# RATE EQUATION SIMULATION OF A SYNCHRONOUSLY PUMPED DYE LASER

L.M. DAVIS, J.D. HARVEY and J.M. PEART
*Physics Department, University of Auckland, Private Bag, Auckland, New Zealand*

Received 20 July 1983
Revised manuscript received 2 February 1984

A simple rate equation model of a standing wave synchronously pumped dye laser yields output pulses which agree qualitatively and quantitatively with recent experimental observations. The shape, amplitude and temporal position of the simulated pulse varies dramatically, not only with cavity length detuning, but also with the gain to loss ratio. Features of pulse formation and stability are predicted which are precluded by the steady-state assumption present in most other models.

## 1. Introduction

The widespread use of the mode locked synchronously pumped dye laser as a source of picosecond light pulses has been paralleled by a widespread interest in the theoretical modelling of these systems [1]. While some early work [2,3] was based on rate equation models which were solved numerically for the output pulse characteristics, most theoretical treatments are based on the self reproducing pulse idea [4,5] in which a single pulse is assumed to circulate without any net change after each round trip of the dye laser cavity. In the model introduced by Haus [6] and in subsequent treatments based on his mode locking equation, a specific form for the modulation loss [7] or pulse shape [8–10] is assumed in order to study output parameters as a function of the operating conditions of the dye laser. These models do not treat the standing wave cavity, in which the dye laser pulse makes two passes through the dye jet per round trip but they do include the effects of a bandwidth limiting filter. It has been asserted [1,10] that the dye laser pulse width is determined by a balance between the temporal compression introduced by the gain modulation, and the temporal expansion produced by the finite band width of the wavelength determining filter. Thus, early rate equation models [2,3] have since received little attention as they did not include the effects of a bandwidth limiting filter and were thought to be incapable of generating stable self reproducing pulses of realistic widths. It has recently been shown, however [13], that a bandwidth limiting filter is not necessary to produce stable pulses from a synchronously pumped dye laser. There are several more recent models [11–18] not derived from Haus's equation which do not assume a form for the dye laser pulse shape, but most [11–16] have been restricted to the treatment of the steady state operating regime of a ring dye laser cavity.

We have examined various theoretical treatments as an aid to understanding the behaviour of a commercially available dye laser system, and our purpose in this communication is to point out several advantages of a rate equation approach. These include the ability to treat a standing wave cavity with two passes through the dye-jet, the ability to investigate any cavity detunings, and to include relaxation of the excited state population by spontaneous emission. In addition the model permits the investigation of the non steady state behaviour of the laser which occurs under certain operating conditions. It is found that the model gives results which are in good agreement with our experimental observations and with those reported by other investigators.

0 030-4018/84/$03.00 © Elsevier Science Publishers B.V.
(North-Holland Physics Publishing Division)

## 2. Theory

Consider a three-mirror standing wave dye laser with cavity losses $L$, excited by a pump laser pulse of intensity $I_p(t)$ at time $t$. As the vibrationally excited states of the ground and first electronic level have sufficiently short lifetimes that their populations may be neglected, the medium may be approximated as a two level system in which the excited state and ground state population densities are given by $N_1$ and $N_0$ respectively. The stimulated emission and absorption cross-sections are given by $\sigma_e$ and $\sigma_a$ respectively, the pump absorption cross section by $\sigma_p$, and $\tau_d$ is the spontaneous decay lifetime for the excited state.

Let $I_l(t)$ and $I_l(t-\tau)$ be the intensity of the intracavity pulse incident on the dye jet at time $t$ for the first and second passes through the dye jet respectively, where $\tau$ is the time elapsed between passes.

