RESEARCH IN NONLINEAR OPTICS

Final Report

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Technical Discussion

The main concern of the research under this grant was to investigate theoretically and experimentally the possibility of passive mode locking and the concomitant generation of ultrashort optical pulses in liquids displaying the Kerr effect. Such liquids are made up of anisotropic molecules which perform an oscillation in the presence of an optical field containing more than one frequency. This oscillation at the beat frequency is equivalent to a time dependent index modulation which in an FM manner generates side bands, thus coupling energy between the laser modes.

A theoretical investigation revealed that a steady state mode-locked solution as appropriate to ultrashort pulses is induced by the Kerr liquids. An experimental investigation using a Q-switched ruby laser ($\sim$ 100 Mw/cm^2) passively mode-locked by the insertion of a Kerr liquid verified the theory. Pulses of $\sim 10^{-11}$sec have been generated when the relaxation time of the liquid was temperature tuned to approximately 10^{-11}sec.

Work in this area is continuing beyond the termination date of the grant. Also, recent work in the Phillips Research Laboratories uses this approach for mode locking ruby lasers.

The details of the theory and experiments are contained in the enclosed reprints.

List of Enclosed Reprints

(1) J. P. Laussade and A. Yariv, "Mode Locking and Ultrashort Laser Pulses by Anisotropic Molecular Liquids", Appl. Phys. Lett. 13, 65 (1968).

(2) J. Comly, E. Garmire, J. P. Laussade, and A. Yariv, "Observation of Mode Locking and Ultrashort Optical Pulses Induced by Anisotropic Molecular Liquids", Appl. Phys. Lett. 13, 176 (1968).

(3) J. P. Laussade, A. Yariv, and J. Comly, "Optical Communication through Random Atmospheric Turbulence", Appl. Optics 8, 1607 (1969).

(4) J. P. Laussade and A. Yariv, "Analysis of Mode Locking and Ultrashort Laser Pulses with a Nonlinear Refractive Index", IEEE J. Quantum Electr. QE-5, 435 (1969).

Reprinted from

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MODE LOCKING AND ULTRASHORT LASER PULSES BY ANISOTROPIC MOLECULAR LIQUIDS*

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(anisotropic molecular liquids; nitrobenzene; 10^{-11}-sec pulses at 126°C; T/E)

Spectral broadening by the nonlinear dielectric response of anisotropic molecules has been observed and explained by Bloembergen and Lallemand.[1] Any two frequencies, say ω_1 and ω_2, present in the incident optical beam exert a torque on the molecules at a frequency $(\omega_1 - \omega_2)$. The resultant oscillation of the average molecular orientation causes a modulation of the dielectric constant at $\omega_1 - \omega_2$. A third frequency ω_3 experiencing this modulation acquires side-band components at $\omega_3 \pm l(\omega_1 - \omega_2)$ $l = 1, 2, 3, \cdots$.

In this Letter we consider theoretically the case of an anisotropic molecular liquid (henceforth designated as AML) placed within the optical resonator of a multimode laser. In this case the frequencies ω_1 and ω_2 correspond to two longitudinal modes so that the side-band energy at $\omega_3 \pm l(\omega_1 - \omega_2)$ coincides in frequency with already oscillating laser modes. The presence of the AML is thus seen to give rise to power exchange between the equispaced laser modes.

In the following we show that this exchange of energy gives rise, as in the internally modulated laser[2,3] to a phase-locked spectrum characteristic of the ultrashort pulse mode of oscillation, where the pulses are separated by the double transit time $2L/c$ and should approach, in the limit, a duration $\tau \sim (\Delta\nu_{\text{gain}})^{-1}$ where $\Delta\nu_{\text{gain}}$ is the linewidth of the amplifying transition. Such pulses are now obtainable from giant pulse lasers containing a saturable absorber.[4,5]

Our task consists of solving for the complex normal mode amplitudes D_n of an optical resonator containing an AML. These amplitudes are defined as in ref. 3 by the following expansion for the total electric and magnetic fields

$$\bar{E}(\bar{r}, t) = \sum_n i\sqrt{\frac{\omega_n}{2\epsilon}} D_n(t)\, e^{-i\omega_n t}\, \bar{E}_n(\bar{r}) + \text{c.c.}$$

$$\bar{H}(\bar{r}, t) = \sum_n \sqrt{\frac{\omega_n}{2\mu_0}} D_n(t)\, e^{-i\omega_n t}\, \bar{H}_n(\bar{r}) + \text{c.c.} \qquad (1)$$

The summation is over the longitudinal modes whose resonant frequencies are ω_n. $\bar{E}_n(\bar{r})$ and $\bar{H}_n(\bar{r})$ are the vector spatial eigenmodes as defined by Slater.[6] In a linear medium the modes D_n are independent of each other. The presence of an AML spoils this independence and forces a unique phase and amplitude relationship upon the modes.

