

Frequency Doubling of cw Ring-Dye-Laser Radiation in Lithium Iodate Crystals

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Abstract. We report on frequency doubling of cw ring-dye-laser radiation in lithium iodate crystals (LiIO_3). The conversion coefficient and the matching angle were measured as a function of the fundamental wavelength in the range 586.3–624 nm. For Type I phase-matching the matching angle $\theta_m = 90^\circ$ for noncritical matching was found at 586.3 nm. At this wavelength the conversion coefficient reaches the maximum value which was observed to be as high as $1.6 \times 10^{-2} \text{ W}^{-1}$ for a LiIO_3 crystal of 1 cm length. From the dependence of the matching angle on the wavelength the refractive indices were determined for the ordinary as well as for the extraordinary ray in the wavelength range from 293 nm up to 355 nm. The influence of focussing on the conversion coefficient is discussed.

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The high output and especially the much higher intracavity power, respectively, of cw tunable single-mode ring dye lasers offers the possibility of extending high-resolution laser spectroscopy into the near uv or ir region by using the nonlinear properties of suited crystals for frequency doubling (Second-Harmonic Generation, SHG), Sum-Frequency Mixing (SFM) and Difference-Frequency Mixing (DFM). Mostly ADA crystals have been applied for SHG in the spectral range of Rhodamine 6G using temperature phase-matching. This paper reports on SHG in LiIO_3 crystals using angle phase-matching.

Lithium Iodate (LiIO_3) forms negative uniaxial crystals of C_6 symmetry with high nonlinear optical susceptibility. Since 1969 [1, 2] these crystals have found broad application in nonlinear optics. The following examples should be mentioned:

- frequency doubling of the radiation of Nd:YAG [3] or ruby lasers [4, 5]

- generation of ir laser light by difference-frequency mixing [6]
- application in a parametric oscillator [20, 21]
- application as correlator for picosecond pulses.

Owing to some favourable features LiIO_3 crystals are very attractive for SHG:

- a high nonlinear coefficient: $d_{31} = (7.31 \pm 0.62)$ pm/V for $\lambda = 488/514.5$ nm $\{d_{31}(\text{LiIO}_3) = (11.3 \pm 3.1) d_{36}(\text{KDP})\}$ [7]
- a very weak temperature dependence of the indices of refraction
- a high damage threshold
- good chemical resistance under laboratory conditions
- optical transmission down to 300 nm
- a high wavelength acceptance.

Up to now LiIO_3 crystal rarely have been used below 355 nm for the generation of uv laser radiation. This is mainly due to the fact, that considerable uv absorption can occur [2, 5]. Improved growing techniques for LiIO_3 crystals can considerably reduce the uv absorption [5]. Several groups demonstrated efficient SHG in the wavelength range of Rh 6G with a pulsed dye laser

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[8] as well as a cw single-mode linear dye laser [9, 10]. Recently two experimental studies of LiIO_3 intracavity SHG have been published. First Majewski [23] generated between two and fifteen mW of uv power in the spectral range of 295–310 nm. The 1 mm crystal was mounted in a waist of the laser resonator. Johnston and Johnston [24] extended the doubling region up to 324 nm, placing the crystal in the collimated arm of the beam in the resonator.

The data known for the indices of refraction and the corresponding Sellmeier coefficients [11–14] are confined to the wavelength range from 355 nm up to 5 μm . We have found that these data cannot be extrapolated to shorter wavelengths because of the onset of absorption.

1. Theory

Neglecting absorption the power P_2 of second-harmonic generation by frequency doubling can be calculated for Gaussian beams according to the theory of Boyd and Kleinman [16]:

$$P_2 = K k_1 d_{\text{eff}}^2 l h(B, \xi) P_1^2 = c P_1^2; \quad (1)$$

where c is called the conversion coefficient for frequency doubling. P_1 is the power of the fundamental wave. The influence of focussing and double refraction is described by the dimensionless function $h(B, \xi)$ where $B = 0.5 \varrho \sqrt{l k_1}$ is the parameter of double refraction and $\xi = l/b$ is the parameter of focussing ($b = w^2 k_1$: confocal parameter).

