Wavelength temperature and angle bandwidths in SHG of focused beams in nonlinear crystals

S. Blit, E. G. Weaver, and F. K. Tittel

The temperature, wavelength, and angle bandwidths in second harmonic generation of focused Gaussian beams in nonlinear crystals are evaluated. Numerical and experimental results are given for ADP as a function of the phase matching angle. These results show higher bandwidths compared with the plane wave case.

I. Introduction

The efficiency of optical second harmonic generation is determined by interaction parameters such as wavelength, direction of propagation in the crystal, and temperature. The efficiency of the harmonic generation decreases from its optimum value by variation of these parameters from the exact phase-matched values. A question of practical importance in designing SHG schemes is the determination of just how large the variation of these parameters can be tolerated before the SHG efficiency decreases appreciably. Parametric bandwidth (BW) as considered in this paper is defined as the variation of a parameter that causes the harmonic power to drop to one half of its maximum phase-matched value.

With the increasing interest in SHG and sum frequency mixing of cw radiation,[1] there is a need to determine the effect of focusing on the parametric bandwidths. Spectral BW and angle BW (sometimes referred to as acceptance angle) were treated in detail recently[2] for the plane wave case using a Taylor series expansion approach based on the sinc function approximation of phase matching of plane parallel waves. A similar approach was also taken by other investigators for specific cases, of angle and wavelength bandwidths,[3–5] and of temperature BW as well.[3,6]

In this paper the wavelength, temperature, and angle bandwidths of SHG of focused Gaussian beams in nonlinear angle phase-matched crystals are evaluated. The calculation is based on the Boyd and Kleinman theory,[7] which replaces the sinc function approximation by a numerically calculated function h, which contains both focusing and angle phase-matching information. This analysis is applied to a specific example using the crystal ADP. Theoretical BWs are calculated for which experimental measurements were performed. Some consideration is also given to the rate of temperature tuning, since in many systems angle tuning is undesirable because of beam deviation and displacement (for example, in intracavity SHG). The calculated and experimental BW obtained demonstrate the typical behavior expected for SHG of focused beams in KDP isomorphs.

II. General Theory

The basic theory for SHG has been developed extensively. For near-field plane parallel interactions the second harmonic power is proportional to the following sinc function:

$$P_2 \propto \left[\frac{\sin^2(\tfrac{1}{2}\Delta kl)}{(\tfrac{1}{2}\Delta kl)^2}\right], \tag{1}$$

where l is the crystal length, and Δk is the phase mismatch given by

$$\Delta k = 2k_1 - k_2. \tag{2}$$

From the Boyd and Kleinman theory for focused Gaussian beams, the expression for the second harmonic power including the effects of focusing, absorption, and phase-matching is given by Eq. (2.22) of Ref. 7:

$$P_2 = KP_1^2\, lk_1 \exp(-\alpha l) h(\sigma,\beta,\kappa,\xi,\mu), \tag{3}$$

where

$$h(\sigma,\beta,\kappa,\xi,\mu) = \frac{\exp(\mu\alpha l)}{4\xi} \int\int_{-\xi(1-\mu)}^{\xi(1+\mu)} d\tau d\tau' \times \frac{\exp[-\kappa(\tau+\tau') + i\sigma(\tau-\tau') - \beta(\tau-\tau')^2]}{(1+i\tau)(1+i\tau')}, \tag{4}$$

When this work was done all authors were with Rice University, Department of Electrical Engineering, Houston, Texas 77001. S. Blit is now with Cornell University, Materials Science Center, Ithaca, New York 14853, and E. G. Weaver is with Hewlett-Packard, Inc., Palo Alto, California 94304.

Received 23 June 1978.
0003-6935/79/050733-04$00.50/0.

$$\left.\begin{aligned} \sigma &= \tfrac{1}{2} b \Delta k, \\ \beta &= \rho/\delta, \\ \kappa &= \tfrac{1}{2}\alpha b, \\ \xi &= l/b, \\ \mu &= (l-2f)/l, \end{aligned}\right\} \quad (5)$$

with the principal parameters defined as
b = confocal parameter of the beam,
ρ = double refraction walk off angle,
δ_0 = diffraction half angle,
f = focal length, and
α = absorption coefficient.

