Reprinted from:
JOURNAL OF APPLIED PHYSICS VOLUME 42, NUMBER 12 NOVEMBER 1971

Efficient Second Harmonic Generation in ADP with Two New Fluorescein Dye Lasers

D. A. Jennings
National Bureau of Standards, Boulder, Colorado 80302

and

A. J. Varga
Ford Scientific Laboratory, Dearborn, Michigan 48121
(Received 15 March 1971; in final form 7 June 1971)

This paper reports two new solutions of fluorescein dyes for flashlamp-pumped dye lasers. Lasers using these dyes have peak powers on the order of kilowatts and are tunable in the 550–580- nm range. Efficient 90° phase-matched second harmonic generation demonstrated with these and other dyes provides a flash-excited uv source tunable from 250 to 290 nm with peak powers on the order of 10 W.

The laser head used in this experiment contained an elliptical cavity with a water-cooled linear flashlamp and a 9 cm × 1 mm i.d. circulating-dye cell at the foci. A 0.1-μF 20-kV low inductance capacitor was discharged through a thyratron to fire the lamp. The electrical input to the lamp was typically 11 J and the flash was about 1 μsec in duration. Mirrors of various reflectances and a 1800 line/mm grating were used to terminate the cavity. Second harmonic generation was accomplished in ADP with the fundamental beam polarized perpendicular to the z axis and propagating at 45° with respect to the x and y axes. The crystal was mounted in a heat sink whose temperature could be varied at 200°C.

Solutions of sodium fluorescein in methanol were prepared in the range 10^{-4}–10^{-2} M. The neutral solutions did not lase when flash-pumped in the above system. The dye solutions in the range above 5×10^{-4} M did lase, however, when they were made alkaline by saturating them with slightly soluble ($< 10^{-4}$ M) Na_2CO_3. Fluorescein converts from the mono-anion form in neutral solution to the di-anion form in alkaline solution.[1] The conversion was verified by the appearance of a strong absorption peak at 496 nm in the absorption spectrum of the solution. The laser output power obtained from the alkaline sodium fluorescein was also improved a factor of 10 by the addition of cyclo-octatetraene (COT) ~ 10^{-2} M.[2] The addition of COT alone to the neutral sodium fluorescein-methanol solution did not allow lasing of the dye. The alkalinity of the solutions was also adjusted with other bases holding the fluorescein and COT concentrations constant. Added NaOH and KOH, for example, up to 10^{-2} M did not result in increased laser power. Much higher base concentration produced some degradation of the output.

Typical results for 3×10^{-4} M sodium fluorescein 10^{-2} M COT Na_2CO_3-saturated methanol solution were a 3.2-J (input to flashlamp) threshold with 70% and 100% reflectors on the cavity. With a grating and a 70% reflector, a tuning range of 549–574 nm was realized. An 11-J input to the flashlamp resulted in a peak laser output of 2 kW corresponding to an energy conversion of 1%.

Similar results were also found with solutions of 2′, 7′ dichlorofluorescein (DCF). A 5×10^{-4} M DCF 2×10^{-2} M COT Na_2CO_3-saturated solution was found to be the optimum for peak power generation with a grating in the cavity. The laser output was 2.6-kW peak with a 0.6-μsec half-width for an 11-J flashlamp input. The tuning range was from 557 to 581 nm. Representative solutions had a threshold under 4 J with 70% and 100% reflectors on the cavity. It was also noted that in the DCP solutions COT had the effect of lengthening the laser pulse in addition to increasing the peak power.

For comparison with these dyes, water and ethanolic solutions of Rhodamine 6G were lased in this system. With 11-J input to the flashlamp the laser exhibited 4% power conversion and 8-W peak power. The usual tuning range, 570–610 nm, was observed. The addition of COT to these dye solutions produced no detectable change in the power or duration of the output pulse. Also, attempts to obtain laser action in the range 560—570 nm by changing solvents and varying the pH were unsuccessful. The Rhodamine 6G solutions exhibited a peak power about four times that obtained with the above fluorescein solutions for similar excitation conditions.