The gain at the dye jet due to stimulated emission is

$$G(t) = N_1(t)\sigma_e - N_0(t)\sigma_a,$$

and the coupled rate equations for the system are

$$\mathrm{d}N_1(t)/\mathrm{d}t = N_0(t)\sigma_p I_p(t) - G(t)[I_l(t) + I_l(t-\tau)] - \tau_d^{-1}N_1(t),$$

$$\mathrm{d}I_l(t)/\mathrm{d}t = cG(t)I_l(t) + c\epsilon\tau_d^{-1}N_1(t),$$

$$\mathrm{d}I_l(t-\tau)/\mathrm{d}t = cG(t)I_l(t-\tau) + c\epsilon\tau_d^{-1}N_1(t),$$

where $c$ is the speed of light and $\epsilon$ is the proportion of spontaneous fluorescence contributing to the dye laser beam. The rate equations may be converted to difference equations by using the first order Taylor expansion and making a lumped gain approximation for each passage through the dye jet of thickness $x$.

$$N_1(t+\Delta t) = N_1(t) + N_0(t)\sigma_p I_p(t)\Delta t - G(t)[I_l(t) + I_l(t-\tau)]\,\Delta t - \tau_d^{-1}N_1(t)\Delta t,$$

$$I_l(t+\Delta t) = I_l(t) + G(t)I_l(t)x + \epsilon\tau_d^{-1}N_1(t)x,$$

$$I_l(t-\tau+\Delta t) = I_l(t-\tau) + G(t)I_l(t-\tau)x + \epsilon\tau_d^{-1}N_1(t)x.$$

The total cavity losses, $L$, including transmission at the output coupler, are also lumped and the intracavity dye laser intensity profile is multiplied by $(1-L)$ with each round trip.

The difference equations are solved by recursively calculating the dye laser intracavity intensity and a gain at each interval of $\Delta t$. An interval of $\Delta t = 0.2$ ps was chosen for the results presented here; intervals between 0.1 ps and 1.0 ps were found to have negligible effect on the solutions. The assumption that the laser pulse has zero intensity, except over an interval 300 ps before and 500 ps after the pump pulse arrives at the dye jet, and that the excited state decays only by spontaneous emission outside this interval, leads to a considerable reduction in computer time and storage requirement. This approximation was found to have no noticeable effect on the solutions generated with a mesh spacing of $\Delta t = 1.0$ ps, and has been used in most of the results presented here.

Detuning of the dye laser cavity length is defined as $\delta t$ = (dye laser round trip time − pump laser round trip time), and is effected by an appropriate shifting and interpolation of the array elements.

After each cavity round trip has been simulated the self reproducing pulse criterion is tested by comparing the pulse with that from the previous round trip according to:

$$\left[\frac{\sum_n [I_l(n\Delta t) - \tilde{I}_l(n\Delta t)]^2}{\sum_n [I_l(n\Delta t)]^2}\right]^{1/2} \times 100 < (0.1)\%$$

where $\tilde{I}_l$ is the pulse intensity profile from the previous round trip, and $n$ is such that the sums are over all the array elements. The number of round trips required for this convergence was typically several hundred but depended on the operating parameters of the dye laser (see sect. 4).

## 3. Experimental

The results predicted by the model described here were compared with the observed behaviour of a CR599 dye laser (Coherent Radiation Palo Alto Calif.) pumped by a CR12 mode-locked argon laser operator in the frequency-tripled configuration [19]. No bandwidth limiting filter was present in the cavity.

The temporal shift of the dye laser pulse as the cavity length was detuned was measured by monitoring the change in temporal position of the dye laser pulse was passed through an adjustable delay line to a fast photodiode (CR299, 150 ps FWHM) and was observed on a sampling oscilloscope (Tektronix 7094 : 7S11 : S2), externally triggered by the pump pulse. As the dye laser cavity was detuned, the delay line length was adjusted to compensate for the temporal shift of the dye laser pulse. A further periodic delay was introduced to the dye laser pulse using a rotating glass slab. Phase-locked signal processing performed on the oscilloscope vertical output, sampled at an arbitrary fixed scan point, enabled the delay line adjustment to be made to within a few picoseconds [20].

The dye laser pulses were also used to excite a cell containing a two photon fluorescence (TPF) dye and the point of maximum TPF signal used to define zero relative detuning and zero relative dye laser pulse shift. No direct or autocorrelator measurements were made of the pulse widths; but they were always negligible compared with the response of the photodiode.