The bandwidth information is contained in the function h. Assuming no absorption ($\alpha = 0$) and assuming the focus is centered within the crystal ($\mu = 0$), the expression for h may be simplified[8] to

$$h(\xi,\beta,\sigma) = \frac{2}{\xi}\int_0^{\xi} \frac{\alpha_1\alpha_2 - \beta_1\beta_2 + x(\alpha_1\beta_2 + x(\alpha_1\beta_2 + \beta_1\alpha_2)}{1+x^2}\,dx, \quad (6)$$

where $x = \tfrac{1}{2}(\tau - \tau')$ is the integration parameter, and

$$\alpha_1 = \tfrac{1}{2}\left[2\pi m + \tan^{-1}\left(\frac{2k-2\xi}{\xi^2 - 2x\xi - 1}\right)\right]$$

$$\alpha_2 = \exp(-4x^2\beta^2 \cos 2x\sigma)$$

$$\beta_1 = -\tfrac{1}{2}\ln\left[\frac{1+\xi^2}{1-(\xi-2x)^2}\right]^{1/2},$$

$$\beta_2 = \exp(-4x^2\beta^2)\sin 2x\sigma,$$

$$m = \text{integer}.$$

The integral in Eq. (6) can be numerically evaluated using Romberg integration to determine h as a function of the three main parameters ξ, β, and σ. ξ is the focal parameter and is determined by the crystal length and the focal length of the lens used for focusing the beam. β contains the walk-off angle information and is a function of λ and the nonlinear material. σ is the phase mismatch parameter.

The full width at half maximum (FWHM) bandwidth of σ is calculated for a given ξ and β by first determining σ_m such that the function $h(\xi,\beta,\sigma)$ is maximized. Then the values $\sigma^+(>\sigma_m)$ and $\sigma^-(<\sigma_m)$ are found which reduce the function $h(\xi,\beta,\sigma)$ to half of its maximum value:

$$h(\xi,\beta,\sigma^+) = h(\xi,\beta,\sigma^-) = \tfrac{1}{2}h(\xi,\beta,\sigma_m). \quad (7)$$

From these values the FWHM bandwidth $\Delta\sigma = \sigma^+ - \sigma^-$ is obtained and is then related to the Δk_{FWHM} bandwidth by

$$\Delta k_{\text{FWHM}} = 2\Delta\sigma/b. \quad (8)$$

Since the dependence of Δk on a nonlinear parameter X is sometimes complicated, Δk can be expanded in a Taylor series:

$$\Delta k = \Delta k_0 + \frac{\partial \Delta k}{\partial X}\Delta X + \tfrac{1}{2}\frac{\partial^2 \Delta k}{\partial X^2}(\Delta X)^2 + \ldots, \quad (9)$$

where ΔX is the FWHM bandwidth of the parameter X. By substituting the value Δk_{FWHM} determined by Eq. (8) and solving for ΔX, one has calculated a bandwidth ΔX of the parameter X, which includes focusing and walk-off. The following evaluation will be concerned with the parameter X representing the wavelength λ, the temperature T, and the internal angle θ. Other parameters could be considered, but these three will be regarded as the principal nonlinear parameters of interest.

In order to determine the spectral BW of the fundamental wavelength, it is required that the partial derivatives of the wave vector mismatch Δk with respect to the wavelength λ_1 be known. Equation (2) can be expressed for type I phase matching in a negative uniaxial crystal as

$$\Delta k = 2\left(\frac{2\pi n_{o1}}{\lambda_1}\right) - \frac{2\pi n_{e2}(\theta)}{\lambda_2}, \quad (10)$$

where the extraordinary index is determined from the phase-match angle by

$$n_{e2}(\theta) = \left(\frac{\cos^2\theta}{n_{o2}^2} + \frac{\sin^2\theta}{n_{e2}^2}\right)^{-1/2}. \quad (11)$$

