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## Efficient intracavity second harmonic generation (I.S.H.G.) using the optimal pump laser focusing method

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**Résumé.** — Une analyse détaillée de la Génération du Second Harmonique Intracavité pour des lasers à faible gain et pour des dye lasers continus en anneau a été effectuée en utilisant la méthode de focalisation optimale du faisceau laser. On a déduit des expressions qui concernent les conditions optimales de focalisation, la longueur optimale du cristal, la puissance de sortie maximale de l'onde du second harmonique et la « transmission non linéaire » des miroirs laser. On a testé expérimentalement la théorie et on obtient ainsi une source de radiation ultraviolette commode pour l'utilisation en spectroscopie.

**Abstract.** — A detailed analysis of Intracavity Second Harmonic Generation (I.S.H.G.) for small-gain and cw-ring dye lasers is given which includes the optimal laser beam focusing method as an integral part. Expressions are derived for optimal focusing conditions, the optimal crystal length, maximal output power of the second harmonic wave and « non-linear transmission » of the laser mirrors. Experimental work is presented to test the theory. An efficient source of u.v. radiation can be realized for spectroscopic applications.

### 1. Introduction.

In recent years, Second Harmonic Generation (S.H.G.) has proved to be a very useful method to generate intense and tunable ultraviolet radiation. For high resolution spectroscopy where continuous radiation is required, it has been shown that a high efficiency harmonic generation can be obtained by inserting the non-linear element into the cavity of the continuous wave laser [1].

The problem of Intracavity Second Harmonic Generation (I.S.H.G.) has been extensively treated [2-4]. Smith [2] studied the case of a small-gain laser with a homogeneously broadened laser transition. In this treatment the intracavity non-linear loss and the plane uniform wave have been used.

Ferguson and Dunn [3] have analysed I.S.H.G. in cw-dye laser, taking into account the thermal absorption with associated thermal phase-mismatching effects and the excited state absorption in the dye. In particular, the role of the crystal length has been studied.

They have concluded that it is not desirable to use a crystal which is as long as possible and that there is an optimal value of the crystal length which depends on the losses of the laser cavity. However, they utilized the formula given by Teschke *et al.* [5] for the operation of dye lasers in the absence of a non-linear crystal to calculate the intracavity dye laser and threshold power. To determine the S.H.G. power, the authors have also resorted to the formulae given by Boyd and Kleinman [6] in the case of the non-linear crystal being located outside the laser cavity. Recently, Kane *et al.* [7] have studied stable behaviour and the dimension of the beam waist in the ring dye laser cavity using Gaussian geometrical optics. However, the non-linear interaction effect coupled with the laser effect in the active medium is not taken into account.

To date, there has been no comprehensive study of I.S.H.G. using the Gaussian pump wave with directly coupled laser medium and parametric interaction.

Gabel and Hercher placed the Second Harmonic Generation crystal inside the cavity of a cw-dye laser. Several other groups continued this work including Hänsch *et al.* and Tittel *et al.* [8]. Many experiments have been carried out which produced very different u.v. powers [9-13].

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Webster *et al.* [10] obtained a maximum power of 120  $\mu\text{W}$  at 250 nm with a single mode ring dye laser using a non critical  $90^\circ$  phase-matching ADP crystal of length 5 cm, whereas Majewski [11] obtained 15 mW at 293 nm using a non phase-matching 0.05 cm thick  $\text{LiIO}_3$  crystal. Eliel *et al.* [12] arrived at a maximum of 25 mW u.v. power using a phase-matching ADA crystal of 2 cm long inside a single mode ring dye laser cavity. Bouvier *et al.* [13] obtained similar results with the same crystal of 1.8 cm long. In all these experiments, nothing is regarded concerning the optimal configuration of the I.S.H.G. system.

In this paper, we present a model for cw-I.S.H.G. using the optimal focusing method. The laser medium manifests here a homogeneously broadened gain transition. The treatment was performed with a Gaussian eigenmode of a coupled laser which is either small gain-(cw-YAG  $\text{Nd}^{3+}$  laser) or cw-dye laser. To produce an u.v. source suitable for spectroscopic uses and to apply our theory in a concrete practical case, we carried out an investigation of the efficiency of I.S.H.G. in a cw ring dye laser using a temperature phase-matched ADA crystal; we also investigated the variation of S.H.G. power with respect to the second harmonic generation wavelength. We will discuss the optimization of two parameters in the system, I.S.H.G. resonator configuration and crystal length. A scheme of I.S.H.G. configuration assumed for the mathematical model is shown in figure 1. The pump (laser) field is considered to be a fundamental Gaussian mode of the highly resonant structure  $M_1 M_2$  and propagates many times through the non-linear crystal. The transmission of the mirrors  $M_1, M_2$  is 100 % for the S.H. wave. Thus the laser and the parametric interaction are directly coupled.

## 2. Theory.

**2.1 EQUATIONS OF MOTION.** — We consider the pump mode of a highly resonant cavity at frequency  $\omega_1$ .  $\omega_2$  is the frequency of S.H.G. wave, satisfying the relation  $\omega_2 = 2\omega_1$  for energy conservation. The method of derivation is similar to that in our previous paper [14] but, in this case, is applied to S.H.G. The total field has the form

$$\mathbf{E}(\mathbf{r}, t) = \sum_{j=1,1,2} E_j(t) \mathbf{e}_j \cos(\omega_j t + \phi_j(t)) U_j(\mathbf{r}) \quad (1)$$

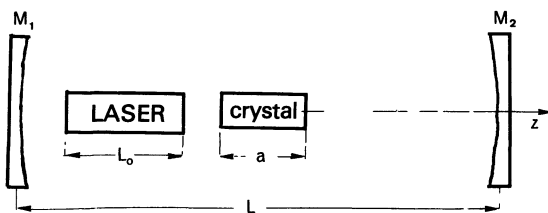


Fig. 1. — Diagram of intracavity second-harmonic generation configuration taken for the mathematical model.

