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69% Efficient continuous-wave second-harmonic generation in lithium-rich lithium niobate

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Lithium-rich lithium niobate fabricated by vapor transport equilibration is used to frequency double the output of an injection-locked Nd:YAG laser. Internal doubling efficiencies as high as 69% and powers of as much as 1.6 W are achieved by resonant external cavity second-harmonic generation. No evidence of photorefractive damage is observed at the operating temperature of 234°C.

Efficient nonlinear frequency conversion of optical radiation requires crystals with extremely low losses and large nonlinear coefficients. Conversion efficiencies exceeding 50% have been obtained by using monolithic magnesium-doped lithium niobate crystal cavities that resonantly enhance the fundamental laser field.[1,2] While lithium niobate crystals grown from melts of the congruent composition have good optical quality and uniform birefringence, they suffer from photorefractive damage[3] when exposed to visible radiation, a problem that is substantially alleviated in MgO-doped crystals.[4] Unfortunately, optical homogeneity problems owing to incongruent incorporation of the MgO are difficult to avoid.[5] In this Letter we describe the generation of as much as 1.6 W of power of cw 532-nm radiation in lithium-rich lithium niobate produced by a vapor transport equilibration (VTE) technique.

In the VTE technique,[6] crystals are annealed in close proximity to a large mass of two-phase powder with a net composition on the lithium-rich side of the $LiNbO_3$–Li_3NbO_4 phase boundary. Given sufficient time, the crystals equilibrate to the phase-boundary composition. The VTE technique produces lithium-rich lithium niobate with excellent homogeneity, which makes feasible the use of crystals several centimeters long[7] that promises efficient doubling at relatively low powers. The phase-matching temperature for doubling 1064-nm radiation is 233.7°C in crystals processed at 1100°C.[8]

Samples were cut from a 2-mm-thick y-cut wafer of the congruent composition and processed at 1100°C for 800 h to the lithium-rich phase boundary by the VTE method.[8] The x faces were antireflection coated for 1064 nm. A crystal with dimensions of 2 mm × 4 mm × 10 mm was placed into an external bow-tie ring cavity[9] with a 32-μm beam waist and input coupler transmission of $t_{IC} = 4.2\%$ at $\lambda = 1064$ nm. The cavity was piezo locked with the Pound–Drever FM technique to an injection-locked, lamp-pumped Nd:YAG laser.[10] The round-trip loss of the cavity at 1064 nm was 1.37%, and 75% of the input laser power was matched to the fundamental transverse mode of the cavity. The generated green power was measured after it passed through the 90% transmitting (at 532 nm) folding mirror.

Figure 1 shows the generated green output power versus the power at 1064 nm incident upon the cavity. The highest green power, 1.6 W, was obtained with 4.7-W incident power, while the maximal overall conversion efficiency, 47%, was achieved with 2.9-W incident power. The potential performance of the doubler is best described by the internal efficiency, which gives the generated power inside the cavity as a percentage of the input power mode matched to the cavity. As shown in Fig. 2, the highest internal efficiency achieved is 69%, which is obtained at 2.2 W of mode-matched fundamental power. The solid curve shown in Fig. 2 was calculated[1] by using the cavity parameters given above and $d_{31} = 4.7$ pm/V.[11]

While the agreement between measurement and theory is excellent for low input powers, the measured efficiencies for higher powers fall below the theoretical values. This drop in conversion efficiency coincided with a degradation of the quality of the TEM_{00} output, probably as a result of thermal gradients in the crystal. The high-power intensity profile was closer to a top-hat profile than to the ideal Gaussian profile. Thermal effects also made it increasingly difficult to lock the cavity to the laser frequency at input powers greater than 2 W. The required oven temperature for peak conversion decreased with circulating power, consistent with crystal heating caused by absorbed power. The dependence was linear with a slope of 70 mK/W at circulating powers of less than 20 W, where the cavity could be well locked to the laser.

The FWHM of the temperature tuning curve for the 1-cm-long crystal for an input power of 27 mW at 1064 nm was 0.52 K compared with a theoretical value of 0.56 K,[12] which confirms the homogeneity of

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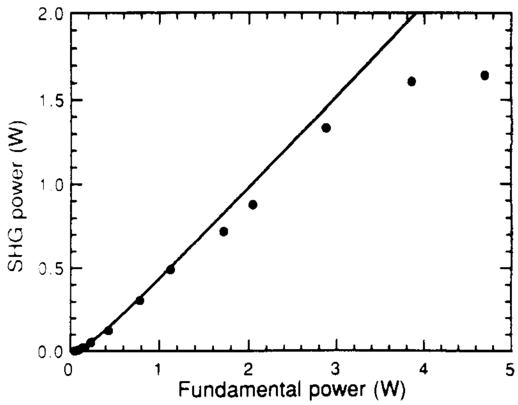

Fig. 1. Generated 532-nm power versus 1064-nm power incident upon an external resonant cavity containing a 1-cm-long lithium-rich lithium niobate crystal. SHG, second-harmonic generation.