The constant K contains some elementary quantities (SI units)

$$K = w_1^2 / \pi n_1^2 n_2 c^3 \varepsilon_0.$$

The other quantities are k_1 : wave number of the fundamental wave in the crystal; d_{eff} : effective nonlinear coefficient, l : crystal length, λ : angle of double refraction, w_1 : waist radius of the Gaussian beam, n_1 : index of refraction of the fundamental wave, and n_2 : index of refraction of the harmonic wave.

The effective nonlinear coefficient d_{eff} depends on the material, the symmetry class of the crystal and on the type of the nonlinear interaction [17]. Kleinman's symmetry condition [18] leads to the following non-vanishing nonlinear coefficients for lithium iodate: d_{15} , d_{31} , and d_{33} with $d_{15} = d_{31}$. For Type I phase-matching [$o(\omega_1) + o(\omega_1) \rightarrow e(\omega_2)$] the effective nonlinear coefficient is given by

$$d_{\text{eff}} = d_{31} \sin \theta_m, \quad (2)$$

where θ_m is the matching angle.

For Type II phase-matching [$o(\omega_1) + e(\omega_1) \rightarrow e(\omega_2)$] the effective nonlinear coefficient depends on the matching angle through $d_{\text{eff}}(\text{II}) = 0.5 d_{14} \sin(2\theta_m)$.

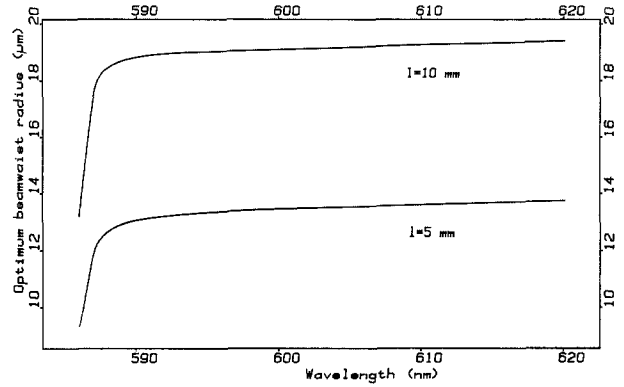


Fig. 1. The optimum SHG waist as a function of the wavelength for two crystal lengths

Kleinman's symmetry condition demands $d_{14} = 0$. Okada and Ieiri [14], however, observed the Type II process and determined the ratio d_{14}/d_{31} to be $(4.3 \pm 0.15) \times 10^{-2}$, so that this process is not suited for SHG of cw laser radiation.

According to (1 and 2) the conversion coefficient reaches its maximum value at $\theta_m = 90^\circ$ (noncritical phase-matching) since also the function $h_m(B, \xi)$ approaches a maximum because of vanishing double refraction. If additionally the optimum focussing parameter $\xi_m = 2.84$ is realized the absolute maximum of the function $h_m(\theta, \xi_m)$ is obtained [16].

In the case of critical phase-matching ($\theta_m < 90^\circ$) the resulting beam walk-off lowers the conversion coefficient drastically. According to Boyd and Kleinman [16] an optimum focussing parameter ξ_m exists for any matching angle in the range $1.39 < \xi_m < 2.84$. In Fig. 1 the optimum waist radius w_{opt} of the Gaussian laser beam is plotted as a function of the fundamental wavelength for two crystals of different length, 5 and 10 mm, respectively. Apart from the region of the boundary wavelength the optimum waist radius is only weakly dependent on the fundamental wavelength as well as on the length of the crystal. The dependence on both quantities can be approximated in each case by $\sqrt{\lambda_1}$ and \sqrt{l} , respectively. So operating with an average waist radius seems to be sufficient in the case $\theta_m < 90^\circ$.