The mode expansion (1) is substituted into Maxwell's equation. The nonlinear dielectric properties of the medium are accounted for by[1]

$$P^{\omega_3+\omega_1-\omega_2}(\bar{r}, t) = \frac{\epsilon_2}{1 + i(\omega_1 - \omega_2)\tau} \times g(\bar{r}) E_1 E^*_2 E_3\, e^{i(\omega_3+\omega_1-\omega_2)t} + \text{c.c.} \quad (2)$$

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where ϵ_2 is the optical Kerr constant, τ is the Debye molecular reorientation time, and $g(\bar{r})$ accounts for the partial filling of the resonator volume by the AML. The equations of motion for the modes D_n are obtained by the usual procedure[7] of dot-multiplying Maxwell's curl equations by $\bar{E}_n(\bar{r})$ and $\bar{H}_n(\bar{r})$ and integrating over the resonator volume. The complete analysis is of very considerable difficulty. In order to avoid getting lost in mathematical detail we limit our consideration to all mode pairs ω_1 and ω_2 separated by some multiple, say s, of the intermode frequency ω so that $\omega_1 - \omega_2 = s\omega$. In addition we assume that: (a) a very large number of oscillating modes exists so that spectral end effects are negligible. This assumption can be shown to be equivalent to the neglect of molecular viscous damping; (b) the gain is the same for all the modes and is equal to the loss. If we define the amplitude and phase of D_n through

$$D^*_n(t) = \beta_n(t)\, e^{in\phi(t)} \tag{3}$$

we obtain

$$\beta'(t) + in\phi'(t)\beta = \frac{-3i\omega_n\epsilon_2 f}{2\epsilon^2 V}\left(1 + \frac{1}{1 + s^2\omega^2\tau^2}\right)\mathscr{E}\beta \tag{4}$$

where V is the resonator volume, f is the geometrical filling factor for the AML cell and ϵ is the total stored electromagnetic energy. Since the right side of Eq. (4) is purely imaginary, we obtain the following solution for the normal mode

$$D^*_n(t) = \beta \exp\left\{-i\omega_n t\left[\frac{3\epsilon_2 f\mathscr{E}}{2\epsilon^2 V}\left(1 + \frac{1}{s^2\omega^2\tau^2}\right)\right]\right\}. \tag{5}$$

The AML is thus seen to give rise to a mode-locked spectrum of equal amplitudes and zero phases, so that the optical envelope consists of a train of ultrashort pulses as discussed above. We notice in passing that the factor within the square brackets of Eq. (5) represents frequency pulling which is proportional to the stored energy $\mathscr{E}$.

As a measure of the strength of the mode coupling we define a circulation time T_0 as the exponential time constant for the circulation of the energy in one mode due to its interaction with all the others. Again, if we limit our attention to mode pairs ω_1, ω_2 such that $\omega_1 - \omega_2 = s\omega$, we obtain the partial contribution to T_0^{-1}

$$\left(\frac{1}{T_0}\right)_s = 6\frac{\omega_n\epsilon_2 f\mathscr{E}}{\epsilon^2 V}\left(\frac{s\omega\tau}{1 + s^2\omega^2\tau^2}\right). \tag{6}$$

Since $T_0^{-1} = \Sigma_s (T_0^{-1})_s$ we expect from Eq. (6) that the shortest circulation time T_0, hence the strongest mode coupling, obtains when the magnitude of the reorientation time τ satisfies

$$\tau \sim \frac{1}{s_{\max}\omega} = \frac{1}{(\Delta\omega)_{\text{gain}}} \tag{7}$$

where $(\Delta\omega)_{\text{gain}}$ is the gain linewidth of the laser transition.

In an experiment performed in collaboration with J. Comly and Dr. E. Garmire, mode locking and pulses of $\sim 10^{-11}$ sec duration were observed. The setup consisted of a Q-switched ruby laser containing a 5-cm nitrobenzene cell in its optical path. Ultrashort pulses were observed only upon heating the nitrobenzene to $T > 126$°C at which point τ becomes small enough ($\sim 10^{-11}$ sec) to satisfy condition (7). The circulation time T_0 at that point is estimated to be less than 10^{-9} sec so that modes lock together in a time short compared to the duration of a single giant pulse.

