QUANTUM TECHNOLOGY, INC.
Quantum Park • 108 Commerce Street • Suite 101 / Lake Mary, Florida 32746-6212 / U.S.A.
Telephone: (407) 333-9348 / Cable: QUANTECH / Telex: 372-7798 / FAX: (407) 333-9352

DATA SHEET 714

SEPTEMBER 1989

SUM FREQUENCY MIXING AND SECOND HARMONIC GENERATION

Second Harmonic Generation and Sum Frequency Mixing as a Source of Tunable, Coherent Ultraviolet Radiation

Introduction

Dye lasers provide a convenient source of tunable radiation at wavelengths extending from the near infrared to the near ultraviolet. Tunable radiation at wavelengths below those directly attainable using dye lasers can be generated, using a suitable non-linear medium, either by frequency doubling the output of a dye laser operating in the visible, or by mixing the output of a dye laser with the output of a second laser. These techniques, referred to as second harmonic generation (SHG) and sum frequency mixing (SFM) respectively, have provided pulsed radiation at wavelengths extending down to 185 nm, and CW radiation at wavelengths down to 211 nm.

Efficient mixing of radiation at ω_1 and ω_2 to produce radiation at the sum frequency ω_3, where

$$\omega_3 = \omega_1 + \omega_2,$$

can only occur when the phase matching condition[1]

$$\omega_3 n_{\omega_3} = \omega_1 n_{\omega_1} + \omega_2 n_{\omega_2}$$

is satisfied, where n_ω is the refractive index of the non-linear medium at ω. SHG represents the special case where

$$\omega_1 = \omega_2 = \omega, \qquad \omega_3 = 2\omega$$

Phasematching can be achieved in many non-linear crystals with an appropriate choice of input beam polarizations, direction of propagation and crystal temperature because of their birefringence. Phasematching schemes are employed in which the interacting fundamental waves have both parallel and orthogonal polarizations. These are referred to as type I and type II phasematching respectively.

The expected output powers with focussed Gaussian input beams can be calculated using the theory developed by G.D. Boyd and D.A. Kleinman.[2] For unfocussed input beams the output power can be estimated using the plane wave coupled-amplitude equations.[1] Under small signal conditions, the output power P_{ω_3} is proportional to the product of the input powers, P_{ω_1} and P_{ω_2}. The conversion efficiency, $P_{\omega_3}/P_{\omega_1}P_{\omega_2}$, depends on many parameters including the non-linear coefficient of the medium, the direction of propagation of the interacting beams, the length of the non-linear crystal, and the linewidth and focussing of the input beams.[3]

Second Harmonic Generation

The phasematched generation of tunable SH radiation can be accomplished by tuning the input wavelength and adjusting either the direction of propagation of the input beam or the temperature of the non-linear crystal. These techniques are termed angle tuning and temperature tuning, respectively. In Figure 1 are shown, as a function of the angle of propagation of the input beam, the input wavelengths that yield phasematched type I or type II SHG in a number of non-linear crystals including potassium pentaborate (KB5),[4] lithium formate monohydrate (LFM) and urea,[5] and potassium dihydrogen phosphate (KDP) and its isomorphs. It is evident from Figure 1 that radiation spanning a broad range of UV wavelengths can be generated by SHG in an appropriate non-linear medium, although the SH conversion efficiency that can be realized depends on the fundamental wavelength. This results, in part, from differences in the non-linear coefficients for the different media. The non-linear coefficient for KB5 is, for example, much less than those of the KDP isomorphs.[4] Thus although KB5 will provide the shortest SH wavelengths, down to 217 nm, the conversion efficiency is low. Lithium formate monohydrate (LFM), will provide phasematched SHG down to ~230 nm.[6] In addition, for a given material, the SH conversion efficiency depends critically on the direction of propagation of the fundamental beam as a result of double refraction effects and the angle dependence of the effective non-linear coefficient of the medium.[1] For type I phasematched SHG in KB5 and the KDP isomorphs, angle tuning to longer wavelengths leads to a marked decrease in the SH conversion efficiency. In the case of KDP a crystal 2.5 cm long will, under optimum focussing conditions, provide a conversion efficiency of $\sim 10^{-3}\ W^{-1}$ at a phasematching angle θ of 90°, but only $\sim 5\ 10^{-5}\ W^{-1}$ at $\theta = 60°$. This decrease can, in the case of the KDP isomorphs, be avoided in certain instances by use of temperature tuning, which makes possible efficient 90° phasematched SHG in each material over a sizeable range of fundamental wavelengths. Ammonium dihydrogen phosphate (ADP) and ammonium dihydrogen arsenate (ADA) have particularly large 90° phasematched temperature tuning ranges. This is illustrated in Figure 2, which shows the fundamental wavelengths that produce 90° phasematched SHG in these materials as a function of crystal temperature.[7,8]

