# V. QUANTUM ELECTRONICS

## A. Laser Applications

### Academic and Research Staff

Prof. Shaoul Ezekiel
Dr. Kent H. Casleton
Dr. Lloyd A. Hackel

### Graduate Students

Salvatore R. Balsamo
Thiruvenkatasamy Govindaraj
Robert E. Grove
Frederick Y-F. Wu

### 1. OBSERVATION OF MAGNETIC OCTOPOLE AND SCALAR SPIN-SPIN INTERACTIONS IN $I_2$ USING LASER SPECTROSCOPY

U. S. Air Force – Office of Scientific Research (Contract F44620-71-C-0051)

Shaoul Ezekiel, Lloyd A. Hackel, Kent H. Casleton

Magnetic octopole and scalar spin-spin interactions have been observed in $I_2^{127}$ by using laser-molecular beam techniques for precise measurement (one part in $10^{11}$) of the hyperfine spectrum on the P(13) 43-0 line. The values of the coupling strengths obtained from fitting the spectrum to 5 kHz are as follows: for the electric quadrupole, $eQq' = -554,094 \pm 13$ kHz, $eQq'' = -2,448,025 \pm 10$ kHz; spin-rotation, $C'-C'' = 186.71 \pm 0.10$ kHz; tensor spin-spin, $D_t'-D_t'' = -100.5 \pm 1.0$ kHz; scalar spin-spin, $D_s'-D_s'' = -2.72 \pm 1.0$ kHz; and magnetic octopole, $\Omega m'-\Omega m'' = -2.17 \pm 0.70$ kHz.

The observation of the magnetic octopole interaction, which to our knowledge is the first such observation in excited electronic states, determines a coupling strength that is the difference between upper and lower state values. An improvement in the data of at least an order of magnitude would be necessary in order to determine the constants for each state. Such an improvement is certainly possible, since the present long-term stability of the molecular beam stabilized argon laser is $1 \times 10^{-13}$ for integration times of 200 seconds.

As the precision increases further, we could consider the hexadecapole interaction as the next significant term in the Hamiltonian. In fact, we have already calculated the matrix elements for this interaction and have estimated the coupling strengths. Preliminary attempts to fit the data by including the hexadecapole term showed that the effect of this interaction is indeed small. The shift in the line positions is expected to be on the order of a few hundred hertz so that an improvement in the data of from one to two orders of magnitude would be required to observe the effect of the hexadecapole.

With the advent of dye lasers, it should now be possible to perform similar highly precise measurements over the entire optical region of the spectrum. We have already

observed the dye laser excited hyperfine structure in a molecular beam of $I_2$ with a resolution of one part in $10^9$. The measurement of the coupling strengths that are due to higher order nuclear moments would therefore make possible a more complete study of electric and magnetic field distributions in excited electronic states of atoms and molecules. Further details may be found in the references.[1-3]

References

1. L. A. Hackel, D. G. Youmans, and S. Ezekiel, "Molecular Beam Stabilized Argon Laser," Proc. Fifth International Conference on Atomic Masses and Fundamental Constants, Paris, June 2-6, 1975.
2. L. A. Hackel, K. H. Casleton, S. G. Kukolich, and S. Ezekiel, "Observation of Magnetic Octopole and Scalar Spin-Spin Interactions in $I_2$ Using Laser Spectroscopy" (to appear in Phys. Rev. Letters).
3. K. H. Casleton, L. A. Hackel, and S. Ezekiel, "Magnetic Octopole Interaction in $I_2$," Proc. Second Laser Spectroscopy Conference, Megève, France, June 23-28, 1975.