A detailed description of the experiment is in preparation.

The authors are indebted to J. Comly and Dr. E. Garmire for a number of stimulating discussions.

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[5] A. J. DeMaria, D. A. Stetser, and H. Heynau, *Appl. Phys. Letters* **8**, 174 (1966).
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OBSERVATION OF MODE LOCKING AND ULTRASHORT OPTICAL PULSES INDUCED BY ANTISOTROPIC MOLECULAR LIQUIDS*

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Mode locking and ultrashort pulses have been produced in a giant-pulse ruby laser with heated nitrobenzene ($T > 110$°C) or α-chloronaphthalene ($T > 62$°C) inside the optical resonator. 10^{-11}-sec pulses were observed with the two-photon fluorescence technique.

In a previous article[1] two of the present authors analyzed the effect of anisotropic molecular liquid on a multilongitudinal mode laser. It was shown that for sufficiently high oscillation power and at a temperature where the molecular reorientation time τ is comparable to the inverse of the laser gain linewidth $\Delta\nu_{\text{gain}}$, the dielectric nonlinearity of the anisotropic molecular liquid should give rise to mode coupling and, consequently, to ultrashort pulses.

In this Letter we describe the experimental observation of mode locking and ultrashort pulses in a giant-pulse ruby laser induced by the presence of

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either α-chloronaphthalene or nitrobenzene inside the optical resonator.

The experimental setup consisted of a giant-pulse ruby laser employing a rotating prism with an average power density of about 10 MW/cm² and a 1-m cavity. Internal reflections were minimized through the use of Brewster-angle surfaces and a wedge output mirror. A Brewster-angle, 5-cm cell containing the liquid was placed inside the resonator. The liquid temperature could be controlled over the range −95°C to 140°C.

In Fig. 1 we show a typical two-photon fluorescence[2] display of the output pulse at .694 μ with *no* liquid within the resonator. The (longitudinal) uniformity of the exposure indicates that no ultrashort pulses are present. The introduction of α-chloronaphthalene inside the resonator gives rise to ultrashort pulses as seen in Fig. 2. The temperature of the liquid is 62°C, and the relaxation time τ is estimated to be in the range 2×10^{-11} to 4×10^{-11} sec. The pulse duration is estimated from Fig. 2 to be $\tau_{\text{pulse}} \sim 1.2 \times 10^{-11}$ sec.

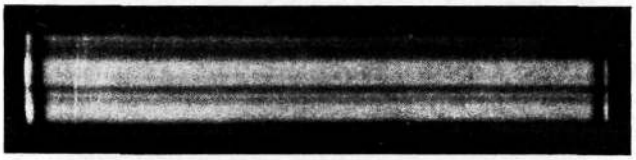

Fig. 1. The two-photon fluorescence display of rhodamine 6G with no anisotropic liquid inside the laser resonator. The longitudinal uniformity indicates the lack of ultrashort pulses. The mirror was placed so that pulses separated by a round-trip cavity time would overlap in the dye.

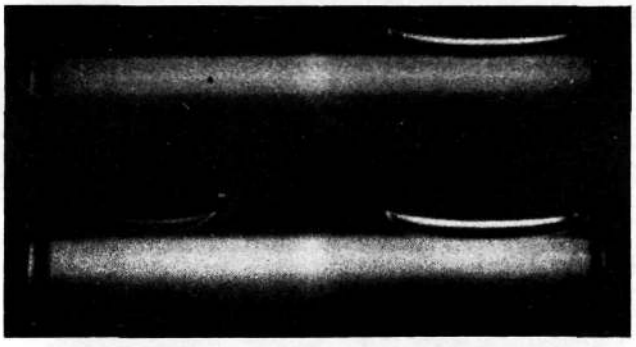

Fig. 2. Typical two-photon fluorescence displays obtained when a 5-cm cell containing α-chloronaphthalene at 62°C is present inside the resonator. The pulse length is approximately 1.2×10^{-11} sec.

Similar results were obtained using nitrobenzene except that ultrashort pulses occurred reliably *only* when its temperature was raised above 110°C. At this temperature, $\tau \sim 1.3 \times 10^{-11}$ sec (compared to a room temperature value of 4.3×10^{-11} sec). The gain linewidth $(\Delta\nu)_{\text{gain}}$ was measured with a Fabry-Perot etalon at ~1.8 cm⁻¹. The expected pulse width is[3] $\sim(1/(\Delta\nu)_{\text{gain}})\ 1.8 \times 10^{-11}$ sec, which is comparable to the measured value $\tau_{\text{pulse}} \sim 1.2 \times 10^{-11}$, indicating mode locking across the full gain line-width.