## 4. Results

We have performed many simulations under different operating conditions and the following results are representative of the trends which we have observed.

The shortest single pulses are obtained for a low gain to loss ratio and small positive detunings, with short pump pulses. The model yields pulses of a few picoseconds width when pumping close to threshold, but these have low power and would be difficult to obtain experimentally [21].

As the dye laser is pumped higher above threshold, or as the reflectance of the output coupler is increased to increase the gain to loss ratio, we find that larger positive detunings are required in order to obtain single pulses. These pulses are broader, but converge faster and are not so critically dependent on operating conditions (such as detuning).

Fig. 1(a) shows predicted pulse intensity profiles for three values of detuning, with all other operating conditions held constant. As the dye laser cavity is shortened the dye laser pulse shifts to earlier times with respect to the pump pulse, and its width decreases. If the cavity is shortened further the pulse does not converge to a steady stage single pulse profile (as determined by the criterion in the previous section) but continues to evolve slowly. For these cavity detunings, which are tens of microns shorter than the length required for the minimum pulse width, the model predicts unstable pulses in an experimental laser system, which is subject to small pump pulse and cavity length fluctuations. Shortening the cavity length further yields stable (convergent) solutions with satellite pulses. All of these trends are in good agreement with streak camera observations [22] in which pulse profiles were observed without the usual time averaging constraint of autocorrelators. They are also consistent with our own observations with a fast photodiode and oscilloscope, and the observations of others [21,23,24].

Fig. 1(b) shows the excited state population profiles corresponding to the pulses of fig. 1(a). The form of these is in agreement with our own observations of the fluorescent intensity of the dye spot [20] and those of others (7) and highlights the importance of the second pass through the dye jet.

The apparatus described in section 3 was used to measure the dye laser pulse shift as a function of cavity length detuning, for stable single pulses, and fig. 2 shows that the computer simulated results, using corresponding operating parameters, gave pulse shifts in good agreement. The computer simulated curve in fig. 2 is positioned by finding the simulated pulse which gives the maximum value of average intensity squared; this pulse corresponds to the experimental pulse which produces maximum TPF and which defines zero relative detuning and zero relative shift. Note that this point is at a substantial positive absolute detuning. Also, as the gain to loss ratio is decreased, it shifts towards zero absolute detuning in agreement with an experimentally observed shift.

Fig. 3 shows pulse intensity profiles for four values of pump pulse intensity, with all other operating conditions held constant. For a fixed positive cavity detuning, double, triple, or higher multiple pulsing is obtained if the pump intensity is increased. If the pump intensity is reduced, the dye laser pulse shifts to later times with respect to the pump pulse and becomes broader. Similar results are obtained when the gain/loss ratio of the dye laser is varied by holding the pump intensity constant but changing the cavity losses.

Our computer simulations allow the evolution of a pulse from spontaneous emission to be followed.