If the indices of refraction of the crystal are represented in the form of a Sellmeier equation,

$$n_{ij}^2 = S_{i1} + \frac{S_{i2}}{\lambda_j^2 - \dfrac{1}{S_{i3}}} + \frac{S_{i4}\lambda_j^2}{S_{i5}\lambda_j^2 - 1}, \quad (12)$$

where S_{i1-5} are constants, $i = o$ (ordinary) or e (extraordinary), and $j = 1$ (fundamental) or 2 (harmonic). Then $\partial\Delta k/\partial\lambda_1$ can be determined by substituting Eq. (11) into Eq. (10), differentiating, and then substituting $n_{o1} = n_{e2}(\theta)$ as

$$\frac{\partial \Delta k}{\partial \lambda} = \frac{2\pi}{\lambda_2}\left[\tfrac{1}{2}\left(\frac{n_{o1}^3}{n_{o2}^3}\cos^2\theta\,\frac{\partial n_{o2}}{\partial \lambda_2} + \frac{n_{o1}^3}{n_{e2}^3}\sin^2\theta\,\frac{\partial n_{e2}}{\partial \lambda_2}\right) - \frac{\partial n_{o1}}{\partial \lambda_1}\right], \quad (13)$$

where the derivatives $\partial n_{o1}/\partial\lambda_1$, $\partial n_{o2}/\partial\lambda_2$, and $\partial n_{e2}/\partial\lambda_2$ can be calculated from Eq. (12). The expression for $\partial^2\Delta k/\partial\lambda_1^2$ is determined by extending the same process. Thus, the first-order derivatives for θ and T are

$$\frac{\partial \Delta k}{\partial \theta} = \frac{2\pi}{\lambda_2}\left[n_{e2}^3(\theta)\cos\theta\sin\theta\left(\frac{1}{n_{o2}^2} - \frac{1}{n_{e2}^2}\right)\right], \quad (14)$$

$$\frac{\partial \Delta k}{\partial T} = \frac{2\pi}{\lambda_2}\left(\frac{n_{o1}^3}{n_{o2}^3}\cos^2\theta\,\frac{\partial n_{o2}}{\partial T} + \frac{n_{o1}^3}{n_{e2}^3}\sin^2\theta\,\frac{\partial n_{e2}}{\partial T} - \frac{2 n_{o1}}{\partial T}\right). \quad (15)$$

These equations can now be applied to calculate the bandwidths expected for a typical experiment.

To apply the preceeding calculation, the experimental parameters such as crystal length, focal length of the lens, phase-matching angle, etc., which characterize the parametric interaction, are converted into the appropriate parameters ξ and β using Eq. (5). Two possibilities for the focusing lens will be considered: first, the optimum focal length, which varies as a function of λ and gives the maximum SHG efficiency for each phase-matching angle; second, a fixed focal length lens, which is close to the optimum over a wide range of wavelengths.

III. Results

The preceding analysis is applied to SHG in a 2.5-cm long ADP crystal at 25°C. The focus of the beam is centered in the crystal, and the fixed focal length is taken as 7.5 cm (the optimum at 88.5° and 43° phase-matching angles). Index of refraction data for ADP from Zernike[9] provide the necessary Sellmeier equations to describe $n(\lambda)$. The effects of temperature on the index of refraction are taken into account using the data from Phillips.[10] These sets of data are then used to calculate values for the first derivatives in Eqs. (13)–(15) as well as for the corresponding second partial derivatives. Then these values are used in Eq. (9) along with the value of Δk_{FWHM} [determined for the appropriate conditions in Eq. (8)] to calculate the bandwidths $\Delta\lambda$, ΔT, and $\Delta\theta$. For comparison, the unfocused bandwidths are calculated from Eq. (9) with Δk_{FWHM} determined from Eq. (1) ($l\Delta k/2 = 1.39$).

For the case of $\Delta\lambda$, the first-order term of the expansion is dominant to the extent that higher order terms can be neglected. The results of the calculations of the wavelength bandwidth using the first-order term are shown in Fig. 1(a) as a function of θ_m (and λ_1). $\Delta\lambda$ increases with θ_m and is larger than calculated with the sinc function approximation.

Using the temperature data[10] only the first-order terms are important for determining ΔT. The calculated results of ΔT vs $\theta_m(\lambda)$ are shown in Fig. 1(b). ΔT increases monotonically with θ_m with values much greater than those usually associated with unfocused SHG.