A number of interesting observations made in the course of this investigation concerns the process of stimulated Raman emission (SRE) in the anisotropic molecular liquid inside the optical resonator. When the cell containing α-chloronaphthalene was placed outside the resonator and in the absence of ultrashort pulses, we observed self-trapping[4] accompanied by SRE in the α-chloronaphthalene. When the cell is introduced inside the resonator no SRE is observed. This is most likely due to the increase in threshold for self-trapping which occurs when τ_{pulse} becomes comparable to or shorter than the molecular relaxation time τ.[5]

With nitrobenzene inside the resonator we observed SRE both at $T < 110$°C, where ultrashort pulses are infrequent, and at $T > 110$°C where ultrashort pulses occur regularly.

The following observations suggest that in both of these cases the SRE takes place *without* self-trapping:

(a) a very nearly exponential dependence of the Raman intensity on that of the laser, with a gain coefficient comparable to the theoretical value; and

(b) the lack of SRE in α-chloronaphthalene which has nearly the same threshold for self-trapping and relaxation time as nitrobenzene.

In Fig. 3 we see on the oscillograph both the laser output and the Raman light generated from a nitrobenzene cell at 90°C inside the cavity. Note the fast rise of Raman output with small increase of laser power. Furthermore, the Raman output is accompanied by substantial smoothing in the laser intensity fluctuations, suggesting the appearance of parametric saturation.[6]

The use of a nonlinear dielectric constant inside the laser cavity may be a very practical way of generating ultrashort pulses. Present techniques generally use a saturable absorber, which tends to enhance the filamentary structure of the laser output, often causing damage to laser components. The liquids used here reduced the filamentary nature of the beam even when compared to that without any liquid. This can be seen by comparing the transverse structures in Figs. 1 and 2. We found no damage to components when ultrashort pulses were generated by the anisotropic molecular liquids,

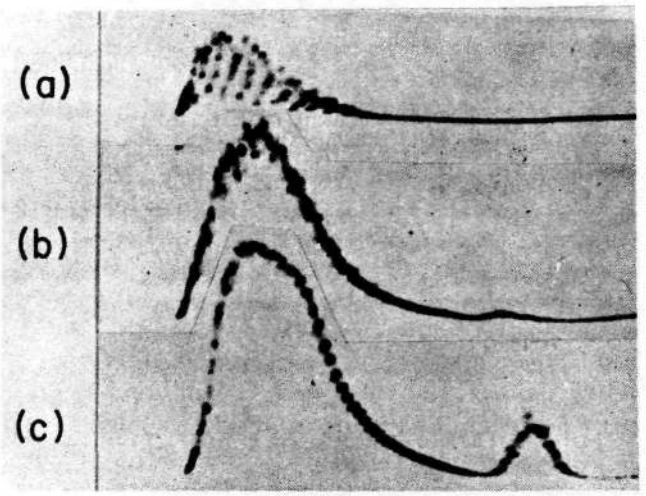

Fig. 3. Laser oscillation and stimulated Raman emission from nitrobenzene at 90°C inside the laser cavity. (*a*) Single pulse showing the large fluctuations in the laser output below the onset of SRE with a width of 40 nsec. (*b*) At an increased pumping level the very beginning of SRE is seen in the small pulse after the main laser pulse. The SRE pulse is delayed electronically by 10^{-7} sec so that a simultaneous single-trace display is possible. (*c*) At even higher pumping level; the laser intensity fluctuations are largely smoothed out.

while it was a frequent problem with saturable absorbers.

The bandwidth of the nonlinear dielectric coupling is determined by the molecular reorientation time, which can be varied by temperature tuning or by changing liquids. The possibility of using this effect for producing ultrashort pulses of variable length is being investigated.

The authors acknowledge the excellent technical assistance of D. Armstrong.

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Optical Communication Through Random Atmospheric Turbulence

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In this paper we compare the theoretical performances of two schemes of optical communication through the atmospheric turbulence: (1) heterodyne detection and (2) video detection. The signal-to-noise ratios (S/N) in the output current of a detector are expressed for both schemes in terms of the correlation function of the refractive index fluctuations of the turbulence. The results of a separate theoretical analysis of optical wave propagation through a random turbulence are used in order to obtain a numerical estimate of the performance criterion $(S/N)_{(2)}/(S/N)_{(1)}$ in terms of the length of propagation through the atmosphere, the turbulence strength, the wavelength of the optical wave, and the diameter of the receiving aperture.