where  $\mathbf{e}_j$  is the polarization unit vector,  $U_j(\mathbf{r})$  the spatial mode and  $E_j(t)$  the peak electric field (V/m). If only the  $\text{TEM}_{00}$  mode of the pump laser is resonant, we can write  $U_1(\mathbf{r})$  as a sum of right- and left-travelling Gaussian beams, i.e.

$$U_1(\mathbf{r}) = u_1^*(\mathbf{r}) + u_1(\mathbf{r}). \quad (2)$$

We shall consider the case in which the pump laser beam is an extraordinary wave with walk-off angle  $\rho = 0$  whereas the S.H. wave is an ordinary wave. Boyd and Kleinman [6] have shown that the result is the same in the inverse case. These latter wrote the one-way travelling pump laser wave with the Gaussian form

$$E_1(\mathbf{r}, t) = E_1(t) u_1(\mathbf{r}) = E_1(t) \frac{\exp(jk_1 Z)}{1 + j\tau} \times \exp\left\{-\frac{X^2 + Y^2}{W_0^2(1 + j\tau)}\right\} \quad (3)$$

with the centre of the non-linear crystal located at the origin of a Cartesian coordinate system  $(X, Y, Z)$ , centred in the laser cavity with the  $Z$ -axis along the longitudinal axis

$$\tau = \frac{2Z}{b}$$

and  $W_0$  is the Gaussian beam waist of the pump wave in the crystal centre;  $b (= k_1 W_0^2)$  the confocal parameter and  $k_1 = n\omega_1/C$  the wave vector of the pump wave;  $n_1$  refractive index. The amplitude of the S.H. wave can be determined using equation (3) together with the parabolic equation deduced in previous work published by one of us [14, 15]

$$\left[\frac{\partial}{\partial Z} - \frac{j}{2k_2} \left(\frac{\partial^2}{\partial X^2} + \frac{\partial^2}{\partial Y^2}\right)\right] E_2(X, Y, Z, t) = j\gamma E_1^2(X, Y, Z, t) e^{-j\Delta k Z}. \quad (4)$$

The coupling constant  $\gamma$  is

$$\gamma = \frac{4\pi\omega_2 d_{\text{eff}}}{Cn_2} \quad (5)$$

where  $d_{\text{eff}}$ , the non-linear effective susceptibility, is given by

$$2d_{\text{eff}} = \chi \quad (6)$$

and the phase-mismatching is

$$\Delta k = 2k_1 - k_2. \quad (7)$$

The method exposed in [14] allows us with the help of Green's function, to obtain the solution of the equa-

tion (4)

$$E_2(X, Y, Z, t) = j\gamma \frac{E_1^2(t)}{1 + j\tau} e^{j2k_1 Z} \times \int_{-a/2}^Z dZ' \frac{\exp \Delta k Z'}{1 + j\tau'} \exp \left\{ -\frac{2(X^2 + Y^2)}{W_0^2(1 + j\tau')} \right\} \quad (8)$$

where  $a$  is the crystal length.

The expression (8) defines the field of the S.H. wave and agrees with the result given by Boyd and Kleinman [6], showing the spatial mode of the S.H. wave

$$U_2(r) = j \frac{e^{jk_2 Z}}{1 + j\tau} \int_{-a/2}^Z dZ' \frac{e^{j\Delta k Z'}}{1 + j\tau'} \times \exp \left\{ -2 \frac{X^2 + Y^2}{W_0^2(1 + j\tau')} \right\}. \quad (9)$$

Taking into account polarizations in the laser medium and in the non-linear crystal, using the treatment given in [14], Lamb's equation [16] and the expressions (1), (2), (3), (8) and (9) we have the following equation of motion :

$$\dot{E}_1(t) = -\frac{1}{2} \frac{\omega_1}{Q_1} E_1(t) - \frac{1}{2} \omega_1 \chi_1'' E_1(t) - \frac{1}{2} \cdot \frac{\omega_1}{\epsilon_1^N} \cdot \frac{\gamma}{8} \cdot \frac{a}{L} \chi b E_1^3 h^{(2)}(\sigma, \xi) \quad (10)$$

$$\dot{\phi}_1(t) = \frac{\omega_1 \chi_1'}{2} + \frac{1}{2} \cdot \frac{\omega_1}{\epsilon_1^N} \cdot \frac{\gamma}{8} \cdot \frac{a}{L} \chi b E_1^2 h^{(2)'}(\sigma, \xi). \quad (11)$$

Here  $Q_1$  is the cavity quality factor for the pump laser mode ;  $\epsilon_1^N$  dielectric constant for the non-linear crystal ;  $\chi_1''$  accounts for the gain and saturation due to the laser medium ;  $\chi_1'$  results in frequency shifts and mode pulling and pushing effects ;  $h^{(2)}(\sigma, \xi)$  and  $h^{(2)'}(\sigma, \xi)$ -focusing functions defined by

$$h^{(2)}(\sigma, \xi) = \Re e(2\xi)^{-1} \times \int_{-\xi}^{+\xi} d\tau \int_{-\xi}^{\tau} d\tau' \frac{e^{-j\sigma(\tau-\tau')}}{(1-j\tau)(1+j\tau')} \quad (12)$$

$$h^{(2)'}(\sigma, \xi) = \Im m(2\xi)^{-1} \times \int_{-\xi}^{+\xi} d\tau \int_{-\xi}^{\tau} d\tau' \frac{e^{-j\sigma(\tau-\tau')}}{(1-j\tau)(1+j\tau')} \quad (13)$$

with

$$\xi = \frac{a}{b}, \text{ the focusing parameter}$$

$$\sigma = \frac{\Delta k b}{2}, \text{ the phase-mismatching parameter}$$

$L$  the pump laser cavity length.