Sum Frequency Mixing

UV radiation can also be generated by the use of SFM. The wavelength combinations λ_1, λ_2 that produce efficient 90° phasematched type I SFM in room temperature ADA, ADP, and KDP,[8,9] together with those that produce phasematched SFM during propagation along the b axis of a KB5 crystal,[10] are shown in Figure 3. It is evident from Figure 3 that shorter output wavelengths can be generated in a given material by use of SFM than by direct SHG. Further, the directions of propagation chosen in Figure 3 provide the highest UV conversion efficiencies that can be achieved in these materials and, with an appropriate choise of λ_1 and λ_2, these conversion efficiencies can be maintained over an extended range of output wavelengths. A tunable output can also be generated by tuning just one of the input wavelengths and using either angle or temperature tuning to maintain phasematching. Again, ADP is particularly well suited to temperature tuning. To demonstrate this Figure 3 includes a shaded region which is bounded by the wavelength combinations λ_1 and λ_2 that produce 90° phasematched SFM in ADP at ±100°C. It is evident that, even with one input wavelength fixed, temperature tuning permits efficient SFM over a sizeable range of output wavelengths. In addition, temperature (and angle) tuning affords some flexibility in the wavelength combinations that may be used to generate a particular output wavelength.

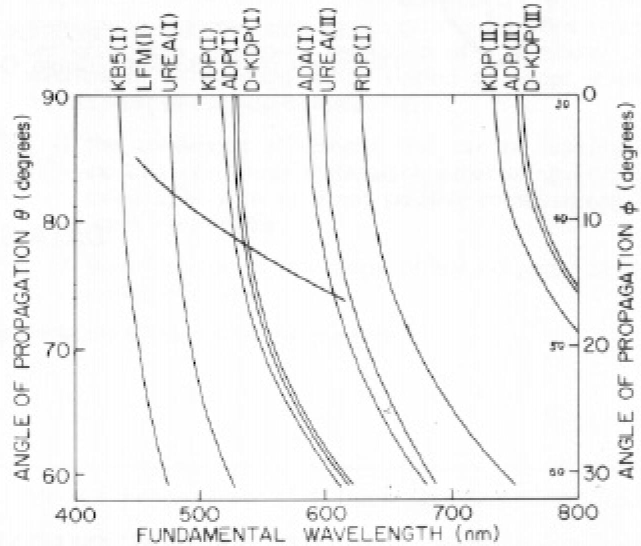

Figure 1

Angle tuning curves for type I and type II phasematched SHG in several non-linear crystals. The data for KB5 pertain to propagation in the ab plane at an angle ϕ to the b axis, the data for the KDP isomorphs to propagation at an angle θ to the optic axis.

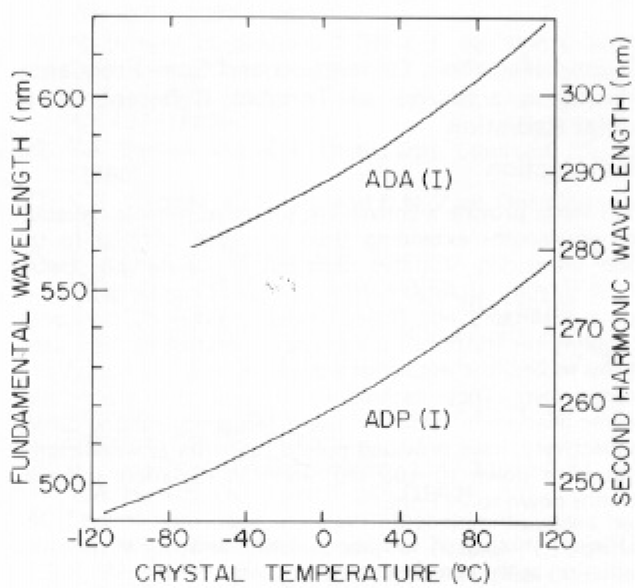

Figure 2

Temperature tuning curves for 90° phasematched type I SHG in ADP and ADA.