I. Introduction

In this paper we consider the effect of propagation through a turbulent atmosphere on two archetypes of optical communication schemes: (1) a superheterodyne and (2) a video communication system.

We are aiming our analysis specifically at electrooptic modulation systems which, fundamentally, control the *phase* of the optical carrier. This modulation is often converted into amplitude modulation by means of polarizers, or recovered as phase modulated intermediate frequency signal in a heterodyne detection system. Since our main interest in this analysis is in the loss of signal information due to random *phase* fluctuations and not in the many possible detection schemes, we define, somewhat arbitrarily, our signal power as that of the modulation frequency sidebands in the output of the optical nonlinear detector. In the case of the heterodyne system, the detector output, being phase modulated, requires further demodulation to recover the signal. It is assumed that this process does not change the signal-to-noise ratio materially, so that the comparison between the video and heterodyne schemes can be carried out at the same point of the communication link, i.e., at the output of the first (optical) detector.

To be specific, in scheme (1) the linearly polarized output field of a laser oscillating at ω_0 is phase modulated at a frequency ω_m. After propagating through a turbulent atmosphere, it is incident on the surface of a nonlinear optical detector along with a local oscillator signal[1] whose frequency is $\omega_0 - \Delta\omega$. For the purposes of this analysis, we define the signal as the components of the detector current oscillating at $(\Delta\omega \pm \omega_m)$.

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In scheme (2), the video communication system, the phase modulation at ω_m is impressed on one of the two mutually orthogonal polarizations which initially add up to form the linearly polarized laser field. These two polarizations, having traversed together the turbulent path, are mixed in the nonlinear detector to produce an output current at ω_m to which we refer as the signal. From the point of view of signal-to-noise ratio, the best we can do is have the main source of noise in the output of the detector be the shot noise generated by the local oscillator in case (1) or the optical signal itself in case (2).[2,3]

The main purpose of this work is to apply the results of a theoretical analysis of propagation through random atmospheric turbulence[4] to evaluate the signal-to-noise ratio following detection in the cases of a superheterodyne receiver and a video receiver as described above. The relative performance of these systems is calculated as a function of optical wavelength, the path length, the receiver diameter, and the strength of the turbulence.

II. Signal-to-Noise Ratio in Optical Mixing Detection

In the most general case, we assume that in the plane of the detector the reference and the information signals have random amplitudes and phases due to the random fluctuations of the refractive index along the propagation path in the atmosphere. At a point defined by the coordinates $(L,\mathbf{r})$ in the plane of the detector, the modulated optical electric field is

$$E_S(L,\mathbf{r}) = A_S(L,\mathbf{r}) \exp\{i[\omega_0 t + \phi_m(t) + \phi_s(L,\mathbf{r})]\}, \quad (1)$$

where L is the length of the communication link, $A_S(L,\mathbf{r})$ are the random amplitude and phase due to the turbulent nature of the transmission medium, ω_0 is the frequency of the optical wave, $\phi_m(t)$ is the modulation

phase containing the information. We consider the case of harmonic phase modulation so that $\phi_m(t) = \delta \sin \omega_m(t)$, where δ is the modulation index and ω_m is the (angular) modulation frequency.

The complex reference electric field is taken as

$$E_R(L,\mathbf{r}) = A_R(L,\mathbf{r}) \exp\{i[(\omega_0 - \Delta\omega)t + \phi + \phi_R(L,\mathbf{r})]\}, \quad (2)$$

where $A_R(L,\mathbf{r})$ and $\phi_R(L,\mathbf{r})$ are the random amplitude and phase due to the turbulent transmission medium; ϕ is a constant phase factor; and $\Delta\omega$ is the frequency offset from the carrier frequency. (For video detection, $\Delta\omega = 0$.) The total electric field in the plane of the detector is the sum of the information and reference electric fields:

$$E_T(L,\mathbf{r}) = E_S(L,\mathbf{r}) + E_R(L,\mathbf{r}), \quad (3)$$

and the output current of the nonlinear detector (after averaging over a few optical periods) is taken as

$$i(t) = \frac{\eta q}{2h\nu} \int_\Sigma d\mathbf{r} E_T(L,\mathbf{r}) E_T^*(L,\mathbf{r}), \quad (4)$$

where η is the quantum efficiency of the detector, ν equals $\omega_0/2\pi$, q is the electronic charge, and Σ is the area of the detector.