## 2.2 INTRACAVITY S.H.G. IN SMALL-GAIN LASER. —

For the case of a small-gain laser (cw-YAG Nd<sup>3+</sup> laser), using (10), (11) and the treatment described in [14] we have the following equations of motion :

$$\frac{dP_1}{d\tau_1} = \left[ -\alpha_1 + g_0 \left( 1 - \frac{16\beta}{Cn_1 W_0^2} P_1 \right) - \frac{128\pi^2 \omega_1^2 d_{\text{eff}}^2 a}{C^3 n_1^2 n_2} \cdot \frac{b}{W_0^2} h^{(2)} P_1 \right] P_1 \quad (14)$$

$$\frac{d\phi_1}{d\tau_1} = -\frac{\omega_1 L}{2C} \chi_1' - \frac{64\pi^2 \omega_1^2 d_{\text{eff}}^2 a}{C^3 n_1^2 n_2} \cdot \frac{b}{W_0^2} h^{(2)'} P_1 \quad (15)$$

where  $\alpha_1 = (L/C)(\omega_1/Q_1)$  is the single-pass power loss for the pump laser mode and  $\tau_1$  a new time variable defined by  $Ct/L$  ;  $g_0$  the single-pass unsaturated power gain and  $\beta$  a parameter accounting for the effect of saturation.  $P_1 (= Cn_1 W_0^2/16 \times |E_1|^2)$  is the pump laser power. We see that the equations (14), (15) illustrate the time dependence of the power and phase of the pump laser wave. Equation (8) defines the power of the S.H. wave when all the characteristics of the pump laser wave are known. In general, the equations (14), (15) and the function  $h^{(2)}(\sigma, \xi)$  cannot be solved analytically, but only by means of computers. The function  $h^{(2)}(\sigma_{\text{opt}}, \xi)$  is designated as a focusing function for calculating the optimal S.H.G. power, and can be solved in the particular case  $\xi \ll 1$  where it assumes the form

$$h^{(2)}(\sigma, \xi) \underset{\xi \ll 1}{=} \frac{1 - \cos 2(\sigma - 1)\xi}{2(\sigma - 1)^2 \xi} = \xi \left[ \frac{\sin(\sigma - 1)\xi}{(\sigma - 1)\xi} \right]^2 \quad (16)$$

or  $h^{(2)} \simeq \xi$  at  $\sigma_{\text{opt}} = 1$ .

The function  $h^{(2)}(\sigma_{\text{opt}}, \xi)$  has been calculated by a computer in the general case and the results are plotted in figure 2.

### Discussion.

In steady-state regime we have

$$-\alpha_1 + g_0 - \frac{16g_0\beta}{Cn_1 W_0^2} P_1 - \frac{128\pi^2 \omega_1^2 d_{\text{eff}}^2 a}{C^3 n_1^2 n_2} \times \frac{b}{W_0^2} h^{(2)}(\sigma, \xi) P_1 = 0 \quad (17)$$

$$\frac{d\phi_1}{d\tau_1} = 0 \rightarrow \phi_1 = \frac{\pi}{2} = \text{Const.} \quad (18)$$

a) In the absence of the non-linear crystal ( $d_{\text{eff}} = 0$ ), expression (17) gives the laser power in the resonator as

$$P_1 = P_{01} = \frac{g_0 - \alpha_1}{16g_0\beta} Cn_1 W_0^2. \quad (19)$$

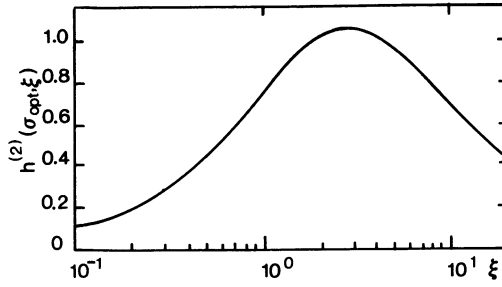


Fig. 2. — Plot of  $h^{(2)}(\sigma_{opt}, \xi)$  as a function of  $\xi$ .

Equation (19) corresponds to the case where the mirrors of the resonator have a reflection coefficient of 100 % for the laser frequency.

b) In the presence of the S.H.G., the laser power

in the resonator is

$$P_1 = \frac{P_{01}}{1 + \frac{8 \pi^2 \omega_1^2 n_1 a^2}{C^2 g_0 \beta} \cdot \frac{h^{(2)}(\sigma, \xi)}{\xi} \cdot \frac{d_{eff}^2}{n_1^2 n_2}} \quad (20)$$

We see that the pump power  $P_1$  is a simple hyperbolic function of the non-linear coefficient  $K = d_{eff}^2/n_1^2 n_2$  (see Fig. 3).

c) The power of the second-harmonic wave is

$$P_2 = \frac{1}{8} \frac{C n_2}{\pi} \int_{-\infty}^{+\infty} dX dY \left| E_2 \left( X, Y, Z = \frac{a}{2}, t \right) \right|^2 \quad (21)$$

Using (8), (5) and (20) the power  $P_2$  of the second-harmonic wave is given by

$$P_2 = \frac{128 \pi^2 \omega_1^2 a K k_1 P_{01}^2}{C^3 h^{(2)}(\sigma, \xi) \left[ \frac{1}{h^{(2)}(\sigma, \xi)} + \frac{8 \pi^2 \omega_1^2 a^2 n_1 K}{\xi} \right]^2} \frac{|H(\sigma, \xi)|^2}{\xi h^{(2)}(\sigma, \xi)} \quad (22)$$

where  $H(\sigma, \xi) = \int_{-\xi}^{+\xi} d\tau \frac{e^{-j\sigma\tau}}{1+j\tau}$ ; this is Boyd's and Kleinman's focusing function in [6]. In general  $|H(\sigma, \xi)|^2$  cannot be solved analytically but only by numerical means [6]. Calculation shows, however, that  $|H(\sigma, \xi)|^2/\xi h^{(2)} \simeq 4$ . Thus equation (22) becomes

$$P_2 = \frac{128 \pi^2 4 \omega_1^2 a K k_1 P_{01}^2}{C^3 h^{(2)}(\sigma, \xi) \left[ \frac{1}{h^{(2)}(\sigma, \xi)} + \frac{8 \pi^2 \omega_1^2 a^2 n_1 K}{\xi} \right]^2} \quad (22')$$