For demonstrating the influence of focussing on the conversion coefficient Fig. 2 shows (for the case of $\lambda_1 = 600$ nm) the dependence of h_m on the ratio w/w_0 . If the waist radius w differs from w_0 by as much as 50% the generated power of the second harmonic is reduced by less than 20%. Provided the magnitude w_L and the position of the laser waist is known, the optimum waist radius w_0 in the crystal can be realized with an appropriate lens according to the well known relation [19]

$$\frac{w_L^2}{w_0^2} = \frac{d_1 - f}{d_2 - f}. \quad (3)$$

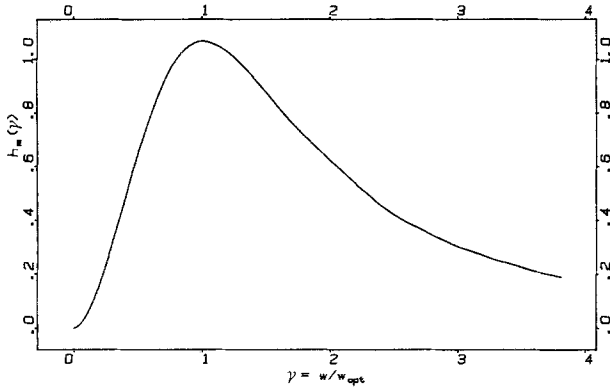


Fig. 2. The influence of the focussing on h_m and hence on the second harmonic power. The double-refraction is neglected ($B=0$)

The focal length f of the lens must be chosen larger than the critical length $f_0 = w_0 w_L \pi / \lambda$. Then the distance d_1 between the laser waist and the lens determines the position of the waist w_0 at the distance d_2 from the transforming lens. The presence of the crystal does not alter the magnitude of the waist but shifts its position towards the imaging lens by $a \cdot (n-1)/n$ where a is the distance of the waist from the entrance face of a hypothetical crystal with $n=1$.

2. Experimental

Our experimental arrangement is shown in Fig. 3. The radiation of a tunable cw ring dye laser (Coherent Mod. 699-21) was focussed into the LiIO_3 crystal by the high-reflecting mirror M1 ($f=11.4$ cm). We used two lithium iodate crystals (supplied by Gsaenger-Optische Komponenten) of different lengths, 5.2 and 10 mm, respectively, with uncoated faces. One crystal (5.2 mm) was cut at an angle of 75° , the other at an angle of 67° with respect to the optical axis. The mirror M2 served as a refocussing element for the fundamental as well as for the harmonic beam. The crystal was mounted on a rotator for adjusting the exact matching angle. The uncertainty in determining the

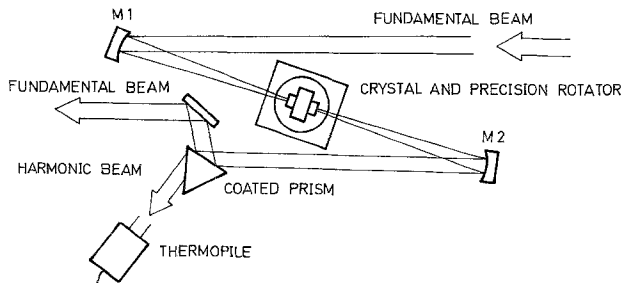


Fig. 3. Experimental set up for the measurement of the conversion coefficient

matching angles was $\pm 0.1^\circ$. The rotator was attached to a translator in order to optimize the position of the waist in the crystal. The optical axis lay in the image plane and the polarization perpendicularly to it. The matching angle was found by adjusting the crystal to optimum uv power.

The fundamental and the harmonic beam were separated by a Brewster prism whose entrance surface was coated for highly reflecting the fundamental beam. The correct waist radius was realized by choosing the appropriate distance between the laser head and the doubling unit.