The output current $i(t)$ has a dc part i_{DC} and oscillating components at sideband frequencies $\Delta\omega \pm \omega_m$, where $m = \pm 1, \pm 2, \ldots$. Here we consider only the part of the current oscillating at frequencies $\Delta\omega \pm \omega_m$. Since this oscillating component is the replica of the modulated information, we call it $i_S(t)$. From Eqs. (1)–(4) with $\phi = \pi/2$,† we find

$$i_{DC} = \frac{\eta q}{2h\nu} \int_\Sigma d\mathbf{r}[A_S^2(L,\mathbf{r}) + A_R^2(L,\mathbf{r})], \quad (5)$$

and

$$i_S(t) = \frac{\eta q}{h\nu} 2J_1(\delta) \sin\omega_m t \int_\Sigma d\mathbf{r} A_R(L,\mathbf{r}) A_S(L,\mathbf{r}) \times \cos[\Delta\omega t + \Delta\phi(L,\mathbf{r})], \quad (6)$$

where $J_1(\delta)$ is the first order Bessel function and $\Delta\phi(L,\mathbf{r})$ is the random phase difference between the information and the reference signal:

$$\Delta\phi(L,\mathbf{r}) = \phi_S(L,\mathbf{r}) - \phi_R(L,\mathbf{r}). \quad (7)$$

Equations (5) and (6) are used below to derive expressions for the signal-to-noise ratio in heterodyne and video communication schemes.

III. Signal-to-Noise Ratio for Superheterodyne Detection

In a heterodyne detection scheme, the amplitude of the reference signal is constant over the plane of the detector $A_R(L,\mathbf{r}) = A_R$ and the phase does not contain any fluctuations $\phi_R(L,\mathbf{r}) = 0$. In this case, $\Delta\phi(L,\mathbf{r}) = \phi_{(1)}(L,\mathbf{r})$ where $\phi_1(L,\mathbf{r})$ is the phase of the modulated optical signal. The subscript (1) refers to heterodyne detection.

From Eq. (6) the signal part of the current is

$$i_S(t) = \frac{\eta q}{h\nu} 2J_1(\delta) A_R \sin\omega_m t \int_\Sigma d\mathbf{r} A_{(1)}(L,\mathbf{r}) \times \cos[\Delta\omega t + \phi_{(1)}(L,\mathbf{r})]; \quad (8)$$

$i_S(t)$, as expressed by Eq. (8), is the result of the beating of both sidebands of the signal beam at frequencies $\omega_0 + \omega_m$ and $\omega_0 - \omega_m$ with the reference beam at frequency $\omega_0 - \Delta\omega$. In the remainder of this analysis, we use a slightly different expression for $i_S(t)$. We consider the current obtained by twice the contribution of a single sideband. It is expressed as

$$i_S(t) = \frac{\eta q}{h\nu} 2J_1(\delta) A_R \int_\Sigma d\mathbf{r} A_{(1)}(L,\mathbf{r}) \sin[(\Delta\omega + \omega_m) \times t + \phi_{(1)}(L,\mathbf{r})]. \quad (9)$$

It is simpler to relate $i_S(t)$ as expressed by Eq. (9) to known functions describing the turbulence, and the final result for S/N is within a factor of one-half of the correct result which could be obtained from Eq. (8).

The signal power in the output current of the detector $S_{(1)}(t)$ is defined as $S_{(1)}(t) = i_S^2(t)$. From Eq. (9),

$$S_{(1)}(t) = \left(\frac{\eta q}{h\nu}\right)^2 4J_1^2(\delta) A_R^2 \int_\Sigma d\mathbf{r}_1 \int_\Sigma d\mathbf{r}_2 A_{(1)}(L,\mathbf{r}_1) A_{(1)}(L,\mathbf{r}_2) \times \sin[(\Delta\omega + \omega_m)t + \phi_{(1)}(L,\mathbf{r}_1)] \sin[(\Delta\omega + \omega_m)t + \phi_{(1)}(L,\mathbf{r}_2)]. \quad (10)$$

The signal power $S_{(1)}$ averaged over a time large compared with $2\pi/(\Delta\omega + \omega_m)$ is

$$S_{(1)} = \left(\frac{\eta q}{h\nu}\right)^2 2J_1^2(\delta) A_R \int_\Sigma d\mathbf{r}_1 \int_\Sigma d\mathbf{r}_2 A_{(1)}(L,\mathbf{r}_1) A_{(1)}(L,\mathbf{r}_2) \times \cos[\phi_{(1)}(L,\mathbf{r}_1) - \phi_{(1)}(L,\mathbf{r}_2)], \quad (11)$$

and the total statistically averaged power in the output current of the detector is expressed as