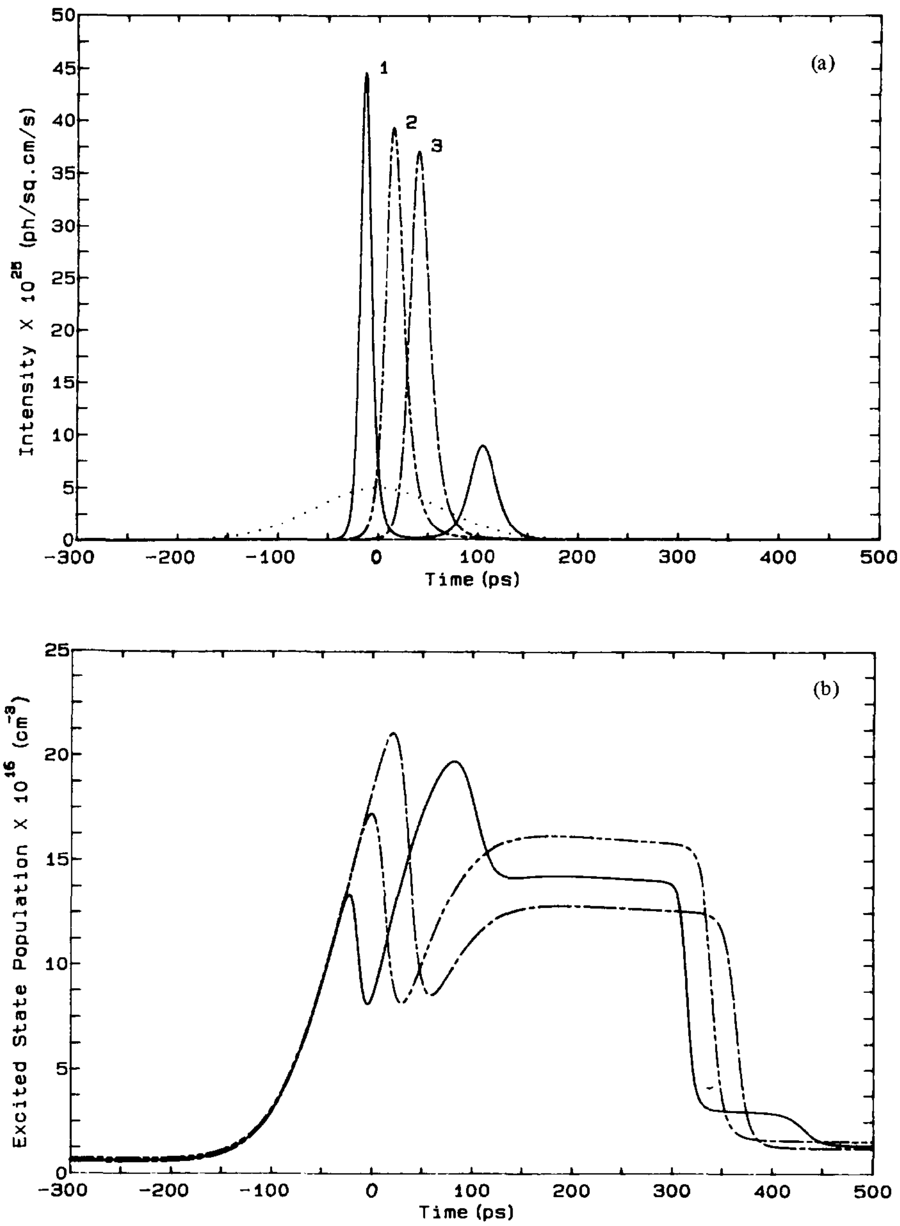


Fig. 1. (a) Intracavity pulse intensity profiles for different detunings. —— (1) Detuning = +1.00 ps (double pulse). – -- – -- (2) Detuning = +2.35 ps (good single pulse). – - – - (3) Detuning = +2.50 ps (broader single pulse). . . . . . Pump pulses (shown as dotted curve). FWHM = 150 ps, peak intensity = 5.0 × $10^{25}$ photons/cm$^2$/s (corresponds to a real pump power of about 1.0 W), cavity losses = 0.45. (b) Corresponding excited state population profiles. These show the depletion of the excited state population as the dye laser pulse passes through the dey jet. Note also the large depletion with the second pass through the dye jet.