In order to determine the conversion coefficients we used three thermopiles. The fundamental power was measured with a calibrated thermopile (Coherent Mod. 210), the second-harmonic power was monitored with a sensitive, thermally compensated thermopile with a short response time (Kipp + Zonen Mod. CA1). Both devices were compared with a calibrated thermopile [Laser Instruments Mod. 17S, calibrated by the Physikalische-Technische Bundesanstalt (PTB)].

3. Results

3.1. Determination of the Indices of Refraction

Using the 10 mm lithium iodate crystal we measured the matching angle θ_m as a function of the fundamental wavelength between 586 and 624 nm. In this region of beginning absorption for the second harmonic the indices of refraction were deduced from the matching angles which are related according to

$$\sin^2 \theta_m(I) = \frac{[n_o(\omega)]^{-2} - [n_o(2\omega)]^{-2}}{[n_e(2\omega)]^{-2} - [n_o(2\omega)]^{-2}}. \quad (4)$$

This expression holds for Type I phase-matching in negative uniaxial crystals [17]. $n_o(\omega)$, $n_o(2\omega)$, and $n_e(2\omega)$ are the indices of refraction for the ordinary ray at the fundamental frequency ω and at the second-harmonic frequency 2ω , respectively, and for the extraordinary ray at 2ω . Except for $n_o(\omega)$ all other indices of refraction are unknown in the spectral range investigated. $n_o(\omega)$ was taken from a 3-parameter Sellmeier equation

$$n_o^2(\lambda) = A + \frac{B}{\lambda^2 - C}, \quad (5)$$

where $A=3.4095$, $B=0.047664 \mu\text{m}^2$ and $C=0.033991 \mu\text{m}^2$ (in the spectral range $0.3547 \mu\text{m} < \lambda < 0.644 \mu\text{m}$) [22].

In a first step we started the determination of the wavelength dependence of the unknown indices of refraction $n_o(2\omega)$ and $n_e(2\omega)$ by using Sellmeier equations according to (5) and by fitting the six unknown parameters to the experimental data in a least squares

Table 1. Sellmeier coefficients for LiIO_3 in the spectral range of $293 \text{ nm} \leq \lambda \leq 335 \text{ nm}$

| | n_o | n_e |
|---|-----------------------------|--------------------------|
| A | 3.425834 | 2.913366 |
| B | $0.046664 \mu\text{m}^2$ | $0.034267 \mu\text{m}^2$ |
| C | $0.032155 \mu\text{m}^2$ | $0.033230 \mu\text{m}^2$ |
| D | $0.010334 \mu\text{m}^{-2}$ | $0.0 \mu\text{m}^{-2}$ |

procedure. Since the result for n_e agreed with known values at 354.7, 404.7, and 454.5 nm within ± 0.0007 , the parameters for n_e were kept fixed in the second step. For improving the precision we used the following four parameter Sellmeier equation for n_o

$$n_o^2 = A + \frac{B}{\lambda^2 - C} - D\lambda^2. \quad (6)$$

As a result of the repeated fitting procedure the discrepancy between the (directly) measured and our calculated indices of refraction near 400 nm is less than ± 0.0007 . At the fundamental wavelength of $\lambda = 709.4 \text{ nm}$ the fit was linked to the directly measured indices by using the calculated matching angle of 50.91° as an additional experimental point.

Table 1 summarizes the Sellmeier coefficients determined. The boundary fundamental wavelength for the case of 90° -phase-matching (Type I) in LiIO_3 crystals was found to be 586.3 nm. Figure 4 shows the experimentally determined matching angles and the result of the fit (solid line) as a function of the fundamental wavelength.

3.2. Determination of the Conversion Coefficients

The theoretical conversion coefficients were calculated according to (1) using our index data and the nonlinear coefficient $d_{31} = (7.31 \pm 0.62) \text{ pm/V}$ [7]. It has been shown [5] that under proper growing conditions the LiIO_3 crystals are totally transparent above 300 nm. So we did not take into account any absorption of the harmonic wave. For $\lambda < 300 \text{ nm}$ a steep absorption edge exists at about 290 nm. Figure 5 depicts the comparison between the theoretical conversion coefficient and the experimental data for the 10 mm LiIO_3 crystal. These experimental results have been corrected for reflection losses of the fundamental wave at the incident surface and reflection losses of the harmonic wave at the exit surface. The results have been obtained using the ring laser without intracavity etalons. Single-mode measurements yielded the same results within our error limits. This is due to the fact that even in broad band operation the ring laser oscillates in a single mode but with considerable frequency jitter. Thus our results can be regarded as single-mode conversion coefficients.