$$\langle S_{(1)} \rangle = \left(\frac{\eta q}{h\nu}\right)^2 2J_1^2(\delta) A_R^2 \int_\Sigma \int_\Sigma d\mathbf{r}_1 d\mathbf{r}_2 \langle u_{(1)}(L,\mathbf{r}_1) u^*_{(1)}(L,\mathbf{r}_2) \rangle, \quad (12)$$

where the $\langle\ \rangle$ denote ensemble averages and $u_{(1)}(L,\mathbf{r})$ is the wave function $u_{(1)}(L,\mathbf{r}) = A_{(1)}(L,\mathbf{r}) \exp[i\phi_{(1)}(L,\mathbf{r})]$ of the optical beam carrying the information. $\langle S_{(1)} \rangle$ is thus a function of the two-point correlation of the wave function in the plane of the detector.

In a detector where the largest source of noise is the shot noise generated by the local oscillator (reference) beam, the noise power is

$$N_{(1)} = 2qBi_{DC} = (\eta q^2/h\nu) B A_R^2 (\pi D^2/4), \quad (13)$$

where Eq. (5) has been used; B is the bandwidth of the circuit following the detector and D is the diameter of the detector. From Eqs. (12) and (13), the signal-to-noise ratio for a heterodyne detection system is

$$(S/N)_{(1)} = \langle S_{(1)} \rangle / N_{(1)} = \frac{\eta}{h\nu B} 2J_1^2(\delta) \frac{1}{(\pi D^2/4)} \int_\Sigma d\mathbf{r}_1 \int_\Sigma d\mathbf{r}_2 \times \langle u_{(1)}(L,\mathbf{r}_1) u^*_{(1)}(L,\mathbf{r}_2) \rangle. \quad (14)$$

† In the case of the video scheme, the optical bias $\phi = \pi/2$, along with an output polarizer, is used to convert the phase modulation to amplitude modulation. In the heterodyne case, the choice of ϕ is immaterial and $\phi = \pi/2$ is used for the sake of definiteness.

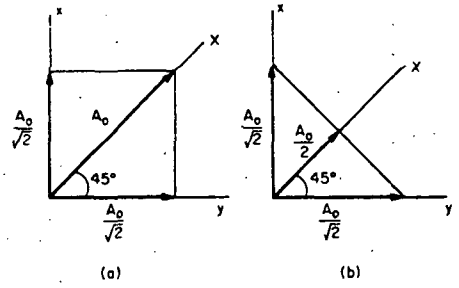

Fig. 1. Configuration of the polarizations in the case of a video communication scheme. The direction X is the direction of the laser polarization and of the parallel polarizer.

IV. Signal-to-Noise Ratio for Video Communication

In the video communication scheme, the amplitude and phase fluctuations of both the information signal and the reference signal are the same, since they follow the same atmospheric path:

$$\phi_R(L,\mathbf{r}) = \phi_S(L,\mathbf{r}) \text{ or } \Delta\phi(L,\mathbf{r}) = 0. \tag{15}$$

We call $A_{(2)}(L,\mathbf{r}) = A_S(L,\mathbf{r}) = A_R(L,\mathbf{r})$ the common amplitude. The subscript (2) refers to the part of the beam carrying the information. With the help of Eqs. (6) and (15), the total statistically averaged signal power in the output current of the detector in a video communication scheme can be expressed as

$$\langle S_{(2)}\rangle = \left(\frac{\eta q}{h\nu}\right)^2 2J_1^2(\delta) \int_\Sigma\int_\Sigma d\mathbf{r}_1 d\mathbf{r}_2 \langle I_{(2)}(L,\mathbf{r}_1) I_{(2)}(L,\mathbf{r}_2)\rangle, \tag{16}$$

where $I_{(2)}(L,\mathbf{r}) = A_{(2)}^2(L,\mathbf{r})$ is the intensity (W/m^2) of the optical wave at the plane of the detector. $\langle I_{(2)}(L,\mathbf{r}_1) I_{(2)}(L,\mathbf{r}_2)\rangle$ is the two-point correlation function of the intensity fluctuations in the plane of the detector.