JSEP

## 2. MEASUREMENT OF THE SPECTRUM OF SPONTANEOUS EMISSION INDUCED BY MONOCHROMATIC EXCITATION

Joint Services Electronics Program (Contract DAAB07-74-C-0630)

Shaoul Ezekiel, Robert E. Grove, Frederick Y-F. Wu

We are conducting experiments with our stable single-frequency cw dye laser to make precision measurements of the spectrum of the resonance fluorescence induced by monochromatic fields. In our first experiment we are using a highly collimated atomic beam of sodium atoms that is excited orthogonally by a single-frequency cw dye laser. The laser-induced fluorescence perpendicular to both laser and atomic beams is collimated by means of small apertures and then passed through a scanning Fabry-Perot interferometer that has a 1-MHz inherent linewidth. We have already observed the predicted three-peak spectrum in preliminary experiments but our techniques have to be greatly refined before quantitative measurements can be made.

JSEP

# V. QUANTUM ELECTRONICS

## B. Gaseous Lasers

### Academic and Research Staff

Prof. William P. Allis
Prof. E. Victor George
Prof. Hermann A. Haus
Prof. Abraham Szöke
Dr. Arthur H. M. Ross
Dr. Ehud Zamir
John J. McCarthy
William J. Mulligan

### Graduate Students

Peter L. Hagelstein
John L. Miller
Charles W. Werner
David W. Wildman

## 1. DETERMINATION OF ELECTRON DENSITY FROM FREE-FREE ABSORPTION MEASUREMENTS OF A $CO_2$ LASER BEAM

JSEP

University of California, Livermore (Subcontract 7877409)
Joint Services Electronics Program (Contract DAAB07-74-C-0630)

E. Victor George, Walter P. Lapatovich, Charles W. Werner, Ehud Zamir

Recent advances in the development of high-pressure rare-gas excimer lasers have made apparent the necessity of understanding the basic mechanisms that are operative in the kinetic scheme of these systems. Of primary importance is a knowledge both of the magnitude and temporal evolution of the electron density, since many kinetic processes depend directly upon this parameter. Certain reaction rates, notably those that contribute to the production of electrons in the plasma afterglow, may be determined from the decay of the electron density at late times. We report here a determination of the electron density by measuring the temporal evolution of the free-free absorption caused by inverse Bremsstrahlung of a single-mode $CO_2$ laser beam passing through the active plasma region. The classical expression for a free-free absorption coefficient is

$$\alpha(\omega) = \frac{4\pi}{3c}\left(\frac{\omega_p}{\omega}\right)^2 \int_0^\infty \frac{\nu_m(v)}{1+\left(\frac{\nu_m(v)}{\omega}\right)^2} v^3 \frac{\partial f(v)}{\partial v}\, dv,$$

where $\omega_p$ is the plasma frequency, $\omega$ is the frequency of the probing laser, $\nu_m(v)$ is the collision frequency for momentum transfer between electrons and atoms, and f(v) is the electron distribution function normalized to unity. Kinetic modeling of the rare-gas systems indicates that the electron temperature lies between 0.5 eV and 1.5 eV. For this temperature range the absorption coefficient is a fairly strong function of the electron temperature. For this reason, interpretation of experimental results was carried

JSEP

JSEP

out by comparison with the predictions of a theoretical model.

Measurements of free-free adsorption in He, Ar, Kr, and Xe were carried out for several values of electron-beam energy deposition, at different pressures and distances from the input foil.

Typically, the decay of the free-free absorption, in particular at large energy depositions and high pressure, includes a fast component and a much slower component at later times. The fast component is anticipated because of the large dissociative recombination rate at these electron densities. The slow component may be explained by considering a reaction such as $Xe_2^* + Xe_2^* \rightarrow Xe_2^+ + 2Xe + e$, which elevates the electron density at later times, before the excimer population has decayed via radiation.

A comparison was made between theoretical and experimental absorption for various values of the rate coefficient for this process. The best fit was obtained for the value $8 \times 10^{-11}$ $cm^3/s$, at room temperature.

The peak theoretical value of electron density for 300 psia of Xe and 30 kV charging voltage of the Febetron 706 electron gun was $9.6 \times 10^{15}$ $cm^{-3}$.