From equation (22) we see that  $P_2$  is proportional to the square of the intracavity power of the pump laser, the crystal length  $a$ ; the focusing function  $h^{(2)}(\sigma, \xi)$ ; and the non-linear coefficient  $K$ . The power  $P_2$  of S.H. wave reaches a maximum at  $(dP_2/dK)_{K=K_{opt}} = 0$ . Or

$$K = K_{opt} = \frac{g_0 C^2 \xi \beta}{8 \pi^2 \omega_1^2 n_1 a^2 h_{opt}^{(2)}(\sigma, \xi)} \quad (23)$$

The expression (23) is the necessary condition for the S.H. power to be a maximum. From equation (23) we see that with a given laser and a given non-linear crystal, we can choose the value of the focusing function  $h^{(2)}(\sigma, \xi)$  such that (23) is satisfied to obtain a maximal value of S.H. power  $P_2$ . This value has the form

$$P_{2max} = \frac{8 g_0 \beta P_{01}^2}{C n_1 W_0^2} \quad (24)$$

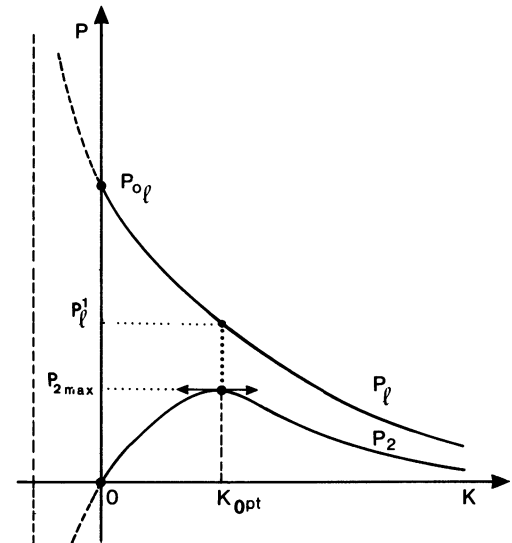


Fig. 3. — Dependence of the second-harmonic generation and laser power on the coefficient  $K$  (or  $\kappa$ ).

Figure 3 shows the dependence of  $P_2$  upon  $K$ .

We shall demonstrate that, in the case of  $P_2$  being maximized, its value is equal to the maximal output power of the pump laser when the laser pump works alone (that is without the S.H. crystal) with mirrors having optimal transmission coefficients. We can write (24) in the form

$$\frac{P_{2max}}{P_{01}} = \frac{8 g_0 \beta P_{01}}{C n_1 W_0^2} \quad (25)$$

Using (19), expression (25) becomes

$$\frac{P_{2\max}}{P_{01}} = T = \frac{g_0 - \alpha_1}{2}. \tag{26}$$

Thus  $T$  can be labelled as the « non-linear transmission coefficient » of the laser mirrors. We write (26) in the form

$$T = \frac{1}{2}(\sqrt{g_0 \alpha_1} - \alpha_1) \left( \sqrt{\frac{g_0}{\alpha_1}} + 1 \right). \tag{27}$$

According to the condition  $(g_0/\alpha_1)^2 - 1 \ll 1$  used by Rigrod [17], the following expression can be obtained :

$$\frac{1}{2} \left( \sqrt{\frac{g_0}{\alpha_1}} + 1 \right) \simeq 1. \tag{28}$$

Inserting (28) into (27) we get

$$T = \frac{P_{2\max}}{P_{01}} = \sqrt{g_0 \alpha_1} - \alpha_1.$$

It is to be noted that, in the absence of the S.H. crystal, the mirror's transmission coefficient  $T_{\text{opt}}$  resulting in the maximal power output from a laser oscillator is the same result as given in [17]

$$T_{\text{opt}} = \sqrt{g_0 \alpha_1} - \alpha_1.$$

In this way, we have demonstrated that, at the maximum S.H.G. power condition, the power of S.H.

wave is equal to the maximum pump laser output power in the absence of the non-linear crystal. This is in total agreement with the result of Smith [2].

**2.3 INTRACAVITY SECOND-HARMONIC GENERATION IN A CW-RING DYE LASER WITH THE OPTIMAL FOCUSING LASER BEAM.** — In this section, we shall determine the equations of motion, the power of the S.H.G. directly coupled dye laser and the maximal S.H.G. power under optimal focusing conditions. To simplify the problem, we will consider that the single-pass loss in the laser cavity is composed entirely of non-useful components, such as incomplete absorption of the pump radiation of the dye jet, non-radiative processes such as scattering, singlet and triplet absorption, also extraneous reflection and diffusion at the optical elements and so forth.

The properties of the dye laser used here are the same as in [5], [18], i.e. the dye is considered to be homogeneously broadened. Since the narrow bandwidth is achieved with the help of a frequency selective tuning element, it is also assumed that the dye laser radiation is essentially monochromatic (eigenmode) whereupon the conditions specified for I.S.H.G. in section 1 are satisfied. Otherwise, the analysis in section 2.1 may be used here for cw-ring dye laser operated with a unidirectional device since the hole burning effect, mode pulling-pushing effects which are absent in this laser, are ignored in that section.

**2.3.1 Equations of motion, power of the dye laser and S.H. wave. Discussion.** — For a small-gain ring dye laser, the equations of motion (14), (15) become

$$\frac{dP_1}{d\tau_1} = \left[ -\alpha_1 + \overline{N}_1(\tau_1) \sigma_1(\lambda_1) L_0 - \frac{128 \pi^2 \omega_1^2 d_{\text{eff}}^2 a}{C^3 n_1^2 n_2} \cdot \frac{b}{W_0^2} h^{(2)}(\sigma, \xi) P_1 \right] P_1 \tag{29}$$

$$\frac{d\phi_1}{d\tau_1} = -\frac{64 \pi^2 \omega_1^2 d_{\text{eff}}^2 a}{C^3 n_1^2 n_2} \cdot \frac{b}{W_0^2} h^{(2)'}(\sigma, \xi) P_1 \tag{30}$$

where  $\sigma_1(\lambda_1)$  is the stimulated cross-section,  $L_0$  the optical path length in the dye jet,  $\overline{N}_1(\tau_1)$  the population inversion in the single excited state,  $P_1$  the power of the dye laser. Using the approximation proposed by Teschke *et al.* [5] the population rate equation can be written as

$$\frac{C}{L} \frac{d}{d\tau_1} \overline{N}_1(\tau_1) = -\frac{\overline{N}_1}{\tau_R} - \frac{\overline{N}_1 \sigma_1(\lambda_1)}{h\nu_1 CS} P_1 + \frac{N_0 \sigma_p P_p}{h\nu_p CS} \tag{31}$$

where  $\sigma_p$  is the ground state absorption cross-section,  $\tau_R$  the fluorescence decay time,  $S$  the area of the pump beam in the dye jet,  $h$  Planck's constant,  $\nu_1 = 1/\lambda_1$  the wavenumber,  $P_p$  the power of the pump laser,  $N_p$  the unexcited number of molecules.