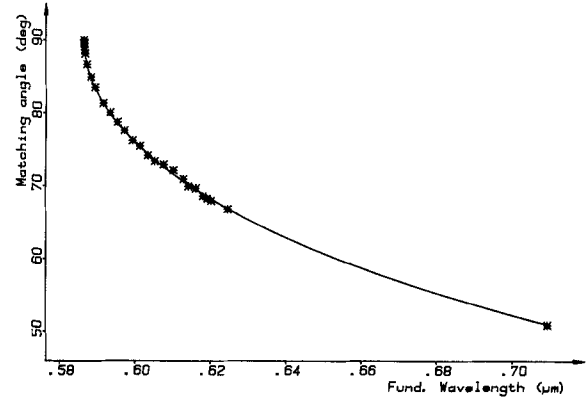


Fig. 4. Experimental matching angles versus the fundamental wavelength (solid line: the least squares fit for the determination of the Sellmeier coefficients)

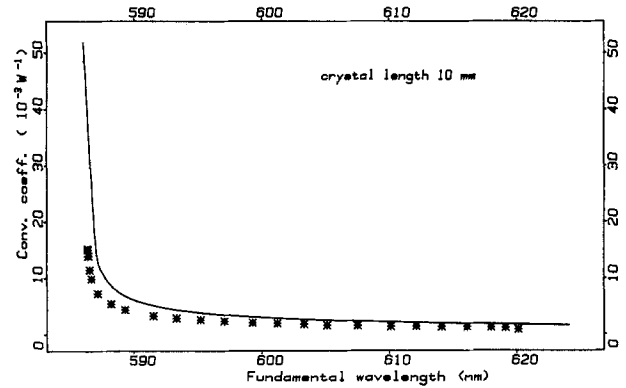


Fig. 5. Comparison between calculated and measured conversion coefficients of a 10 mm LiIO_3 crystal

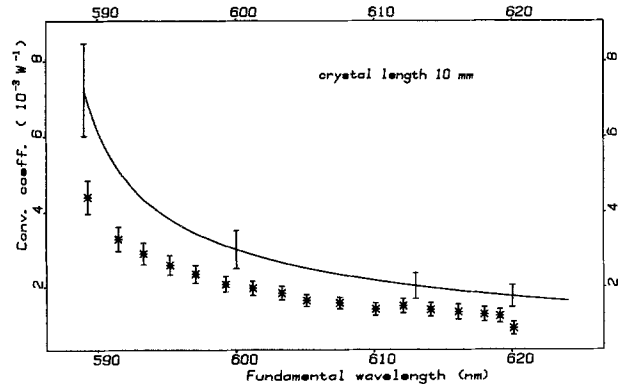


Fig. 6. Section of Fig. 5 with error bars

Figure 6 shows a part of Fig. 5 for better comparison between theory and experiment. The experimental values lie below the theoretical curve. This is due to the following reasons:

- The beam profile of the ring dye laser differed considerably from the TEM_{00} intensity distribution

- The focus was optimized for the centre of the interesting spectral region, thus it was not optimized near 586 nm (Fig. 1)
 - Below 300 nm the absorption of the harmonic wave sets on strongly. Therefore, theoretical and experimental values are diverging.
- Nevertheless, at the limiting wavelength we measured the remarkably high conversion coefficient of $c_{\max} = 1.6 \times 10^{-2} \text{ W}^{-1}$. c remains larger than 10^{-3} W^{-1} up to 620 nm.