## 2. HIGH-PRESSURE $CO_2$ LASER

Joint Services Electronics Program (Contract DAAB07-74-C-0630)

E. Victor George, John L. Miller, Arthur H. M. Ross

A preionization technique and discharge electrode configuration has been developed to produce arc-free discharges with the high-pressure discharge tube[1] in a mixture of $CO_2$:$N_2$:He = 1:1:8 with total pressure to 18 atm (250 psig). The gain of the high-pressure medium has been measured on various lines of the P and R branches of the 10.4 μm and 9.4 μm $CO_2$ bands, at several pressures to 16 atm. The data show an apparent increase in the gain of the P branch (with respect to that of the R branch) of the 10.4 μm band. This gain has been attributed to a contribution from an overlapping "hot" band. The gain measurements, and the high-pressure laser system have been described elsewhere.[2]

To gain further understanding of the details of the gain spectrum of the above-atmospheric-pressure $CO_2$ laser system, absorption measurements have been performed in $CO_2$ and in mixtures of $CO_2$ and He to 18 atm pressure. To understand the data, the theory of collisional broadening of overlapping lines[3] is being applied to $CO_2$. The understanding of the contribution of the "hot" bands and interacting-overlapping line effects serves to explain various anomalies observed in the gain of high-pressure $CO_2$ lasers and allows accurate calculation of the details of the gain spectrum such as the magnitude of the gain between line centers.

JSEP

JSEP

References

1. J. L. Miller, S. M. Thesis, Department of Electrical Engineering, M. I. T., September 1973 (unpublished).

2. J. L. Miller, A. H. M. Ross, and E. V. George, Appl. Phys. Letters 26, 523 (1975).

3. M. Beranger, Phys. Rev. 111, 494 (1958).

JSEP

# V. QUANTUM ELECTRONICS

## C. Nonlinear Phenomena

### Academic and Research Staff

Prof. E. Victor George
Prof. Hermann A. Haus
Prof. Abraham Szöke
Dr. Arthur H. M. Ross

### Graduate Students

Christopher P. Ausschnitt
John L. Miller

JSEP

### 1. LASER LOCKING OVER A "WIDE" FREQUENCY RANGE

Joint Services Electronics Program (Contract DAAB07-74-C-0630)

Hermann A. Haus, Abraham Szöke

Intensity and frequency control of optical pulses is acquiring increased importance as these pulses are used in a variety of ways. It is especially important to provide optical pulses at a fairly high power level, and therefore we are studying the locking of a TEA laser to a stable $CO_2$ oscillator. This can be achieved if the natural frequency of the high-power oscillator is near that of the master oscillator.

We have previously studied the frequency characteristic of the TEA laser during a single pulse.[1,2] It was found that in the early part of the pulse the frequency varies because of the change in the refractive index of the gain medium as a consequence of the change in the population inversion as the energy is extracted from the cavity. In later stages of the pulse, the shock generated by the heating of the gas causes a much larger change in frequency. The frequency change in the early part of the pulse (~100 ns) can be minimized by proper control of the cavity length. We shall measure the cavity length using the transmission of the cavity for the stable $CO_2$ laser, and we shall control the length by using a slow servo.

In the first stage of the experiments the frequency chirp characteristics of the TEA laser pulse were studied as a function of the cavity detuning. A stable $CO_2$ laser was used as a local oscillator (frequency reference). It was found that the chirp rate in the beat note is proportional to the frequency of the beat note itself, and as the gain decreases the frequency increases. In particular, in some cases where the TEA laser happened to operate near line center the beat frequency stays constant within 2.5 MHz.

In the second stage, the length of the cavity will be controlled by feedback, and the oscillator frequency will be controlled through the injection of a cw signal from the stable oscillator. This injection usually achieves both the control of frequency and a shortening between the time of the onset of the discharge and the laser pulse. The filter

JSEP

in this time delay is also reduced. All of these phenomena will be carefully evaluated. JSEP

References

1. W. A. Stiehl, "Measurement of the Spectrum of a TEA $CO_2$ Laser," Quarterly Progress Report No. 106, Research Laboratory of Electronics, M. I. T., July 15, 1972, pp. 63-69.