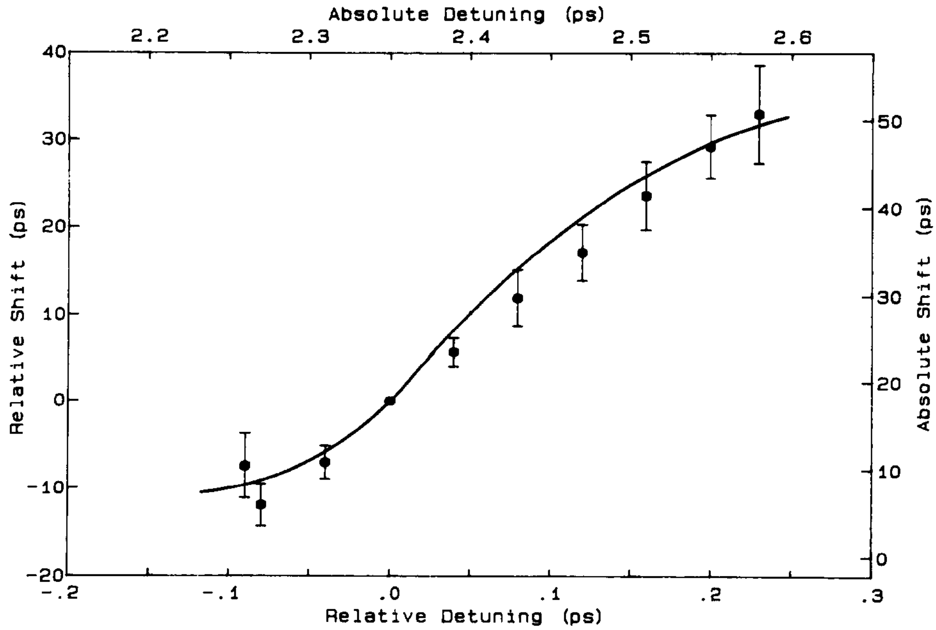


Fig. 2. Pulse shift versus detuning. Experimentally measured relative shift in the temporal position of the dye laser pulse with respect to the pump laser pulse for various values of relative detuning. The smooth curve is generated by the computer simulations and is labelled by the top and right axes. Operating conditions for the dye laser for the experimental run are: Pump pulses FWHM approximately 150 ps, average power = 1.0 watt, output coupler loss = 0.40. No intracavity bandwidth limiting filter in dye laser. Pump laser and dye laser are operated in a frequency tripled configuration [17]. Operating parameters for the dye laser for the simulated curve are: pump pulses FWHM = 150 ps, peak intensity = $5.0 \times 10^{25}$ photon/cm$^2$/s, dye laser cavity loss = 0.45. Dye laser cavity length same as experimental laser (i.e. 4400 ps). Pulse shapes corresponding to absolute detunings of 2.35 and 2.5 ps are shown in fig. 1(a).

