# EXTENDING THE WAVELENGTH RANGE OF FUNDAMENTAL LASER SOURCES

R. A. POPPER, T. NOWICKI, W. RUDERMAN, AND J. RAGAZZO,
*Interactive Radiation, Inc., Northvale, New Jersey 07647 U. S. A.*

The ability to extend the wavelength range of fundamental laser sources provides the opportunity to match a light source to a specific chemical or biological interaction wavelength. The most commonly used technique to alter laser wavelengths involves mixing two optical waves in a nonlinear crystal to produce the sum frequency. Energy is efficiently transferred from the two input waves to the output wave only when a proper phase-matching condition is maintained. Special cases arise when the two input waves have the same optical frequency (i.e., are components of the same wave) or have in themselves a harmonic relationship with a common fundamental source. The first instance results in an output wavelength equal to exactly one-half that of the fundamental input and is called second harmonic generation (SHG). In the second instance, combining the fundamental and second harmonic of a laser will result in third harmonic generation (THG). THG is a two-stage process, a doubling stage followed by a mixing stage.

Phase matching is accomplished by utilizing the birefringence of the nonlinear crystal to overcome the refractive index dispersion from one wavelength to another. The indices are precisely tuned to the values desired for a specific set of wavelengths either by altering the optical propagation direction with respect to the crystal optic axis (angle matching) or by changing the temperature of the crystal and utilizing the subsequent index change (temperature matching). Whichever method is used, the physical properties of individual crystal species impose a range limitation on phase matching that requires that several crystals be used for broad wavelength coverage.

Laser sources used in the study of fast reactions must be characterized by short pulse widths. In addition, to achieve maximum efficiency in harmonic generating and mixing applications, the laser should have a narrow line width, high peak power, and low beam divergence. Spectral brightness ($W/cm^2$ sterad Å) is a single parameter relating the three variables. For efficient harmonic generation and mixing, the laser must exhibit a high spectral brightness. Among the more common and flexible lasers that exhibit high spectral brightness are Nd:YAG, ruby, $N_2$-pumped dye lasers and Nd:YAG-pumped dye lasers. Techniques used to generate short-pulse outputs include Q-switching and mode-locking.

A wavelength region of high interest to biologists, chemists, physicists, and spectroscopists is the ultraviolet (UV), since this is a region of high interaction of light with molecules. Several techniques are employed to extend the wavelength range of tunable or fixed-frequency laser sources to the ultraviolet. SHG is the most direct approach. In those cases where the desired wavelength regions fall within the umbrella of 90° phase-matching of KDP homologues (see figure), an extremely efficient tem-

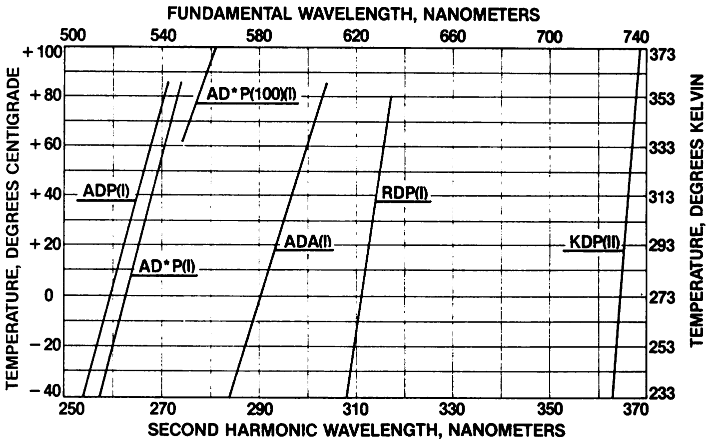


FIGURE 1

perature-tuned SHG approach may be used. Where this is not possible less efficient critical phase matching (angle-tuned) SHG techniques are available.

Mixing techniques frequently offer a great improvement in efficiency over conventional SHG approaches. For example, using a temperature-tuned ammonium dihydrogen arsenate crystal (ADA) to mix the output of a dye laser pumped by the second harmonic of a Nd:YAG laser with that same second harmonic has resulted in highly efficient conversion from 285.1 nm to 302.8 nm. The nonlinear crystal potassium pentaborate (KPB) makes it possible to obtain intense tunable UV radiation down to 196.6 nm by mixing the output of the fourth harmonic (266 nm) of a Nd:YAG laser and a near-infrared dye laser pumped by the second harmonic (532 nm) of the same YAG laser. Five ns pulses with peak powers of 40 kW at 10 pulse/s at 196.6 nm have been achieved.

Parametric conversion (difference-frequency) techniques are used for generating continuously variable wavelengths longer than those of the pump pulses. Continuously tunable radiation from 660 to 2,750 nm can be obtained by parametric conversion of 532 nm radiation (the second harmonic of a mode-locked Nd:YAG laser) in lithium iodate. Pulse widths as short as 2–3 ps with pulse energies of 50 $\mu$J are achievable. In addition, the fourth harmonic (266 nm) of Q-switched or mode-locked Nd:YAG lasers can pump an ammonium dihydrogen phosphate (ADP) parametric amplifier to produce nanosecond or picosecond pulses at visible wavelengths. A synchronous mode-locked tunable dye laser using a frequency-doubled mode-locked Nd-YAG laser pump can be sum and difference mixed with the 1,064-nm and 532-nm pump pulses in lithium iodate, KDP, and ADP. This results in tunable picosecond pulse generation in the ultraviolet from 270 to 432 nm and in the infrared from 1,130 to 5,600 nm.