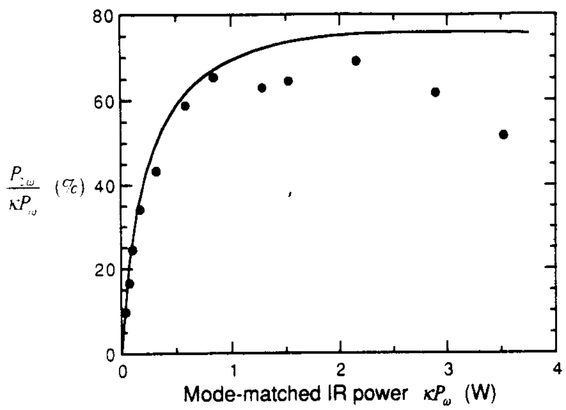

Fig. 2. Internal second-harmonic generation efficiency. The circles are measured data points, and the solid curve indicates the theoretical efficiency.

the sample. The shape of the temperature tuning curve became increasingly asymmetric, as shown in Fig. 3, as the input power was increased. At fundamental powers greater than 0.5 W, this led to a bistable output power as a function of oven temperature. To obtain maximal second-harmonic output power, the peak conversion had to be approached from oven temperatures that were lower than optimal for the applied fundamental power. To understand the temperature tuning behavior of the doubler, heating effects caused by absorption of optical power need to be considered. The circulating fundamental power P_c is related to the cavity parameters by[1]

$$P_c = \kappa \frac{t_{IC} P_\omega}{[1 - \sqrt{(1 - t_{IC}) t_c}\,]^2}, \quad (1)$$

where P_ω is the incident laser power, κ is the mode-matching efficiency, t_{IC} is the transmission of the input coupler at the fundamental wavelength, and t_c is the round-trip transmission factor of the cavity. The temperature of the crystal volume where the beams travel is higher than the oven temperature by the ratio of heat deposited over the thermal resistance K. In a one-dimensional approximation,

$$T_{crystal} = T_{oven} + \left[\alpha_\omega l P_c + \int_0^l \alpha_{2\omega} P_{2\omega}(z) dz\right] \Big/ K, \quad (2)$$

where l is the crystal length and α_ω and $\alpha_{2\omega}$ are the absorption coefficients for the fundamental and second harmonic, respectively. The second-harmonic power generated, including the effects of thermally induced phase mismatch, is given[13] by $P_{2\omega}(l)$, where

$$P_{2\omega}(z) = \Gamma(z) P_c^2,$$
$$\Gamma(z) = \Gamma_0 \operatorname{sinc}^2\left[\left(\pi \frac{T_{crystal} - T_{pm}}{\Delta T}\right)\left(\frac{z}{l}\right)^2\right]. \quad (3)$$

The function sinc(x) is defined as $\sin(x)/(x)$, and ΔT is the temperature difference between the phase-matching peak T_{pm} and the first zero, 0.59 K. The nonlinear conversion factor Γ_0 is calculated from the focusing parameter and the nonlinear coefficient.[1] The round-trip transmission factor t_c at the fundamental wavelength is

$$t_c = (1 - r_m)(1 - \Gamma P_c), \quad (4)$$

where $r_m = 1.37\%$ is the loss due to spurious reflections, absorption, and cavity misalignment. The term ΓP_c describes the pump depletion loss resulting from the SHG process.

The temperature tuning curve $P_{2\omega}(T_{oven}, P_\omega)$ is obtained by employing a numerical root-finding routine on Eqs. (1)–(4). The asymmetry of the tuning curve caused by fundamental absorption has the opposite sign from the one caused by second-harmonic absorption. Figure 3 shows the experimental tuning curve (circles) for an input power P_ω = 780 mW. The solid curve has been calculated by assuming no absorption at the second-harmonic wavelength and the value $\alpha_\omega l/K$ = 70 mK/W for the fundamental wavelength absorption as suggested by the measured oven temperature for peak efficiency. The agreement is good, which confirms the assumption that the temperature rise owing to second-harmonic absorption is negligible. The dominance of fundamental wave absorption is not surprising since the circulating fundamental power is many times larger than the second-harmonic power. On the peak of the tuning curve of Fig. 3, the heat deposited owing to the fundamental absorption is $\alpha_\omega l P_c = (16\ \text{W})\alpha_\omega l$,