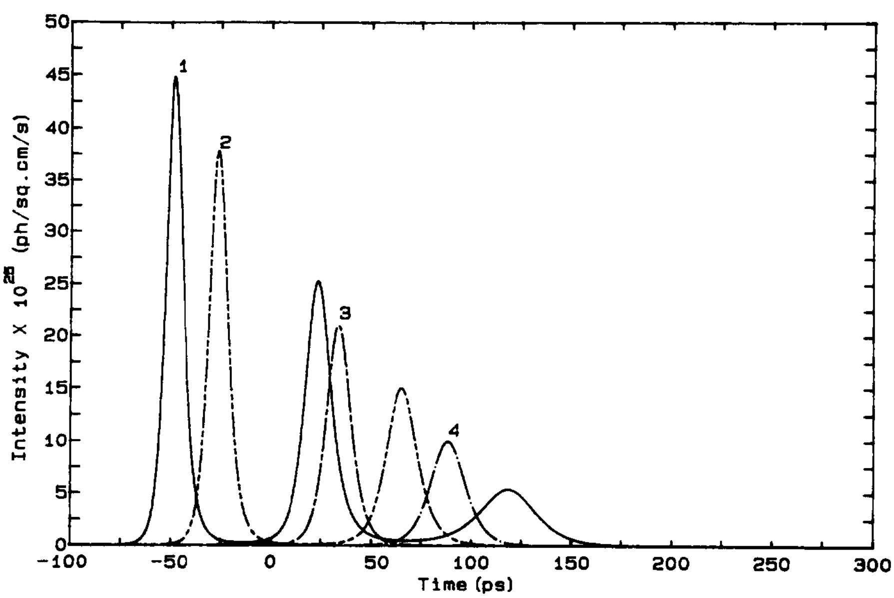


Fig. 3. Intracavity pulse intensity profiles as a function of pump intensity. Dye laser detuning = 0.6 ps, cavity losses = 0.45, pump pulses FWHM = 150 ps. —— (1) (triple pulse) pump peak intensity = $6.0 \times 10^{25}$ ph/cm$^2$/s. — -- — -- (2) (double pulse) pump peak intensity = $4.0 \times 10^{25}$ ph/cm$^2$/s. - - - - (3) (good single pulse) pump peak intensity = $2.0 \times 10^{24}$ ph/cm$^2$/s. — . — . (4) (broader single pulse) pump peak intensity = $1.5 \times 10^{25}$ ph/cm$^2$/s.

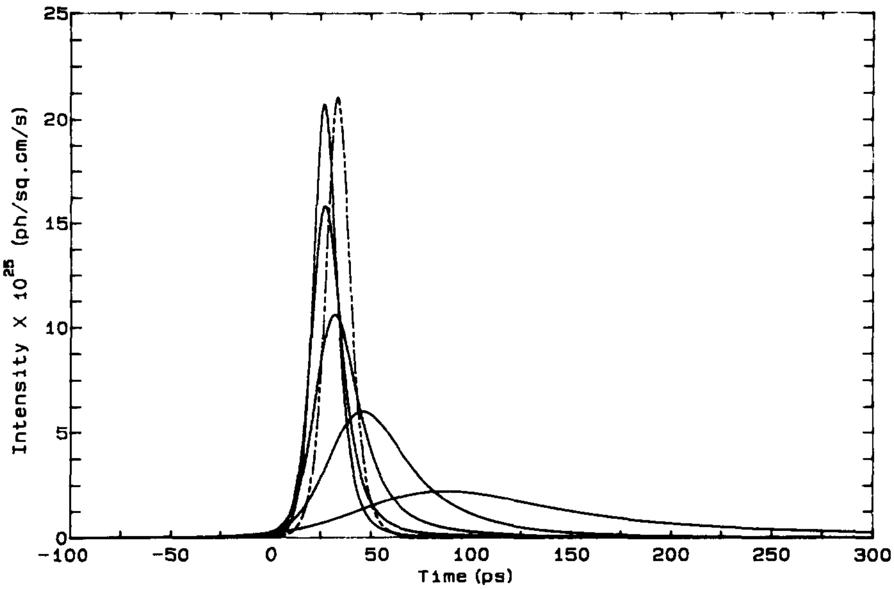


Fig. 4. Pulse evolution. Evolution of a pulse from spontaneous emission and pulse shaping effects are shown by plotting the pulse every 25 round trips, for the first 125 round trips. The final steady state pulse (after 760 round trips) is the dashed curve. Operating parameters (same as curve 3 of fig. 3) are: dye laser detuning = 0.6 ps, cavity losses = 0.45, pump pulse FWHM = 150 ps, peak intensity = $2.0 \times 10^{25}$ photons/cm$^2$/s.

Pulse evolution depends on the operating parameters, but fig. 4 is representative of the evolution of a good single pulse. The pulse builds up from spontaneous emission, with the leading edge experiencing gain and the trailing edge loss. The pulse attains its shape in about a hundred round trips, but then evolves more slowly to its final steady state position.

The overall efficiency of the dye laser may be estimated if the loss due solely to the output coupler is separated from other intracavity losses, and the time integral of the dye laser pulse passed by the output coupler is divided by the time integral of the pump laser pulse. Estimates for varying operating conditions yield efficiencies in the range of 20–70% which are in approximate agreement with the observed efficiencies of our laser (up to 40% when well aligned).

## 5. Conclusions

Our investigations indicate that numerical solution of a simple rate equation model for the standing wave dye laser yields pulse profiles which are in good qualitative and reasonable quantitative agreement with experiment. The complex changes in the pulse properties when operating conditions are varied, are also in accord with available experimental data. The model indicates, in agreement with previous work [22], that the best operating conditions for the dye laser cover a range of cavity detunings a few micrometers longer than the position of maximum second harmonic (or TPF) generation.