2. W. A. Stiehl and P. W. Hoff, "Measurement of the Spectrum of a Helical TEA $CO_2$ Laser," Appl. Phys. Letters 22, 680 (1973).

## 2. SHORT-PULSE SATURATION OF HIGH-PRESSURE $SF_6$

Joint Services Electronics Program (Contract DAAB07-74-C-0630)

Christopher P. Ausschnitt

It is well known that $SF_6$ produces mode locking in the high-pressure $CO_2$ laser.[1,2] We believe the mode-locking mechanism to be nonlinear saturation of the $SF_6$ absorption on the time scale of the mode-locked pulses. Thus the absorption must be due, in part, to a set of levels having a relaxation time of the order of or less than the typical width of a mode-locked pulse (2 ns). In other words, the attenuation experienced by an intense 2-ns pulse in propagating through a cell of $SF_6$ should be less than that experienced by a quasi-continuous signal of the same energy.

We have completed a set of experiments that verify this hypothesis. By alternately irradiating a cell of $SF_6$, 1 mm long, with the single-mode output and the mode-locked output of a high-pressure TEA $CO_2$ laser we have measured the difference in transmission of the cell for equal values of the total energy in the TEA pulse. The single-mode and mode-locked output of the TEA laser are shown in Fig. V-1. Single-mode operation was achieved by means of an intracavity cell, 2.5 cm long, of low-pressure $SF_6$ (<1 Torr). Mode locking was produced by an intracavity acousto-optic modulator tuned to the cavity mode spacing at approximately 42 MHz. The mode-locked pulses, 2 ns long, were spaced 24 ns apart; this gives approximately a 12:1 peak power enhancement over the single-mode TEA pulse of equal energy.

The output of the TEA laser was passed through a 3-mm aperture to ensure a nearly uniform intensity distribution across the beam and focused by a salt lens to a 0.2 mm spot inside the $SF_6$ cell. A second lens recollimated the beam before it was detected by a Ge:Au detector. Different values of input intensity were achieved by placing calibrated $CaF_2$ attenuators in the path of the beam. From the detector the signal was amplified with an amplifier that was slow (~5 μs) compared with the TEA pulse (~250 ns) so that its response depended on the total energy in the TEA pulse. The amplifier output was then fed into a 400-channel pulse height analyzer that displayed the distribution of JSEP

JSEP transmitted TEA pulse energy for successive shots of the laser. The use of the analyzer enabled us to distinguish between shot-to-shot fluctuations in the laser output energy and differences in absorber transmission.

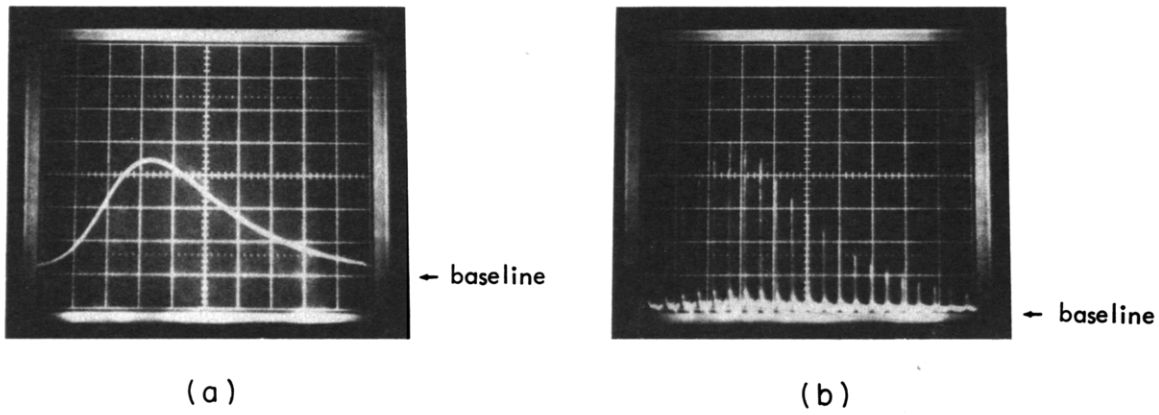


Fig. V-1. TEA $CO_2$ laser output.
(a) Single mode. Time scale: 50 ns/div. Amplitude scale: 2 kW/div.
(b) Mode locked. Time scale: 50 ns/div. Amplitude scale: 10 kW/div.

