O}$ . The lithium deficiency of CLT causes native defects that are widely accepted to be one  $\text{Ta}^{4+}$  antisite defect ( $\text{Ta}_{\text{Li}^{5+}}^{4+}$ ) and four lithium vacancies.<sup>9</sup> The concentration of native  $\text{Ta}^{4+}$  antisite defects closely correlates with the magnitude of PRD and GRIIRA<sup>10</sup> with the magnitude of the ferroelectric coercive field.<sup>11</sup> Thus, SLT has reduced sensitivity to PRD and GRIIRA and has a lower coercive field. We have chosen to produce our SLT by the VTE method.<sup>4</sup> The VTE method produces crystals with a coercive field as low as 100 V/mm, consistent with a composition of 49.99 mol.%  $\text{Li}_2\text{O}$ .<sup>11</sup>

The original substrates for the VTE process were surface-acoustic-wave-grade, 50 mm diameter, 1 mm thick, 25°-rotated, x-cut CLT wafers from Sawyer Technical Materials, LLC. The VTE process was carried out at 1360°C for 120 h. After the VTE processing, it was necessary to uniformly pole the VLT crystals before reproducible PP was possible. The uniform poling process was performed at 185°C with graphite electrodes and applied fields as high as 1000 V/mm to remove the head-to-head domains that result from the VTE process. To ensure complete polarization, the applied field was reversed several times. Afterwards, the crystals were annealed in air at 620°C for 10 h.

The “apparent” coercive field (the applied voltage divided by the wafer thickness) and spontaneous polarization (the charge per unit area transferred in tracing a complete hysteresis loop) at room temperature were 315 V/mm and 27  $\mu\text{C}/\text{cm}^2$ , respectively. For rotated-cut materials, the apparent coercive field and the spontaneous polarization are the conventional z-cut coercive field and the spontaneous polarization scaled by the geometrical projection of the surface normal onto the  $z$ -axis of the crystal, respectively. Projecting the coercive field and the spontaneous polarization onto the direction of the ferroelectric axis yields 133 V/mm and 63  $\mu\text{C}/\text{cm}^2$ , respectively. These values are consistent with those for similarly processed z-cut substrates.

Before PP, the crystals were ground and polished on both sides to a final dimension of 0.5 mm in thickness. PP was carried out at 120°C with aluminum electrodes evaporated over a photoresist pattern. The reduced thickness and elevated PP temperature served to increase nucleation and improve overall domain quality. Poling was performed with a single poling pulse for a duration of 15 ms at 540 V/mm at a period of 8  $\mu\text{m}$  for first-order QPM SHG of 532 nm radiation. Samples were then polished along the  $+y$

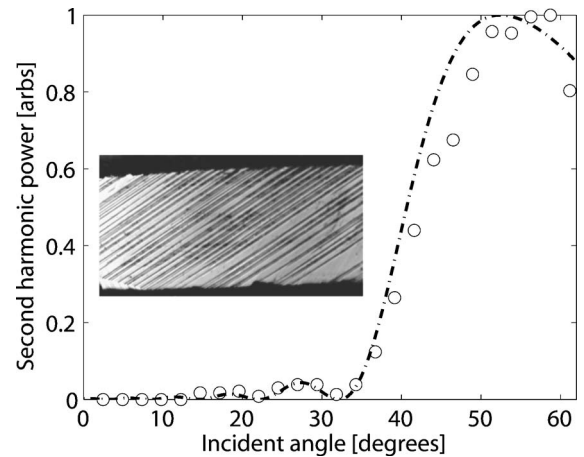


Fig. 2. Angular tuning curve of SHG power (in arbitrary units) as a function of incident fundamental angle. The dotted-dashed curve represents the theoretical angular tuning curve and the circles represent measured data. Inset, photograph of 25°-rotated domains ( $+y$  surface) revealed by chemical etching in hydrofluoric acid. The domain period is 8  $\mu\text{m}$ .