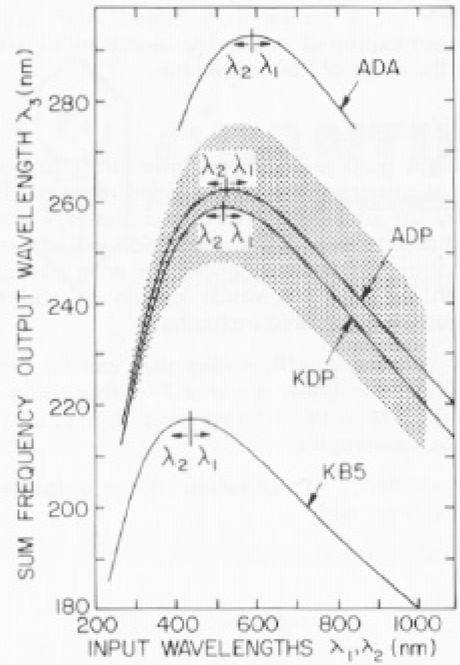

Figure 3

Wavelength combinations λ_1, λ_2 that provide phasematched type I SFM during propagation along the b axis of a KB5 crystal and 90° phasematched type I SFM in room temperature ADA, ADP, and KDP. The significance of the shaded region is discussed in the text.

Pulsed UV Generation

SHG techniques are widely used to provide pulsed, tunable UV radiation.[4-7,10] However, in certain cases radiation at wavelengths that can be generated by frequency doubling can be more efficiently generated by use of SFM, especially if Ruby or Nd/YAG pump lasers are employed. This is possible because SFM may permit better utilization of the pump laser outputs, or use of a more efficient non-linear medium or more favorable phasematching angle. For example, tunable radiation in the range 236-250 nm may be obtained at megawatt power levels by mixing, in a 90° phasematched, temperature-tuned ADP crystal, the second harmonic output of a Ruby laser and the output of an infrared dye laser pumped by the fundamental output of the laser.[11] This approach yields higher output powers than can be obtained by simply frequency doubling the output of a dye laser pumped by the SH of the Ruby laser because it allows the use of both outputs of the pump laser together with an efficient non-linear medium and favorable crystal orientation.

Radiation at wavelengths below those attainable by SHG has also been generated by SFM. UV radiation in the range 208-234 nm has been produced by mixing, in angle and temperature tuned ADP and KDP, the fundamental output of a Nd/YAG laser and the output of a frequency doubled dye laser.[12] Wavelengths as short as 185 nm have been obtained by mixing in KB5.[10] Although this material has a small non-linear coefficient, its high damage threshold permits the use of large input power densities, and, in consequence, sizeable output powers can be obtained.

CW UV Generation

Tunable CW UV radiation can be conveniently generated by frequency doubling the output of a CW dye laser using an angle or temperature tuned non-linear crystal located external to the laser cavity. The resultant UV powers, typically ~0.1 to 1.0 mW, are sufficient for many spectroscopic applications. However, recent experiments have shown that much larger output powers can be obtained by locating the frequency doubling crystal at an auxiliary focus within the laser cavity, thereby taking advantage of the high circulating power in the cavity. Although initial investigations were undertaken using standing wave lasers, most recent studies employ traveling wave, ring dye lasers as these provide easier mode discrimination and higher circulating powers. Radiation tunable from 314 ro 318 nm, with output powers of ~15 mW has been obtained by intracavity SHG using a Rhodamine B laser.[13] Somewhat larger output powers, and a tuning range of 285 to 311 nm have been obtained by intracavity SHG using Rhodamine 6G (Rh6G) lasers,[8,14-20] and by cavity enhanced external frequency doubling.[21] Radiation at wavelengths below 270 nm has been generated by intracavity SHG using Coumarin dye lasers, but the output powers obtained were small, typically ~0.1 mW.[22,23]

CW radiation tunable across a broad UV wavelength range can be simply generated by mixing the output of a commercial dye laser with the various output lines from a krypton or argon ion laser in a non-linear crystal located outside both laser cavities. Because ion lasers provide lines at wavelengths between the ultraviolet and infrared, a wide range of sum frequency wavelengths can be obtained without changing the dye or optics in the dye laser and without use of short lived or less efficient dyes. This is illustrated in Figure 4 which shows the output tuning ranges that can be obtained by mixing the output of a Rh6G or Oxazine 1 dye laser with selected argon or krypton ion laser lines. For Oxazine 1 the tuning range extends down to ~226 nm, and for Rhodamine 6G extends from 211 nm to 406 nm.