Finally, $\Delta\theta$ is evaluated and is shown in Fig. 1(c). Unlike ΔT and $\Delta\lambda$, the calculation of $\Delta\theta$ near 90° phase matching requires the second order expansion terms from Eq. (9) in order to correct for singularity from the first-order terms [Eq. (14)]. $\Delta\theta$ is seen to decrease quite sharply from a maximum at $\theta_m = 90°$ and then levels off at about an 85° phase-matching angle. In all three cases, both the fixed and optimized focal length behaviors follow each other closely with the fixed focal length exhibiting a slightly larger bandwidth except very near a 90° phase-matching angle.

These results were also checked experimentally. The radiation of a cw rhodamine 6G tunable dye laser and the 531-nm, 647-nm, 753-nm lines of a Kr laser were frequency doubled in two ADP crystals mounted in a temperature stabilized cells. The z-cuts of the crystals were 65° and 80°. The angle bandwidth was determined by accurately rotating the cell using a stepping motor driven rotary mount with a resolution of 0.07 mrad. The dye laser wavelength, having a bandwidth of 0.5 Å, was tuned to determine the wavelength bandwidth. To find the temperature bandwidth the temperature of the liquid filled cell, stabilized to 0.1°C, was varied very slowly to allow for homogeneous temperature distribution in the crystal. The recollimated and filtered uv beam was measured with a uv enhanced silicon photodiode and recorded as a function of the variable parameter. The experimental results for the wavelength, temperature, and angle bandwidths are given in Fig. 1. It is seen that the experimental results are in satisfactory agreement with the behavior of predicted values. However, in each case the measured bandwidth is larger than the predicted value. This discrepancy is expected to originate from material and thermal inhomogeneities in the crystal as well as from the finite linewidth of the source. Also, close to 90° phase matching, a behavior, very similar to the predicted, was observed in ADA. These BW predictions are seen to be an improvement over those resulting from the plane parallel theories.

Temperature tuning at 90° results in high efficiency but is relatively slow in scanning. This can be seen from Fig. 1(c) where the temperature bandwidth is minimal at a 90° phase-matching angle. In fact, all the systems employing 90° phase matching use temperature stabilization of the crystal to retain a constant uv power at a constant wavelength. Also, the nearer the crystal to a 90° phase-matching angle, the more prone it is to an