4. Discussion

The Sellmeier coefficients determined in this work reproduce the course of the indices of refraction n_o and n_e in the spectral region 293–454 nm. Using these coefficients the limiting wavelength for Type I frequency doubling turns out to be 586 nm. This is in good agreement with the experiment, which gives 586.3 nm while the Sellmeier coefficients known so far predict a limiting wavelength of 580 nm. Our investigation demonstrates that LiIO_3 is well suited for external frequency doubling with cw dye lasers. Experiments with LiIO_3 in an external cavity are in progress in our group. The influence of the crystal tuning on the enhancement of the cavity has been studied. As a preliminary result 370 μW uv power has been generated with 190 mW input power at 594 nm and an enhancement factor of 9. The small enhancement is due to the fact, that the crystal cut is not optimized relative to the polarization and the phase-matching conditions in the cavity. Tuning the wavelength at fixed crystal position the width at half maximum of the tuning curve was found to be 2.7 nm, which is large compared to the tuning width of temperature matched crystals.

References

1. G. Nath, S. Haussuehl: Appl. Phys. Lett. **14**, 154–156 (1969)
2. F.R. Nash, J.G. Bergmann, G.D. Boyd, E.H. Turner: J. Appl. Phys. **40**, 5201–5206 (1969)
3. U. Deserno, G. Nath: Phys. Lett. **30 A**, 483–484 (1969)
4. G. Nath, S. Haussuehl: Phys. Lett. **29 A**, 91–92 (1969)
5. G. Nath, H. Mehmanesch, M. Gsaenger: Appl. Phys. Lett. **17**, 286–288 (1970)
6. B. Wellegehausen, D. Friede, H. Vogt, S. Shahdin: Appl. Phys. **11**, 363–370 (1976)
7. M.M. Choy, R.L. Byer: Phys. Rev. B **14**, 1693–1706 (1976)
8. V.I. Stroganov, V.I. Trunov, A.A. Chernenko, A.N. Izrailenko: Sov. J. Quant. Electron. **6**, 601–602 (1976)
9. A. Fischer, R. Kullmer, W. Demtroeder: Chem. Phys. **83**, 415–428 (1984)
10. I.M. Beterov, V.I. Stroganov, V.I. Trunov, B.Ya. Yurshin: Sov. J. Quant. Electron. **5**, 1329–1331 (1976)
11. S. Umegaki, S.I. Tanaka, T. Uchiyama, S. Yabmuoto: Opt. Commun. **3**, 244 (1971)
12. J.M. Crettez, J. Comte, E. Coquet: Opt. Commun. **6**, 244 (1972)
13. R.L. Herbst: Quoted in [7]
14. M. Okada, S. Ieiri: Phys. Lett. **34 A**, 63–64 (1971)
15. S. Umegaki, S. Tanaka: Jpn. J. Appl. Phys. **16**, 775–781 (1977)
16. G.D. Boyd, D.A. Kleinman: J. Appl. Phys. **39**, 3597–3640 (1968)
17. F. Zernike, J.E. Midwinter: *Applied Nonlinear Optics* (Wiley, New York 1973)
18. D.A. Kleinman: Phys. Rev. **126**, 1977–1979 (1962)
19. H. Kogelnik, T. Li: Proc. IEEE **54**, 1312–1329 (1966)
20. L.S. Goldberg: Appl. Phys. Lett. **17**, 489–491 (1970)
21. A.J. Campillo, C.L. Tang: Appl. Phys. Lett. **16**, 242–244 (1970)
22. Information sheet on LiIO_3 , Cleveland Crystals, Inc.
23. W.A. Majewski: Opt. Commun. **35**, 201–206 (1983)
24. T.F. Johnston, T.J. Johnston: In *Laser Spectroscopy VI*, ed. by H.P. Weber, W. Luety, Springer Ser. Opt. Sci. **40** (Springer, Berlin, Heidelberg 1983) pp. 417–418; Coherent, Inc. Information sheet