Efficient second harmonic generation was achieved in ADP with 90° phase matching. This condition provides maximum conversion lengths by eliminating walkoff between the ordinary fundamental and extraordinary second harmonic beams.[3] Of the two nonlinear crystals transparent in the uv region of interest, ADP and KDP, ADP has the larger thermal coefficient of birefringence, $-6.2\times10^{-5}\,{}^\circ K^{-1}$ vs $-1.5\times10^{-5}\,{}^\circ K^{-1}$ [4] and allows 90° phase-matching over a broader spectral range. Measurements of the temperature for 90° phase match in ADP as a function of wavelength are shown in Fig. 1. It is seen from this figure that efficient conversion to wavelengths from 290 to below 250 nm is possible.[5,6] The limits are determined by the inability to bring the crystal to a temperature which allows phase matching. Temperatures above those corresponding to the long-wavelength limit produce thermal decomposition of the ADP.[5] At short wavelengths the rapidly decreasing difference between $n_0(\omega)$ and $n_e(2\omega)$ cannot be compensated with indefinitely lower temperatures.

The second harmonic conversion efficiency of the dye laser–ADP system was measured experimentally and compared with the efficiency calculated from

$$\eta = P_2/P_1 = KP_1 l^2/\omega_0^2, \tag{1}$$

where[3]

$$K = (218\pi^2\omega_1^2/c^3 n_1^2 n_2^2)d^2 \ . \tag{2}$$

P_1 and P_2 are the fundamental and second harmonic

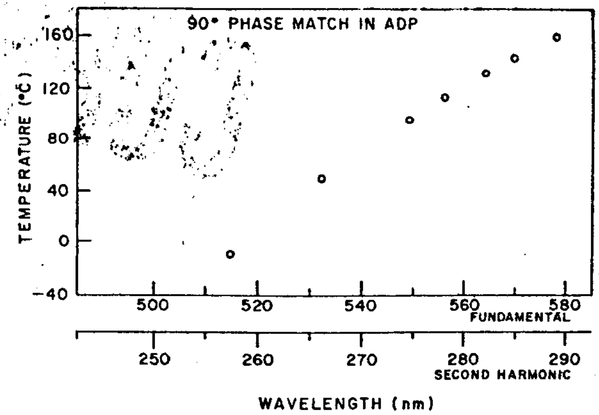

Fig. 1. Temperatures vs wavelength for 90° phase-matched second harmonic generation in ADP. The points at 530 and 514.5 nm were taken from Refs. 5 and 6, respectively.

power in erg/sec; l is the crystal length, 4 cm; w_0 is the beam waist diameter, about 0.2 mm; ω_1 is the frequency of the fundamental; n_1 and n_2 are the appropriate refractive indices for the fundamental and second harmonic; and $d = 1.3\times10^{-9}$ esu is the nonlinear coefficient for ADP. The measured second harmonic conversion efficiency was found to be linear within a factor of 2 for fundamental powers varying over two decades. 400 W at 564 nm produced 6.4-W peak harmonic power at 282 nm.

The above methods add the 250–290-nm range to the 340–709-nm range already reported accessible to flashlamp-pumped, tunable dye lasers.[7-10]

[1] L. Lindqvist, Arkiv Kemi 16, 79 (1960).
[2] R. Pappalardo, H. Samelson, and A. Lempicki, Appl. Phys. Letters 16, 267 (1970).
[3] G. D. Boyd and D. A. Kleinman, J. Appl. Phys. 39, 3597 (1968).
[4] M. Yamazaki and T. Ogawa, J. Opt. Soc. Am. 56, 1407 (1966).
[5] B. G. Huth and Y. C. Kiang, J. Appl. Phys. 40, 4976 (1969).
[6] M. W. Dowley and E. B. Hodges, IEEE J. Quantum Electron. 4, 552 (1968).
[7] P. P. Sorokin and J. R. Lankard, IBM J. Res. Develop. 11, 148 (1967).
[8] B. B. Snavely, Proc. IEEE 57, 1374 (1969).
[9] H. W. Furumoto and H. L. Ceccon, IEEE J. Quantum Electron. QE-6, 262 (1970); J. Appl. Phys. 40, 4204 (1969).
[10] J. B. Marling, D. W. Gregg, and S. J. Thomas, IEEE J. Quantum Electron. QE-6, 572 (1970).