Results of the transmission measurements at various $SF_6$ pressures for the P(20) and P(22) lines of 10.6 μ $CO_2$ output are shown in Fig. V-2. The mode-locked pulse transmission is always higher than that of the single-mode pulse. Furthermore, we observe that the saturation curves appear to be asymptotic to the small-signal absorption values measured by Wood et al.[3] and thus to indicate that the absorption cross section does not change appreciably with pressure.

The complex energy level structure of $SF_6$ makes it difficult to speculate about which set of levels is saturated by the mode-locked pulses. Following in the footsteps of previous authors,[4,5] however, we can model $SF_6$ as a four-level system whose total absorption is composed of ground-state absorption and excited-state absorption. The relaxation rate of ground-state absorption has been measured[6] to be 122 μs-Torr$^{-1}$, which is slow compared with the TEA pulse even at pressures up to 250 Torr. Measurements of the excited-state absorption relaxation time indicate that it is comparable to or less than 1 ns even below 10 Torr.[7] Thus it is reasonable to assume that the higher transmission seen by the mode-locked pulses is caused by the saturation of the excited-state absorption. This hypothesis is also consistent with the fact that the difference in transmission on P(22) is larger than that observed on P(20) where the excited-state absorption is a smaller fraction of the total absorption.[6]
JSEP