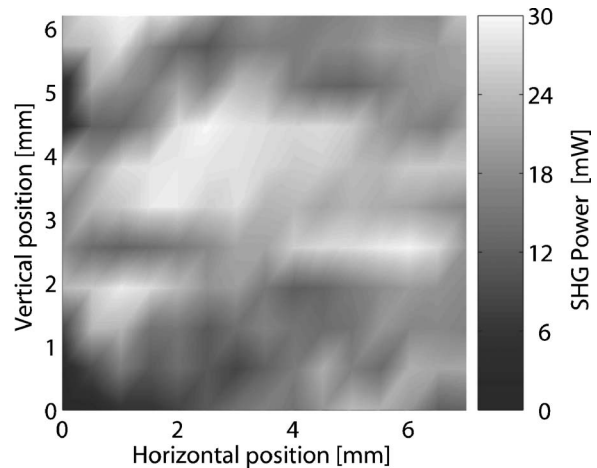


Fig. 3. Spatial map of the generated 532 nm power as a function of position.

surface and chemically etched in hydrofluoric acid for 20 min to reveal domain patterns. A photograph of a chemically etched sample is shown in the inset of Fig. 2. After PP and etching, the aluminum electrodes and photoresist were removed in preparation for SHG measurements.

To examine the local efficiency and the high-energy operation of the substrates two separate SHG measurements were performed. A mode-locked, 9.2 W average-power, 7 ps, 50 MHz repetition-rate, 1064 nm laser was loosely focused to a 135  $\mu\text{m}$  spot ( $1/e^2$  intensity radius, excluding foreshortening due to nonnormal incidence on the sample). An angle tuning curve was measured and fits well with the theoretical calculations. The generated SHG power was then measured as a function of position over a 6 mm by 7 mm area. The results from this experiment are shown in Fig. 3. The maximum SHG power generated was 30 mW and is a factor of 3.5 below theoretical for this loose focus (assuming  $d_{\text{eff}}=11$  pm/V).<sup>12</sup> The discrepancy can be explained by poling errors (mostly merged domains) and domains that did not

propagate the full thickness of the wafer. The laser was then focused to a  $30\ \mu\text{m}$  spot ( $1/e^2$  intensity radius, excluding the foreshortening) and generated a maximum of 540 mW of 532 nm radiation. The experimental conversion efficiency is again a factor of 3.5 below theoretical. Although the uniformity of the sample was poor, further improvements in PP will enable scaling of the usable area for future devices.

To examine the high-energy capacity of a rotated-cut sample, we focused a 1 J/pulse, 7 ns, Q-switched, 1064 nm laser on a  $0.26\ \text{cm}^2$  area of the crystal (including the foreshortening). The damage threshold in z-cut near-stoichiometric tantalate fabricated by VTE was measured to be  $7.3\ \text{J}/\text{cm}^2$  for 12 ns pulses. At the input energy of 1 J, no laser-induced damage was observed and the SHG energy was measured to be 21 mJ, a factor of 4.2 below theoretical.

For a given fluence,  $F$ , the required thickness for an ideally poled sample to reach 50% conversion,  $t_{50\%}$ , is given by

$$t_{50\%} \approx \sqrt{\frac{n_1^2 n_2^2 \tau}{\eta_0^3 \omega_1^2 \epsilon_0^2 d_{\text{eff}}^2 F}} \tanh^{-1}(\sqrt{0.5}) \cos(25^\circ), \quad (1)$$

where  $n_i$  is the refractive index at the first and second harmonic,  $\eta_0$  is the impedance of free space,  $\omega_1$  is the fundamental frequency,  $\epsilon_0$  is the permittivity of free space, and  $\tau$  is pulse width. This equation assumes a flat-top intensity profile in space and time. With a measured damage fluence,  $F_{\text{max}}$ , of  $7.3\ \text{J}/\text{cm}^2$  for 12 ns pulses and a  $\sqrt{\tau}$  scaling of the damage fluence, we expect that for 1-mm-thick crystals, SHG of 532 nm radiation with 50% conversion will be possible for pulse widths less than or equal to 16 ns constraining  $F \leq \frac{1}{2} F_{\text{max}}$  for a flat-top intensity profile in space and time. At  $F = \frac{1}{2} F_{\text{max}}$ , the 50% conversion thickness as a function of pulse length is given by

$$t_{50\%} \approx 0.93 \sqrt[4]{\frac{\tau}{12}}, \quad (2)$$

where  $t_{50\%}$  has units of mm and  $\tau$  has units of ns.

We have presented the realization of a large-area, scalable QPM nonlinear device based on  $25^\circ$ -rotated, x-cut, near-stoichiometric lithium tantalate fabricated by VTE. The device operates at near Brewster's angle, has an efficiency nearly 30% of the theoretical value in the ps regime, and has been shown to support 1 J of pulse energy at 1064 nm. With further improvement of the PP quality and the use of optical-quality crystals, we believe that 50% efficient conversion is possible out to a pulse width of 16 ns for a flat-top intensity profile in space and time.

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