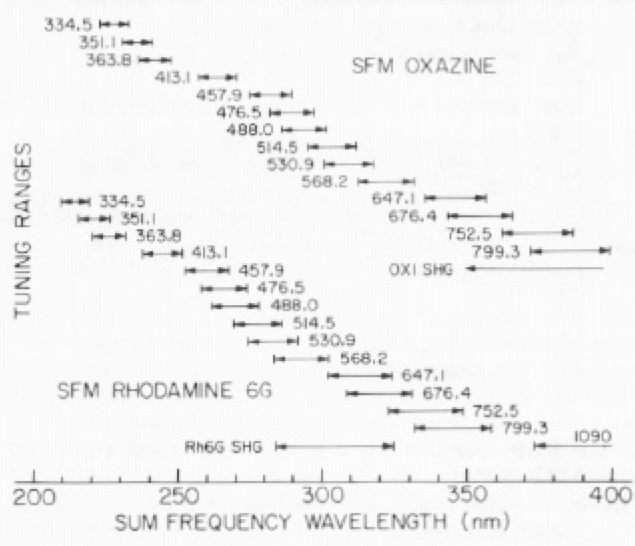

Figure 4

Tuning ranges that can be realized by mixing the output of a Rh6G or Oxazine I laser with selected ion laser lines.

CW radiation tunable from 257 to 357 nm has been obtained, using a Rh6G laser, by extracavity SFM in angle tuned crystals of KDP, ADP, ADA and rubidium dihydrogen phosphate.[24] Given the output powers available from Rh6G and ion lasers, this technique provides output powers $\gtrsim$0.1 mW over much of this tuning range. Tunable radiation at ~247 nm has been generated by mixing the 413 nm line from a krypton ion laser with radiation from a Rh6G laser in a temperature tuned ADP crystal. UV output powers of ~ 1 mW were obtained,[25] much greater than could be realized by extracavity SHG using a Coumarin dye laser. Radiation tunable from 211 to 215 nm has been obtained by mixing, in KB5, the output of a Rh6G laser with the 334.5 nm line of an argon ion laser.[26] Radiation extending to about 233 nm can be produced by mixing with other UV argon ion laser lines. Although the conversion efficiencies are low, they are sufficient to provide output powers of ~50–100 nanowatt.

Significant increases in output power can be obtained by use of intracavity SFM, the mixing crystal being located either in the primary laser cavity itself[27] or in an external, frequency-locked passive ring cavity.[28] Although these techniques have not, as yet, been widely applied, intracavity SFM in a temperature tuned ADP crystal has already been exploited to provide several milliwatts of UV power in the range of 254 to 268 nm.

Summary

As is evident from the foregoing discussion, tunable radiation can be obtained over an extended range of UV wavelengths by use of SHG and SFM techniques. Frequently a given pump laser/dye laser combination will afford a number of options as regards generation of a particular UV wavelength. In selecting which option to adopt several factors must be considered including:

i) the conversion efficiencies that can be realized in existing non-linear materials[29] either using SHG or using SFM with different possible combinations of input wavelengths,

ii) the efficiency of utilization of the output(s) of the pump laser, and

iii) the efficiency of the dye laser.