$$N_p + \overline{N}_1(\tau_1) \simeq N_0 \simeq N_p = \text{Const}. \tag{32}$$

Equation (31) is written for the fundamental Gaussian mode making the assumption that the active region is confined within a distance which is less than the confocal parameter of the beam waist, i.e. the Gaussian beam can be considered to be at constant diameter. In the steady state regime of a cw small-gain dye laser, equations (29), (30), (31) are equal to zero. Using (32) in conjunction with these equations, we have following equation for

the intracavity power of the dye laser

$$\frac{\sigma_1(\lambda_1) \tau_R \kappa h^{(2)}(\sigma, \xi) P_1^2}{h\nu_1 CS} + [\lambda_1 \alpha_1 \sigma_1(\lambda_1) \tau_R + \kappa h^{(2)}(\sigma, \xi)] P_1 - [N_0 L_0 \sigma_p P_p \tau_R \sigma_1(\lambda_1) \lambda_p - \alpha_1 hCS] = 0 \quad (33)$$

where

$$\kappa = \frac{128 \pi^2 \omega_1^3 d_{\text{eff}}^2}{C^3 n_1 n_2} ahS \quad (34)$$

or

$$\kappa(K) = \frac{128 \pi^2 \omega_1^3 n_1}{C^3} KahS. \quad (35)$$

For the general case (small-gain or large-gain dye laser), using Pike's result [18] and (33), the equation which determines the intracavity cw-dye laser becomes

$$\frac{\sigma_1(\lambda_1) \tau_R \kappa h^{(2)}(\sigma, \xi) P_1^2}{h\nu_1 CS} + [\lambda_1 \alpha_1 \sigma_1(\lambda_1) \tau_R + \kappa h^{(2)}(\sigma, \xi)] P_1 - [(1 - e^{-N_0 L_0 \sigma_p}) \lambda_p P_p \tau_R \sigma_1(\lambda_1) - \alpha_1 hCS] = 0. \quad (36)$$

\* In the absence of the non-linear effect ( $\kappa = 0$ ), equation (36) yields

$$P_1 = P_{01} = (P_p - P_{\text{pth}}) \frac{(1 - e^{-N_0 L_0 \sigma_p}) \lambda_p}{\alpha_1 \lambda_1} \quad (37)$$

where

$$P_{\text{pth}} = \frac{\alpha_1 hCS}{(1 - e^{-N_0 L_0 \sigma_p}) \lambda_p \tau_R \sigma_1(\lambda_1)} \quad (38)$$

is the dye laser threshold power;  $\alpha_1$  the total single-pass loss at the laser wavelength including nonuseful components such as scattering, singlet and triplet states absorption, extraneous reflections and so forth.

On the basis of the comparison with the power formulae in [5, 18] we may make a phenomenological correction for the Gaussian beam effect generated in the dye jet [18] to our expressions (37), (38), which become

$$P_{01} = \frac{v}{\sqrt{1+v^2}} (P_p - P_{\text{pth}}) \frac{(1 - e^{-N_0 L_0 \sigma_p}) \lambda_p}{\alpha_1 \lambda_1} \quad (39)$$

$$P_{\text{pth}} = \frac{(1+v) \alpha_1 hCS}{2 v \lambda_p \sigma_1(\lambda_1) \tau_R (1 - e^{-N_0 L_0 \sigma_p})} \quad (40)$$

where the parameter  $v$  is defined as the ratio of the areas of the pump laser and dye laser beam at the jet.

$$v = \frac{S}{\pi W_1^2}.$$

The expressions (39), (40) are similar to Pikes' formulae [18] in the case where the mirrors of the laser resonator have reflection coefficient 100 % at the laser wavelength.

\* In the presence of the S.H.G. effect ( $\kappa \neq 0$ ), using equation (36) and the above correction, we have the following formulae for the intracavity power of the dye laser

$$P_1 = \frac{\frac{v}{\sqrt{v^2+1}}}{2 \kappa(K) \tau_R \sigma_1 \lambda_1 h^{(2)}(\sigma, \xi)} \cdot \left[ -\alpha_1 \lambda_1 \sigma_1 \tau_R - \kappa(K) h^{(2)} + \left\{ [\alpha_1 \lambda_1 \sigma_1 \tau_R + \kappa(K) h^{(2)}]^2 + \frac{4 \tau_R \sigma_1 \lambda_1 \kappa(K) h^{(2)}}{hCS} [2 v \sigma_1 \tau_R \lambda_p P_p (1 - e^{-N_0 L_0 \sigma_p}) - (1+v) \alpha_1 hCS] \right\}^{1/2} \right]. \quad (41)$$

Using (5), (8), (21), (38) and (41), the expression for S.H. output power  $P_2$  is

$$P_2 = \frac{v^2 hCS}{4(1+v^2) \lambda_1^2 \kappa(K) (\sigma_1 \tau_R)^2 h^{(2)}} \left[ -\alpha_1 \lambda_1 \sigma_1 \tau_R + \kappa(K) h^{(2)} + \left\{ [\alpha_1 \lambda_1 \sigma_1 \tau_R + \kappa(K) h^{(2)}]^2 + \frac{4 \lambda_1 \sigma_1 \tau_R \kappa(K) h^{(2)}}{hCS} [2 v \sigma_1 \tau_R P_p (1 - e^{-N_0 L_0 \sigma_p}) - (1+v) \sigma_1 hCS] \right\}^{1/2} \right]^2. \quad (42)$$