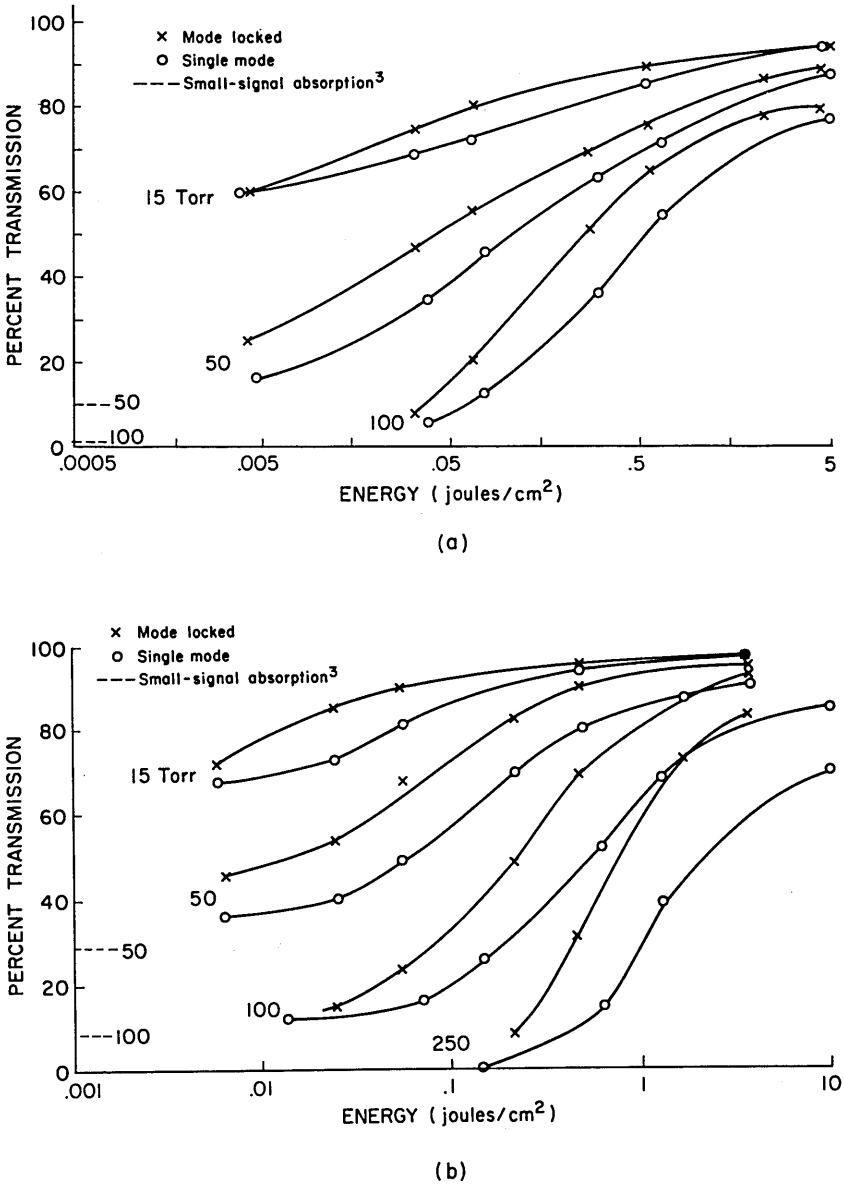


Fig. V-2. (a) Transmission on P(20) line.
(b) Transmission on P(22) line.

Work is in progress to glean information regarding the saturation intensity and relaxation time of the excited-state population from the transmission measurements.

References

1. A. Nurmikko, T. A. DeTemple, and S. E. Schwarz, Appl. Phys. Letters 18, 130 (1971).

2. R. Fortin, F. Rheault, J. Gilbert, M. Blanchard, and J. L. Lachambre, Can. J. Phys. 51, 414 (1973).

JSEP 3. O. R. Wood, P. L. Gordon, and S. E. Schwarz, IEEE J. Quantum Electron., Vol. QE-5, No. 10, p. 502, October 1969.

4. H. Brunet, IEEE J. Quantum Electron., Vol. QE-6, No. 11, p. 678, November 1970.

5. U. P. Oppenheim and Y. J. Kaufman, IEEE J. Quantum Electron., Vol. QE-10, No. 7, p. 533, July 1974.

6. J. I. Steinfeld, I. Burak, D. G. Sutton, and A. V. Nowak, J. Chem. Phys. 52, 5421 (1970).

7. J. J. Armstrong and O. L. Gaddy, IEEE J. Quantum Electron., Vol. QE-8, No. 10, p. 797, October 1972.

## 3. TRANSIENT BUILDUP OF FAST SATURABLE ABSORBER MODE LOCKING

Joint Services Electronics Program (Contract DAAB07-74-C-0630)

Christopher P. Ausschnitt

In previous investigations[1-3] the statistical nature of the onset of passive mode locking has been emphasized. We use a mode-locking equation derived by Haus[4] to show that the buildup of fast absorber mode locking in the homogeneously broadened laser is, in fact, deterministic and that the only stable steady-state solution is the hyperbolic secant pulse found by Haus. We assume that lasing initiates on the longitudinal mode nearest line center, so that the initial output of the laser is cw. This assumption is consistent with the well-known experimental observation that stable passive mode locking occurs only when the laser is operated near threshold.[5]

The equation governing mode locking by the fast saturable absorber has been shown by Haus[4] to be

$$-\frac{2}{\Delta\omega_c T_R}\frac{\partial v(t,n)}{\partial n} = \left[1 + q - q\frac{|v(t,n)|^2}{P_A} - g(n)\left(1 + \frac{1}{\omega_L^2}\frac{\partial^2}{\partial t^2}\right)\right] v(t,n), \qquad (1)$$

where v is the field envelope; t the time "local" to a single cavity round-trip transit time; n the integer that counts the number of round-trip transits, treated as a continuum variable over many transits; $T_R$ the round-trip transit time in the cavity; $\Delta\omega_c$ the cavity-mode linewidth; q the small-signal absorber loss; $P_A$ the saturation power of the absorber; g the saturated gain; and $\omega_L$ the laser medium half-width. The saturated gain is related to the power in a single round-trip transit by