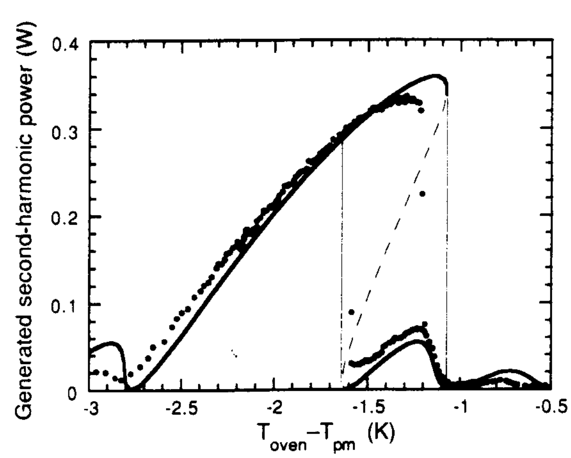

Fig. 3. Output power versus oven temperature showing asymmetry caused by heating by absorbed optical power in a 1-cm-long sample with 780 mW of fundamental power incident upon the external resonant cavity. At low input powers, the peak conversion occurs at T_{pm} = 234°C.

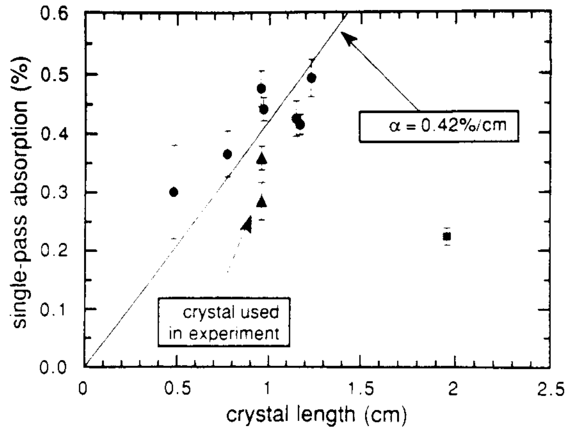

Fig. 4. Measured absorption losses in lithium-rich VTE samples at the ordinary polarization for a wavelength of 1064 nm. The circles and the square are for samples fabricated from two *z*-cut wafers A and B, respectively. The triangles represent crystals made from a *y*-cut wafer.

whereas it is $\alpha_{2\omega}l/3P_{2\omega} = (0.12\ \text{W})\alpha_{2\omega}l$ for the second harmonic.

The power of the circulating field and the obtainable efficiency strongly depend on the crystal absorption at the fundamental wavelength. A laser calorimeter was used to study the absorption for the ordinary rays at 1064 nm in the lithium-rich VTE crystals in more detail.[14] Figure 4 shows the results for a number of different samples. Crystals from three wafers processed in three different VTE runs were used. The circles show the absorption data of antireflection-coated samples cut from the 2-mm-thick *z*-cut wafer A. The best fit to those data is shown as a solid line and yields a fundamental wave absorption coefficient $\alpha_\omega = 0.42\%/\text{cm}$. The absorption data of two samples manufactured from a processed *y*-cut wafer are shown as the triangles. The crystal used in the doubling experiment described above had a single-pass absorption of 0.28%.

The uncoated sample cut from the *z*-cut wafer B showed the smallest absorption (0.11%/cm), as depicted by the square, which is comparable with that of congruent material.[15] The absorption coefficients for the different wafers vary considerably for reasons still under investigation. The 2-cm-long crystal has 0.4 times the absorption of the crystal used in the second-harmonic generation experiment, which should result in improved performance of the doubler when that crystal is used. The degradation of the beam profile owing to thermal gradients and the drop in conversion efficiency can be expected to occur at power levels 2.5 times higher when a 1-cm-long crystal made from this material is used. Lowering the crystal losses also leads to a steeper rise in efficiency with power, and the doubler should work efficiently at fundamental power levels less than 1 W. To double several watts of fundamental power efficiently, shorter crystals should be used. Owing to the large nonlinear coefficient of lithium niobate, the conversion efficiency will not decrease, while thermal loading will be reduced substantially.

To characterize qualitatively the resistance of lithium-rich lithium niobate to photorefractive damage, we operated the doubler at an input power of 1.8 W and generated 830 mW of 532-nm radiation for 100 min. Over this interval the output power varied by ±5%, comparable with the stability of the 1064-nm laser.

In conclusion, lithium-rich crystals fabricated by the VTE technique are promising for frequency-doubling applications. 1.6 W of power of 532-nm radiation has been generated in an external resonant cavity. The maximum internal efficiency for a 1-cm-long crystal was 69% for a cavity-matched input power of 2.2 W. Higher powers with similar conversion efficiencies should be achievable by using shorter crystals. The operation of the frequency doubler at temperatures near 234°C eliminates the effects of photorefractivity on the performance of the device. Absorption losses as low as 0.11%/cm at the fundamental wavelength have been observed.

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