The S.H.G. power reaches a maximum at  $(dP_2/d\kappa)_{\kappa=\kappa_{opt}} = 0$ , thus

$$\kappa_{opt} h_{opt}^{(2)}(\sigma, \xi) = \alpha_1 \sigma_1(\lambda_1) \tau_R \lambda_1. \quad (43)$$

Using (35), (43) becomes

$$\frac{128 \pi^2 \omega_1^3 n_1}{C^3} KhSah^{(2)}(\sigma, \xi) = \alpha_1 \sigma_1(\lambda_1) \tau_R \lambda_1. \quad (44)$$

Equations (43) and (44) like expression (23) define the optimal focusing condition for maximal S.H.G. power, the value of which is

$$P_{2max} = \frac{2 \lambda_p hC}{\lambda_1 \tau_R} \cdot \frac{v^2}{1+v^2} \left( \sqrt{\frac{2 v \tau_R P_p}{hC} (1 - e^{-N_0 L_0 \sigma_p})} - \sqrt{\frac{(1+v) \alpha_1 S}{\lambda_p \sigma_1(\lambda_1)}} \right)^2 \quad (45)$$

and the intracavity power of the dye laser in this case is

$$P_1 = P_1^1 = \frac{2 v}{1+v} \cdot \frac{P_{01}}{\left( \frac{P_p}{P_{pth}} \right)^{1/2} + 1}. \quad (46)$$

Equations (41), (42) show that the powers of the dye laser and S.H. wave are a simple function of  $K$ . Figure 3 illustrates their  $K$ -dependence on a different scale.

When the thermal effect in the S.H.G. crystal has to be taken into account, we can correct the power formula (45) using the result obtained by Ferguson *et al.* [4] so that

$$P_{2max} = \frac{2 hC v^2 \lambda_p h_T}{\tau_R (1+v^2) \lambda_1} \left( \sqrt{\frac{2 v \tau_R P_p (1 - e^{-N_0 L_0 \sigma_p})}{hC}} - \sqrt{\frac{(1+v) \alpha_1 S}{\lambda_p \sigma_1(\lambda_1)}} \right)^2 \quad (47)$$

where  $h_T(P_1^1)$  is the thermal function which depends on the intracavity power of dye laser. Equation (47) shows that the S.H. wave maximal power  $P_{2max}$  is a function of the single-pass power loss  $\alpha_1$ ,  $\lambda_1$  and  $\sigma_1(\lambda_1)$  and it exists only when  $P_p > P_{pth}$ .

With a given I.S.H.G. system having a threshold  $P_{pth}$ , its single pass loss  $\alpha_1$  is defined by (40) and the optimal value of focusing function  $h^{(2)}(\sigma, \xi)$  by (43), thus the optimal configuration of this system is achieved. If a system is not at the optimal focusing

condition (43) it is necessary to use formula (42) to determine its S.H.G. power.

In the case of a I.S.H.G. system using ADA crystal ( $\rho = 0$ ,  $d_{eff} = 7.4 \times 10^{-8}$  ues [19];  $n_o = n_e = 1.57$  [20];  $a = 1.8$  cm) and Rh 6G dye ( $\tau_R = 5 \times 10^{-9}$  s;  $N_0 = 1.2 \times 10^{18}$  molecules/cm<sup>3</sup>;  $\sigma_1(\lambda_1 = 0.586 \mu\text{m}) = 1.62 \times 10^{-16}$  cm<sup>2</sup> [18]) with  $\alpha_1 = 0.62$ ;  $S = \pi(1.4)^2 \times 10^{-6}$  cm<sup>2</sup>, at wavelength  $\lambda_1 = 0.586 \mu\text{m}$ , the optimal value of the focusing function  $h^{(2)}(\sigma, \xi)$  given by (43) is  $h_{opt}^{(2)} = 0.1$ . The dependence of the maximal S.H.G. power of the above system on the S.H. wavelength as expressed in equation (47) is shown in figure 4 for different values of  $\alpha_1$ . The working conditions being : a 3 watts argon ion laser, operating at 514.5 nm in single transversal mode with a pump mirror of curvature radius of 5 cm, is used to pump a jet of rhodamine 6G dye whose thickness is 0.2 mm. The pump beam makes a Brewster angle ( $57^\circ 2$  for 514.5 nm) with the dye jet, giving a resultant optical path length  $L_0 = 0.37$  mm and a pump beam area radius of

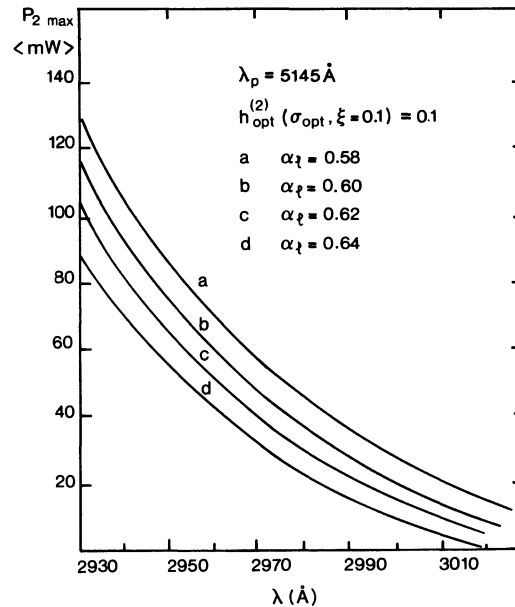


Fig. 4. — Dependence on the second-harmonic generation maximal power on its wavelength.  $P_{2max}$  versus  $\lambda_1/2$  for various values of single-pass loss  $\alpha_1$  calculated from (47). The values of  $h_{opt}^{(2)}$  and  $\alpha_1$  are  $h_{opt}^{(2)} = 0.1$ .