$$g(n) = \frac{g_o(n)}{1 + P(n)/P_L}, \qquad (2)$$

JSEP

where $g_o(n)$ is the known small-signal gain, $P_L$ is the saturation power of the homogeneously broadened laser medium, and the power is

$$P(n) = \frac{1}{T_R} \int_{-T_R/2}^{T_R/2} |v(t, n)|^2 \, dt. \tag{3}$$

Haus has shown that a stable steady-state solution ($\partial v/\partial n = 0$) to (1) is a single pulse

$$v(t) = \frac{v_o}{\cosh t/\tau_p}, \tag{4}$$

where the pulse parameters $v_o$ and $\tau_p$ are determined by the system parameters through the eigenvalue relations

$$1 + q - g = \frac{q}{2} \frac{v_o^2}{P_A} = \frac{g}{\omega_L^2 \tau_p^2} \tag{5}$$

in conjunction with (2). We show here that (4) is the only stable steady-state solution to (1). Thus all other solutions evolve into (4).

A logical guess for the general solution to (5) is of the form of a slowly varying envelope times a waveform v' describing the field in each transit

$$v(t,n) = \exp\left(\int^n a(n)\, dn\right) v'(t,n). \tag{6}$$

Thus we approximate the continuously varying waveform by a succession of steady-state waveforms in each transit that evolve slowly over many transits. For the solution (6), the left-hand side of (1) becomes

$$\frac{\partial v}{\partial n} = \left(a + \frac{1}{v'} \frac{\partial v'}{\partial n}\right) v. \tag{7}$$

To be consistent with our slowly varying envelope approximation, we assume

$$a \gg \frac{1}{v'} \frac{\partial v'}{\partial n} \tag{8}$$

so that (7) becomes

$$\frac{\partial v}{\partial n} \approx a v. \tag{9}$$

Equation 1 can now be normalized to the form

$$\left[1 + \beta(n) - \frac{\partial^2}{\partial x^2}\right] y(x,n) = 2y^3(x,n), \tag{10}$$

where

JSEP

$$y(x,n) = \frac{v\left(\frac{t}{\tau_p}, n\right)}{V_o} \tag{11}$$

$$V_o = \sqrt{\frac{2P_A[1+q-g(n)]}{q}} \tag{12}$$

$$\tau_p(n) = \frac{1}{\omega_L}\sqrt{\frac{g(n)}{1+q-g(n)}} \tag{13}$$

$$\beta(n) = \frac{\frac{2}{\Delta\omega_c T_R}\alpha(n)}{1+q-g(n)}. \tag{14}$$

Equation 10 integrates to the elliptic form

$$\int_{y(0,n)}^{y(x,n)} \frac{dy}{\sqrt{P(y)}} = x(n), \tag{15}$$

where

$$P(y) = -y^4 + [1+\beta(n)]y^2 + C(n) \tag{16}$$

and C(n) is the constant of integration. The solutions to (10) are determined by the polynomial P(y) which is the potential well of Eq. 10. In particular, the roots of P(y) are the turning points of the function x(y). If we solve for the roots of (16) they will be functions of $\beta$ and C. But from our normalization (11) we have already specified that the maximum value of y must equal but cannot exceed 1. Thus one of the turning points, and hence one of the roots of (16), must lie at y = 1. This requirement imposes the constraint

$$\beta(n) = -C(n) \tag{17}$$

so that the two roots of (16) become

$$y_1^2 = 1$$

$$y_2^2 = -C(n). \tag{18}$$

For the case $-1 < C(n) < 0$ integration of (15) in terms of elliptic functions yields

$$y = \sqrt{1 - [1+C(n)]\,\mathrm{sn}^2\, x(n)} \tag{19}$$

JSEP where sn has modulus 1 + C(n). Equation 19 reduces to the hyperbolic secant solution

of Haus (4) as $C \to 0^-$, and has the period

$$D(n) = 2K[1 + C(n)], \tag{20}$$

where K is the complete elliptic integral of the first kind.

