Angle-Tuned Second-Harmonic Generation in LiIO\textsubscript{3} with Low Losses due to Index Matching

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Abstract. A new technique to reduce the losses of angle-tuned frequency doubling is described. Single-mode dye-laser radiation is injected in an external cavity containing a 7 mm LiIO\textsubscript{3} crystal, which is placed in a cuvette with index matching fluid. In this way, more than 5 mW usable uv power was achieved over a spectral range of more than 10 nm in uv.

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LiIO\textsubscript{3} is frequently used for second-harmonic generation because of its high nonlinear coefficient and its chemical stability. The work of Nath et al. has made the production of crystals possible, which can be used for frequency doubling in the 600/300 nm region. We reported on frequency doubling with LiIO\textsubscript{3} in this spectral region in 1982 [1]. A more comprehensive description is given in [2]. Majewski [3] placed a thin crystal in a modified dye laser and was able to produce uv radiation between 295 and 330 nm. A power of more than 2 mW could be achieved between 295 and 310 nm with a power of 15 mW at the optimum wavelength. Johnston and Johnston [4] placed their 6 mm crystal in the collimated arm of a commercial dye laser, generating uv light between 296 and 324 nm. They obtained more than 2 mW from 305 up to 322 nm with a peak of 5 mW at the optimum wavelength. Wang and Gaubatz used a 1 mm crystal in an external ring cavity [5]. They reported a maximum power of 5 mW at 603 nm with a fundamental power of 320 mW, which corresponds to 4.8 W in the ring cavity.

In our work, we used an external ring cavity with a crystal placed under Brewster’s angle. Angle tuning by tilting the crystal led to considerable reflection losses. Therefore we placed the crystal in a cuvette with an index matching fluid. When the cuvette was placed under Brewster’s angle in the external cavity, the reflection losses were always minimized; furthermore, the crystal was protected against degradation. Several index matching fluids were tested.

1. Theoretical Considerations

LiIO\textsubscript{3} is a negative uniaxial crystal of the symmetry group \(C_6\). Second-harmonic generation can be achieved with type I angle phase matching \((\omega + \omega \rightarrow \epsilon)\). The uv power \(P_2\) is given by (detailed representation and notation see [6] and [2])

\[
P_2 = cP_1^2.
\]

\(P_1\) is the fundamental power and

\[
c = \frac{2\cdot\omega_1^2}{(\pi n_1^2 n_2 c^3 \delta_0)} \cdot k_1 d_{\text{eff}}^2 \cdot L \cdot h_m(B, \zeta)
\]

the conversion coefficient. The latter is in the main characterized by the square of the nonlinear coefficient \(d_{\text{eff}} = d_{31} \sin \Theta_m\) (where \(\Theta_m\) is the phase matching angle); the crystal length \(L\) and the function \(h_m(B, \zeta)\), which describes the influence of focusing and double refraction. Absorption is not taken into account here. The uv power increases with \(L\), but the progress is less than linear because \(h_m\) decreases with \(L\). In many practical cases an increase with \(\sqrt{L}\) is valid. The absorption of the fundamental wave can be neglected, but the absorption of the uv radiation, however, has to be taken into account. It can even cause a decrease of the
uv power with increasing crystal length. The length dependent part of the conversion coefficient has the form

$$f(L) = L \cdot e^{-\frac{L}{a_2^2}} \cdot h_{nm}(L).$$

$L$ is the crystal length, exp$(-a_2 L/2)$ gives the absorption of the uv radiation (slightly simplified, see [6]), and $h_{nm}(L)$ is obtained from $h_{nm}(B)$ given in [6] by the relation $L = 4B^2/(\alpha^2 k_1)$. $h_{nm}$ is the value of $h_{nm}(B, \zeta)$ for optimal focusing conditions. Figure 1 shows the length dependence of the conversion coefficient for small absorption ($\alpha_2(305 \text{ nm}) = 0.015 \text{ mm}^{-1}$) and stronger absorption ($\alpha_2(295.5 \text{ nm}) = 0.14 \text{ mm}^{-1}$). The absorption data is taken from Nath et al. [7]. For wavelength $\lambda > 300 \text{ nm}$ the behaviour is similar to the absorption free case. $f(L)$ increases with about $\sqrt{L}$. Near the limiting wavelength of 293 nm $f(L)$ increases very quickly in the beginning, but there is no more increase for crystal length of about 7 mm.

The second-harmonic generation takes place in an external ring cavity in order to enhance the fundamental power. The fundamental power $P_1$ in the cavity is the product of the incoming power $P_{in}$ and the enhancement factor $A$ (for plane waves [2])

$$P_1 = P_{in} \cdot A = P_{in} \frac{1 - R_1}{(1 - \sqrt{R_1} V)},$$

$R_1$ being the reflectivity of the incoupling mirror $M_1$, $R_2 = R_3 = R_4 = 1$, and $V$ the round-trip loss factor (e.g., $V = 0.96$).

The frequency-doubling unit has to be inserted in the cavity under Brewster's angle for the incoming polarization in order to obtain good enhancement. In addition to this, the electric field vector $\mathbf{E}$ of the incoming radiation has to be perpendicular to the optical axis of the crystal, so that the fundamental wave passes as an ordinary beam (in the case of LiIO$_3$). If this condition is not fulfilled, part of the light passes the crystal as an extraordinary beam which deteriorates enhancement and uv conversion. There are three possibilities for angle tuning in a cavity:

1) A Brewster-cut crystal is placed in the resonator. High power will be obtained for the corresponding wavelength. If the crystal is tuned by tilting it in the plane of the optical axis and the propagation vector $\mathbf{k}$, the fundamental power decreases quickly because of the additional reflection losses.

2) As a second possibility the crystal can be tuned by turning it around the surface normal vector [3]. In this case the Brewster angle is maintained, but the condition $\mathbf{E}$ perpendicular to the optical axis is unfulfilled. This changes the round-trip loss factor by a factor of $\sin^2 \Theta_o$ (where $\Theta_o$ is the angle between $\mathbf{E}$ and the optical axis). This method produces better results than the first one.

3) We propose a third possibility which permits maintaining the Brewster angle for the frequency doubling unit and the condition $\mathbf{E}$ perpendicular to the optical axis: The crystal is placed in a cuvette filled with an index matching fluid. The cuvette is placed in the ring cavity under Brewster's angle. The geometry of the set up is shown in Fig. 2.

![Fig. 2a and b. This figure shows the path of the beam through the cuvette with index matching fluid and the crystal for the case of perfect index matching. (a) shows a horizontal cut in the plane of the beam path and the tilting axis $\Omega$, and (b) represents a vertical cut in plane of the beam path and the optical axis](image-url)