- a)  $\alpha_1 = 0.58$       b)  $\alpha_1 = 0.60$
- c)  $\alpha_1 = 0.62$       d)  $\alpha_1 = 0.64$ .

The other parameter values are given in the text.



0.014 mm at the jet. The value of the function  $h_T(P_1^1)$  is taken from [4], the value of  $v$  is 1, i.e.  $W_1 = 0.014$  mm. We see in figure 4 the rapid decrease of the maximal S.H.G. power caused by the forceful decline of the stimulated emission cross-section  $\sigma_1(\lambda_1)$  [18] when  $\lambda_1$  increases. With a given cavity (i.e. for a given  $b$ ) we can determine the optimal crystal length which satisfies the expression (44) i.e. to develop an optimal I.S.H.G. system; this is in agreement with the conclusion of Ferguson and Dunn [4]. We have calculated the optimal length of the crystal for  $b = 3$  cm, its value is 0.73 cm.

**2.3.2 Non-linear transmission (non-linear coupled loss).** — It can be shown by means of (45) and (39) that the maximal power of the S.H.G. under the optimal focusing condition is identical to the fundamental output power of the dye laser when the dye works alone with mirrors having optimal transmission coefficient.

In the case of  $S = \pi W_1^2$ , using equations (39), (40) and (45) we have the following formula for the « non-linear transmission » :

$$T = \frac{P_{2\max}}{P_{01}} = 2 \alpha_1 \cdot \frac{\sqrt{P_p} - \sqrt{P_{\text{pth}}}}{\sqrt{P_p} + \sqrt{P_{\text{pth}}}}. \quad (48)$$

If the I.S.H.G. system operates close to the threshold (i.e.  $P_p \simeq P_{\text{pth}}$ ) the formula (48) can be written as

$$T = \frac{P_{2\max}}{P_{01}} = \alpha_1 \left( \sqrt{\frac{P_p}{P_{\text{pth}}}} - 1 \right). \quad (49)$$

For a small-gain dye laser, (49) becomes

$$T = \frac{P_{2\max}}{P_{01}} = \sqrt{\frac{2 \alpha_1 N_0 L_0 \sigma_p \tau_R P_p \sigma_1}{h\nu_1 CS}} - \alpha_1. \quad (50)$$

Expression (50) agrees with Walter Koechners' optimal laser output coupling formula [21].

### 3. Experiment.

In this part, we describe the experiment performed both to test the validity of the theory and to produce

an efficient u.v. radiation suited to spectroscopic experiment.

**3.1 EXPERIMENTAL ARRANGEMENT.** — A scheme of the experiment to investigate the I.S.H.G. in cw-ring dye laser is shown in figure 5. A Spectra-Physics 380 D ring dye laser is used in our experiment. The conditions relating to the pump, dye, jet, curvatures of mirrors path length and pump beam area are those previously described at the end of paragraph 2.3.1.

The ring dye laser has a four mirror  $M_1, M_2, M_3, M_4$  cavity with mirrors reflection coefficients of 100 % for visible frequencies. Three curved mirrors with radii of curvature of 3.5 cm, 10 cm, 23 cm are used and the distances between these mirrors are  $d_{12} = 8.5$  cm,  $d_{23} = 34.5$  cm. The fourth mirror is flat and the third mirror  $M_3$  is 75 % transparent to u.v. light (see Fig. 5). There are two waists in the laser cavity. One at the dye jet of 0.014 mm radius and the other in the arm  $M_2 M_3$  of 0.043 mm radius. The tuning elements of the ring dye laser are composed of a birefringent filter and an electronically tunable etalon. The F.S.R. of the scanning etalon is 75 GHz. The stable, high power one way propagation is obtained through the use of an unidirectional device which consists of a Faraday rotator plate and a crystal quartz rotator plate.

For double frequency purpose, a temperature 90° phase-matched ADA crystal measuring 1.8 cm in length is placed at the arm  $M_2 M_3$  in such a way that the centre of the second waist coincides with the centre of the crystal. Since at the second focus of the laser cavity the power density far exceeds the damage threshold of antireflexion coatings, the ADA crystal is placed at the Brewster angle.

The temperature of the crystal is varied at 0.1 °C intervals by an electronically controlled stabilizing oven. The S.H.G. crystal is oriented so that the parametric interaction in it is of type I, i.e. the polarization vector of the pump wave is perpendicular to the optical axis when the polarization of the S.H.G. wave is parallel to it. Using [7] we see that our system operates in stable zone of the cavity.

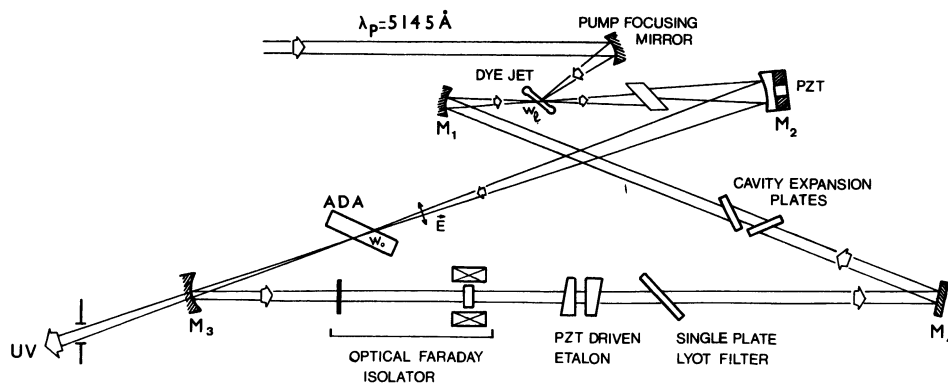


Fig. 5. — The experimental set up.