The form of the polynomial P(y) and the solution (19) for several values of C(n) are shown in Fig. V-3. The solution is a constant initially, then breaks into oscillation, and

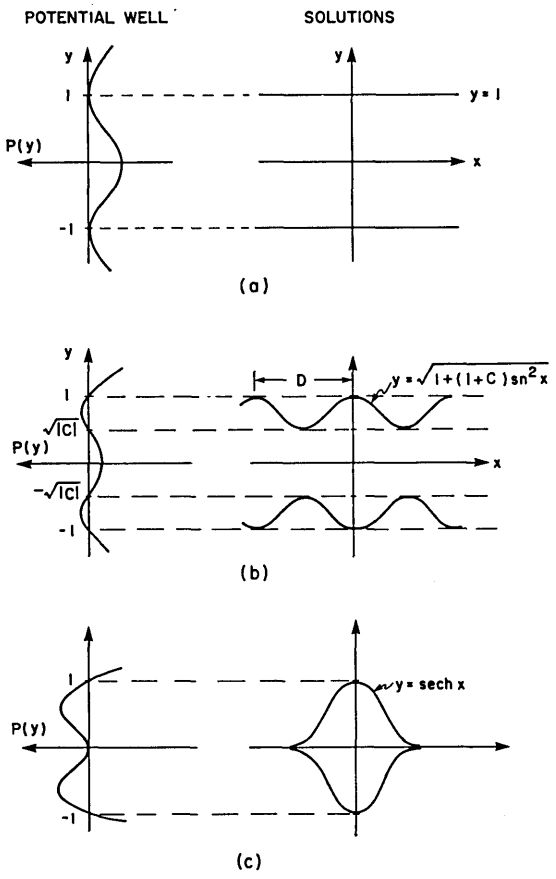


Fig. V-3.
Potential well and solutions for various values of C during the transient buildup. (a) $C = -1$, $D = \pi$. (b) $-1 < C < 0$. (c) $C = 0^-$, $D = \infty$.

ends up as a single pulse as C goes from -1 to 0. Note that for $-1 < C < 0$ Eqs. 17 and 14 require that the solution be still growing. For $C > 0$, however, they require that the solution decay. Thus the solution at $C = 0^-$, that is, Eq. 4, is the only stable steady-state solution.

Making use of (19), we can obtain an expression for the energy in a given period

$$\mathcal{E}(n) = \int_{-D(n)/2}^{D(n)/2} y^2(n)\, dx = 2E[1 + C(n)], \tag{21}$$

where E is the complete elliptic integral of the second kind.

JSEP

By noting that the condition

$$D(n)\,\tau_p(n) = T_R, \tag{22}$$

which originates from our normalization of the time scale, $x = t/\tau_p$, determines the value of C(n) for each value of n, we completely determine the transient buildup of the mode-locking solution (4) from cw.

In contrast to previous analyses[1-3] of the transient buildup of passive mode locking we have shown that the evolution of the output of a laser containing a fast saturable absorber from cw to short pulse is deterministic and does not rely on the selection of a pulse from noise. Furthermore, there is only one stable short-pulse solution. In a future report we shall discuss the precise behavior of the laser near the threshold of cw operation.

References

1. P. G. Kryukov and V. S. Letokhov, IEEE J. Quantum Electron., Vol. QE-8, No. 10, p. 766, October 1972.
2. T. I. Kuzuetsova, Sov. Phys. — JETP 30, 904 (1970).
3. B. Hausherr, E. Mathieu, and H. Weber, IEEE J. Quantum Electron., Vol. QE-9, No. 4, p. 445, April 1973.
4. H. A. Haus, "Theory of Mode Locking with a Fast Saturable Absorber" (to appear in J. Appl. Phys.).
5. R. Fortin, F. Rheault, J. Gilbert, M. Blanchard, and J. L. Lachambre, Can. J. Phys. 51, 414 (1973).

JSEP