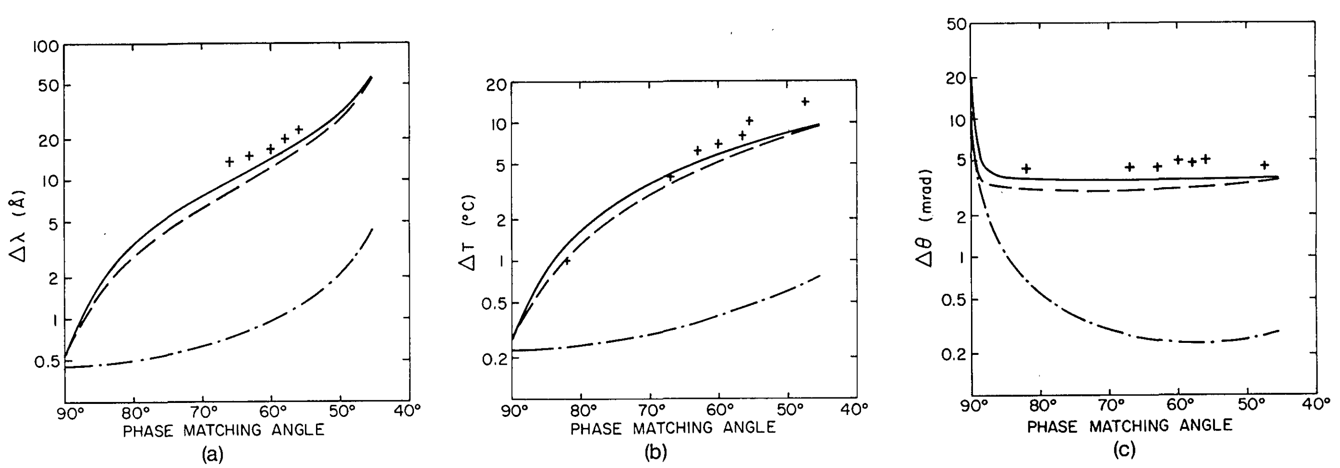

Fig. 1. Calculated and experimental dependence of the FWHM parametric bandwidth as a function of the phase-matching angle for a 25-mm long crystal of ADP: — calculated; beam focused with a 7.5-cm focal length lens; - - - calculated; beam optimally focused (see text); -·-·- calculated; unfocused beam. + Experimental. Beam focused with f = 7.5-cm lens. (a) Temperature; (b) wavelength; (c) angle.

efficiency decrease because of self-heating by high average power beams. The farther the phase-matching angle is from 90°, the higher the temperature bandwidth is, less temperature stabilization is needed, and the rate at which the wavelength can be scanned is higher because thermal inhomogeneities occurring in the crystal have no severe effect on the efficiency. It should be mentioned that for fast temperature scanning, crystals in the shape of thin rods or thin plates would have much better performance over bulky crystals.

IV. Conclusion

In summary, the hybrid of Boyd and Kleinman's focused Gaussian beam nonlinear interaction theory with a simple Taylor expansion approach to calculating parametric bandwidths produces reasonable results which agree with the observed bandwidths in angle-tuned SHG. The principal limitation to the extension of this technique to other nonlinear materials is the lack of reliable index of refraction data as a function of λ and T. This limitation is especially evident for the more exotic KDP isomorphs, such as ADA and RDP, which are so useful in doubling the longer wavelength portion of the visible spectrum. However, on the basis of strong chemical and crystal similarities, the first-order behavior of these more exotic materials can be expected to parallel closely the behavior of the better characterized ADP nonlinear material which has been calculated in this paper. Simple extensions of this approach would allow calculation of the parametric bandwidths for other materials both uniaxial and biaxial as well as for sum and differences frequency nonlinear interactions.

We thank R. Fletcher, S. Ketkar, and C. Pollock for their help in various parts of this work. This work was supported by the Department of Energy, the Robert A. Welch Foundation, and the National Science Foundation.

References

1. S. Blit, F. K. Tittel, and E. G. Weaver, J. Opt. Soc. Am. **67**, 1384 (1977).
2. N. Barnes and V. J. Corcoran, Appl. Opt. **15**, 696 (1976).
3. K. Kato, IEEE J. Quantum Electron. **QE-13**, 622 (1977).
4. J. Kuhl and H. Spitschan, Opt. Commun. **5**, 382 (1972).
5. Y. S. Lier, Appl. Phys. Lett. **31**, 187 (1977).
6. M. Okada and S. Ieiri, IEEE J. Quantum Electron. **QE-7**, 469 (1971).
7. G. D. Boyd and D. A. Kleinman, J. Appl. Phys. **39**, 3597 (1968).
8. E. Sahar, M. Sc. Thesis, Weizmann Institute of Science (1972).
9. F. Zernike, Jr., J. Opt. Soc. Am. **54**, 1215 (1964); **55**, 210 (1965).
10. R. A. Phillips, J. Opt. Soc. Am. **56**, 629 (1966).

Manual to Help Faculty Disclose and Develop Inventions

A manual for research administrators at colleges and universities to help the faculty members disclose and develop their inventions has been completed by Research Corporation, under a 1974 Research Management Improvement program grant from NSF and the National Bureau of Standards. The manual, *Stimulating Invention Disclosures by Faculty Researchers: A Guide for the University Intervention Administrator,* offers guidance to administrators wishing to establish or improve their in-house programs for helping faculty recognize patent potential of their inventions and overcome any misconceptions about university patents.

Based on a 3-year study at 8 selected academic institutions in the U.S., the 50-page report provides information about patent systems and the benefits of getting an invention patented. It offers suggestions on establishing a seminar designed to stimulate interest in invention within an institution. Copies are available from Dr. Willard Marcy, Research Corporation, 405 Lexington Ave., New York N.Y. 10017. Price is $5.25 per copy.