In comparison with previous models, the approach described here has several advantages. Firstly it enables the inclusion of a second pass through the dye jet, and as can be seen from fig. 1(b) this second pass involves substantial amplification and pulse shaping. In addition the model allows the evolution of the laser output from initial spontaneous emission to final stable pulses to be followed. This gives an indication of the stability of the dye laser. Steady state models based on the self reproducing pulse criterion cannot yield this information, although by a suitable extension of these models the pulse evolution can be investigated [18]. Our results yield convergence times similar to those obtained in this way. The approach described here could also be used to model ring dye lasers or cavity dumped systems, or to study the effects of fluctuations in the dye laser operating conditions.

The major approximations involved in the model are the following: the dye molecules are represented as a two level system, the finite thickness of the dye jet has not been taken into account (lumped gain approximation), and the beam profiles are assumed to have a uniform intensity cross section. Of these, we feel that the last approximation is the most important, in view of the dependence of pulse shaping effects upon the temporal gain profile, which will vary over the cross section of the beam. No bandwidth limiting filter has been included in the modelling calculations reported here since no such filter was present in the experimental laser. We note that the inclusion of such a filter was not found to be necessary in order to yield stable solutions. The effects of including a band width limiting filter are being investigated separately.

## Acknowledgement

This work was supported in part by grants from the Cáncer Society of New Zealand, the Medical Research Council of New Zealand and the New Zealand University Grants Committee. The authors are also grateful for assistance from J.S. Patel in the early stages of this work.

## References

[1] G.H.C. New, Phil. Trans. R. Soc. Lond. A298 (1980) 247.
[2] A. Scavennec, Optics Comm. 17 (1976) 14.
[3] Z.A. Yasa and O. Teschke, Optics Comm. 15 (1975) 169.
[4] J.R. Creighton and J.L. Jackson, J. Appl. Phys. 42 (1971) 3409.
[5] D.J. Kuizenga and A.E. Siegman, IEEE J. Quant. Elect. QE-6 (1970) 694.
[6] H.A. Haus, IEEE J. Quant. Elect. QE-11 (1975) 323.
[7] N.J. Frigo, T. Daly and H. Mahr, IEEE J. Quant. Elect. QE-13 (1977) 101.
[8] C.P. Ausschnitt, R.K. Jain and J.P. Heritage, IEEE J. Quant. Elect. QE-15 (1979) 912.
[9] C.P. Ausschnitt and R.K. Jain, Appl. Phys. Lett. 32 (1978) 727.
[10] D.K. Kim, J. Kuhl, R. Lambrich and D. von der Linde, Optics Comm. 27 (1978) 123.
[11] L.W. Casperson, J. Appl. Phys. 54 (1983) 2198.
[12] J. Herrmann and U. Motschmann, Appl. Phys.B 27 (1982) 27.
[13] J.M. Catherall, G.H.C. New and P.M. Radmore, Optics Lett. 7 (1982) 319.
[14] V.A. Nekhaenko, Sov. J. Quant. Elect. 11 (1981) 446.
[15] M. Piche and P.A. Belanger, Optics Comm. 35 (1980) 137.
[16] P.M. Radmore, Phys. Lett. 89A (1982) 4.
[17] Z.A. Yasa, Appl. Phys. B 30 (1983) 135.
[18] J. Herrmann and U. Motschmann, Optics Comm. 40 (1982) 379.
[19] A.I. Ferguson, Optics Comm. 38 (1981) 387.
[20] J.M. Peart, M.Sc Thesis, University of Auckland, New Zealand (1983).
[21] J.P. Ryan, L.S. Goldberg and D.J. Bradley, Optics Comm. 27 (1978) 127.
[22] S.L. Shapiro, R.R. Cavanagh and J.C. Stephenson, Optics Lett. 6 (1981) 470.
[23] P.G. May, W. Sibbett, K. Smith, J.R. Taylor and J.P. Wilson, Optics Comm. 42 (1982) 285.
[24] E.W. Van Stryland, Optics Comm. 31 (1979) 93.