The focusing parameter value of the above system is

$$\xi_{\text{exp}} = \frac{a}{b} = \frac{a}{2 \pi n_1 W_0^2}.$$

Using  $a = 1.8 \text{ cm}$ ;  $\lambda_1 = 0.586 \times 10^{-4} \text{ cm}$ ;  $W_0 = 4.3 \times 10^{-3} \text{ cm}$  and  $n_1 = 1.5$ ; we obtain  $\xi_{\text{exp}} = 0.60$ . The value of the focusing function in this case is  $h_{\text{exp}}^{(2)}(\sigma_{\text{opt}}, \xi_{\text{exp}} = 0.6) = 0.55$  (see Fig. 2). Comparing with the above theoretical value of the focusing function  $h_{\text{opt}}^{(2)} = 0.1$ , we see that our experimental system is not at the optimal focusing conditions. To satisfy the optimal focusing conditions, we have to change either the cavity configuration (mirrors curvatures or distances) or the crystal length.

**3.2 RESULTS AND DISCUSSION.** — To define the single-pass loss  $\alpha_1$  of the I.S.H.G. system, the pump power threshold  $P_{\text{pth}}$  has been measured at the laser wavelength  $\lambda_1 = 0.586 \mu\text{m}$  with a Spectra-Physics wattmeter. The obtained value is 1.8 watt, which means  $\alpha_1 = 0.62$  (using formula (40)). This measured value of the loss is acceptable since, from its definition in 3.1  $\alpha_1$  is the total loss at the laser wavelength. This includes nonuseful components such as 10% of the pump radiation which is not absorbed by the dye, 25% goes into non radiative processes which generate heat [3, 22, 23], approximately 21% is lost at the optical elements in the laser cavity [9] and a 2.5% loss is introduced by a deliberate slight misalignment [5]. With an u.v. detector (a Mole cell with sensibility of 0.16 mV/mW) and a Jobin-Yvon monochromator, the S.H.G. power has been found to be a function of S.H. wavelength at the argon pump laser power of 3 W. Ferguson *et al.* [3] have shown that excited state absorption and thermal effects increase to such an extent that it is better to use a relatively small pump power to reduce saturation effects. In our case a suitable pump power is 3 W at  $\lambda_p = 514.5 \text{ nm}$ . In these experiments, the u.v. wavelength is tuned across a wide range between 2 930 Å and 3 020 Å by means of temperature variation of the ADA crystal. Due to the limited transmission of  $M_3$ , the u.v. output power is lower than the u.v. generated in the crystal (lower than the output power corresponding to 100% u.v. transmission of the mirror  $M_3$ ). Figure 6 shows the S.H.G. power as a function of u.v. wavelength. The shape of the theoretical curve (solid line), which is defined by (42) using the practical value of the focusing function  $h_{\text{exp}}^{(2)} = 0.55$ , matches to the plotted experimental data within the accuracy of the experiment. To obtain the maximum value of the S.H. generated wave, it would be easier to cut the crystal to the optimal length  $a_{\text{opt}} = 0.73 \text{ cm}$  since our resonator has a confocal parameter  $b = 3 \text{ cm}$  inside the S.H.G. crystal. This method would have another advantage in eliminating the thermal effect which becomes a serious problem at large crystal lengths [3]. The laser frequencies can be scanned over 30 GHz and 60 GHz for visible and u.v. radiation respectively, and the use of active stabilization by

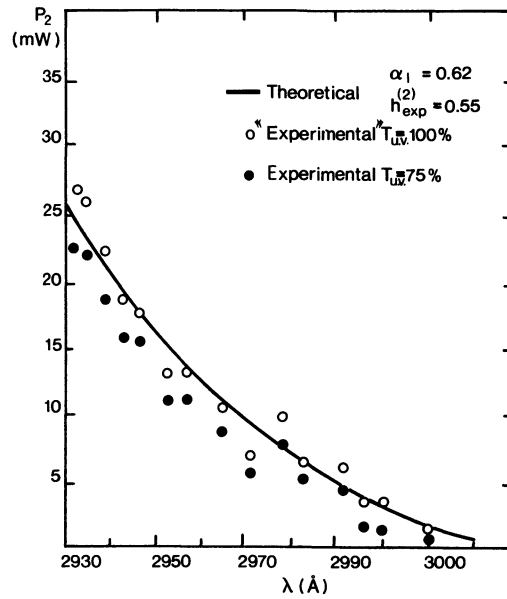


Fig. 6. — Dependence of the second-harmonic generation power on its wavelength. The solid line is the theoretical curve as expressed in (42). The values chosen for  $h^{(2)}$  and  $\alpha_1$  are :  $h_{\text{exp}}^{(2)}(\sigma_{\text{opt}}, \xi = 0.6) = 0.55$ ;  $\alpha_1 = 0.62$ . The other parameter values are the same as those quoted for figure 4. The experimental points ● correspond to u.v. power measured with the 75% u.v. transparency of the  $M_3$  mirror. The « experimental » points ○ correspond to the u.v. power which would be measured if a 100% u.v. transparency would be available. The theoretical curve was obtained with  $T = 100\%$  for the  $M_3$  mirror.

the Spectra-Physics 388 stabilock system in laser reduces the widths to less than 1 MHz and 2 MHz. In other experiments, the above mentioned u.v. laser system has also been used in range 2 940 Å-2 955 Å to investigate the hyperfine structure of the  $f(0_g^+)$  state of iodine [13].

**4. Conclusion.**

A detailed quantitative analysis of the intracavity second-harmonic generation (I.S.H.G.) using the optimal laser beam focusing method has been carried out. With the directly coupled non-linear crystal laser model, we have developed expressions for the optimal focusing condition, maximal output power of second harmonic wave and the « non-linear transmission » of the laser mirrors. Whilst our formulae are general for I.S.H.G. systems using Gaussian laser beams, they give very good agreement with results published by other authors, as discussed in the text. The theory was tested by experimental work using a ring dye laser, and, in this case, the agreement between experimental and theoretical results was highly satisfactory. It has been shown that an optimal focusing I.S.H.G. system increases the efficiency of the S.H. wave output power. This work leads us to make two remarks of practical

importance. Firstly, that it is not necessary to use a very long crystal in order to maximize S.H.G. output power. Secondly, there is no requirement for a large pump input power for the ring dye laser. In the future, our results might be used to improve existing u.v. sources used in spectroscopic studies.

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