References

1. F. Zernike and J.E. Midwinter, Applied Nonlinear Optics, A.A. Ballard, ed. (Wiley, New York, 1973)
2. G.D. Boyd and D.A. Kleinman, J. Appl. Phys. 39, 3597 (1968)
3. S. Blit, E.G. Weaver, and F.K. Tittel, Appl. Opt. 18, 733 (1979)
4. H.J. Dewey, I.E.E.E. J. Quant. Elect. QE12, 304 (1976) C.F. Dewey Jr., W.R. Cook Jr., R.T. Hodgson, and J.J. Wynne, Appl. Phys. Lett. 26, 714 (1975)
5. J.M. Halbout, S. Blit, W. Donaldson, and C.L. Tang, I.E.E.E. J. Quant. Elect. QE15, 1176 (1979)
6. S. Saikan, D. Ouw, and F.P. Schafer, Appl. Optics 18, 193 (1979) F.B. Dunning, F.K. Tittel, and R.F. Stebbings, Opt. Commun. 7, 181 (1973)
7. R.S. Adhav and R.W. Wallace, I.E.E.E. J. Quant. Elect. QE9, 856 (1973)
   D.A. Jennings and A.J. Varga, J. Appl. Phys. 42, 5171 (1971)
   R.K. Jain and T.K. Gustafson, I.E.E.E. J. Quant. Elect. QE12, 555 (1976)
8. C.M. Marshall, R.E. Stickel Jr., F.B. Dunning, and F.K. Tittel, Appl. Optics 19, 1980 (1980)
9. F. Zernicke Jr., J. Opt. Soc. Am. 54 1215 (1964); 55, 210 (1965)
10. R.E. Stickel Jr. and F.B. Dunning, Appl. Optics 17, 981 (1978);16, 2356 (1977); 15, 3131 (1976)
    K. Kato, Appl. Phys. Lett. 30, 583 (1977), see also J.A. Paisner, M. L. Spaeth, D.C. Gerstenberger and I.W. Ruderman, Appl. Phys. Lett. 32, 476 (1978)
11. R.E. Stickel Jr. and F.B. Dunning, Appl. Optics 17, 1313 (1978)
12. G.A. Massey and J.C. Johnson, I.E.E.E. J. Quant. Elect. QE 12, 721 (1976)
13. S. Runge and N. Wolffer, Opt. Commun. 32, 489 (1980)
14. H.W. Schröder, L. Stein, D. Frölich, B. Fugger, and H. Welling, Appl. Phys. 14, 377 (1977)
15. A.I. Ferguson and M.H. Dunn, Opt. Commun. 23, 177 (1977)
16. C.E. Wagstaff and M.H. Dunn, J. Phys. D 12, 355 (1979)
17. B. Couillaud, L.A. Bloomfield, J.E. Lawler, A. Seigel, and T.W. Hansch. Opt. Commun. 35, 359 (1980)
18. E.R. Eliel, W. Hogervorst, K.A.H. Van Leeuwen, and B.H. Post, Opt. Commun. 36, 366 (1981); 39, 41 (1981)
19. S. Runge, A. Pesnelle, M. Perdrix, D. Sevin, N. Wolffer, and G. Watel, Opt. Commun. 42, 45 (1982)
20. T.F. Johnson Jr., R.H. Brady, and W. Proffitt, Appl. Optics 21, 2307 (1982)
21. M. Brieger, H. Büsener, A. Hese, F. van Moers, and A. Renn, Opt. Commun. 38, 423 (1981), see also J.C. Bergquist, H. Hemmati, and M. Itano, Opt. Commun. 43, 437 (1982)
22. S.J. Bastow and M.H. Dunn, Opt. Commun. 35, 259 (1980)
23. C.R. Webster, L. Wöste, and R.N. Zare, Opt. Commun. 35, 435 (1980)
24. S. Blit, E.G. Weaver, F.B. Dunning, and F.K. Tittel, Opt. Lett. I, 58 (1977); Appl. Optics 17, 721 (1978)
25. R.P. Mariella Jr., Opt. Commun. 29, 100 (1979)
26. R.E. Stickel Jr., S. Blit, G.F. Hildebrandt, E.D. Dahl, F.B. Dunning, and F.K. Tittel, Appl. Optics 17, 2270 (1978)
27. Enquan Liu, F.B. Dunning, and F.K. Tittel, Appl. Optics 21, 3415 (1982)
28. B. Couillaud, Ph.Dabkiewicz, L.A. Bloomfield, and T.W. Hansch, Opt. Lett. 7, 265 (1982)
29. Several non-linear materials other than discussed here may also be considered, see, for example, L. Schüler, K. Betzler, H. Hesse, and S. Kapphan, Optics Commun. 43, 157 (1982); J.M. Halbout, and C.L. Tang, I.E.E.E. J. Quant. Elect. QE18, 410 (1982)
30. Quantum Technology Inc., Technical Data Sheets 701, 702, 703, 704, 706, 707, 710, 711, 712, 713, 723, 724