In order to calculate $(S/N)_{(2)}$, we assume that the largest source of noise is the shot noise due to the optical signal itself. This condition corresponds to the ideal mode of operation of a video communication system. In a practical situation, a careful evaluation of all the parameters (areas of the transmitter and the receiver, length of propagation, wavelength, transmitted power, absorption coefficient of the atmosphere, sensitivity and noise equivalent power of the detector) is needed in order to determine whether this condition is satisfied.[2,3]

According to Eq. (5), the statistically averaged signal shot noise power is

$$\langle N_{(2)}\rangle = 2B \frac{\eta q^2}{h\nu} \int_\Sigma d\mathbf{r} \langle I_{(2)}(L,\mathbf{r})\rangle, \tag{17}$$

and $(S/N)_{(2)}$ is given by the following expression:

$$(S/N)_{(2)} = \frac{\eta}{h\nu B} J_1^2(\delta) \frac{\int_\Sigma\int_\Sigma d\mathbf{r}_1 d\mathbf{r}_2 \langle I_{(2)}(L,\mathbf{r}_1) I_{(2)}(L,\mathbf{r}_2)\rangle}{\int_\Sigma d\mathbf{r} \langle I_{(2)}(L,\mathbf{r})\rangle}. \tag{18}$$

The signal-to-noise ratio for video communication is thus a function of the correlation function of the intensity fluctuations in the plane of the detector.

V. Comparison of the Performances

We define the quantity $R = (S/N)_{(2)}/(S/N)_{(1)}$ which is a measure of the performance of a video communication system relative to that of a heterodyne communication system. According to Eqs. (14) and (18),

$$R = \frac{1}{2} \frac{(\pi D^2/4)}{\int_\Sigma d\mathbf{r} B_{u(2)}(L,\mathbf{r},\mathbf{r})} \frac{\int_\Sigma\int_\Sigma d\mathbf{r}_1 d\mathbf{r}_2 B_{I(2)}(L,\mathbf{r}_1,\mathbf{r}_2)}{\int_\Sigma\int_\Sigma d\mathbf{r}_1 d\mathbf{r}_2 B_{u(1)}(L,\mathbf{r}_1,\mathbf{r}_2)} \tag{19}$$

where $B_u(L,\mathbf{r}_1,\mathbf{r}_2) = \langle u(L,\mathbf{r}_1)\, u^*(L,\mathbf{r}_2)\rangle$ is the correlation function of the wave function and $B_I(L,\mathbf{r}_1,\mathbf{r}_2) = \langle I(L,\mathbf{r}_1) I(L,\mathbf{r}_2)\rangle$ is the correlation function of the intensity. The subscripts (1) and (2) refer to the part of the optical beam carrying the information in the heterodyne and video communication schemes, respectively.

In the absence of turbulence $R = \frac{1}{2}(A_{(2)}^2/A_{(1)}^2$, we assume that the same laser optical beam with amplitude A_0 at the plane of the detector is used as a carrier for both communication schemes. In the case of heterodyne detection, the phase of the total laser electric vector is modulated; then $A_{(1)} = A_0$. In the case of video communication, the laser polarization is chosen to have equal components along two perpendicular directions. One polarization component is used as the information signal, the other as the transmitted reference signal; then only the amplitude $A_0/(2)^{\frac{1}{2}}$ is modulated [see Fig. 1(a)]. A polarizer is used at the detector to mix the information and the reference signals. If the polarizer is parallel to the laser polarization, the amplitude of the modulated optical signal in this direction is $A_{(2)} = [A_0/(2)^{\frac{1}{2}}][1/(2)^{\frac{1}{2}}] = (A_0/2)$ [see Fig. 1(b)]. Then in the absence of turbulence, $R = \frac{1}{8}$; i.e., the S/N for the superheterodyne system is larger by a factor of eight than that of a video system.

In order to calculate R in the presence of atmospheric turbulence, we use the results of a theoretical analysis of optical wave propagation through a random medium which is reported elsewhere.[4]

Assuming that the refractive index fluctuations are a homogeneous[5] gaussian random process, the wave function correlation function is expressed in terms of the

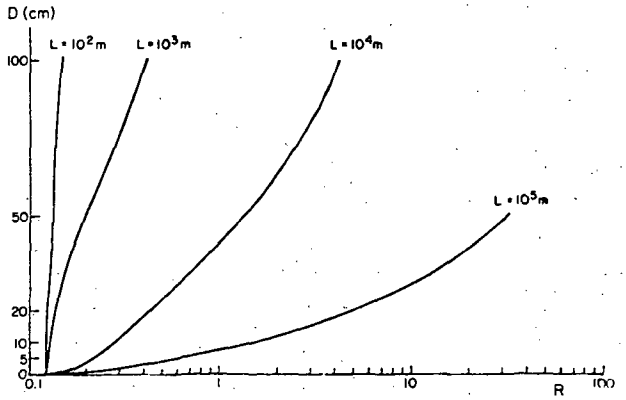

Fig. 2. R vs D for a weak turbulence $C_n = 10^{-8}\,m^{-1/3}$ at $\lambda = 1\mu$.

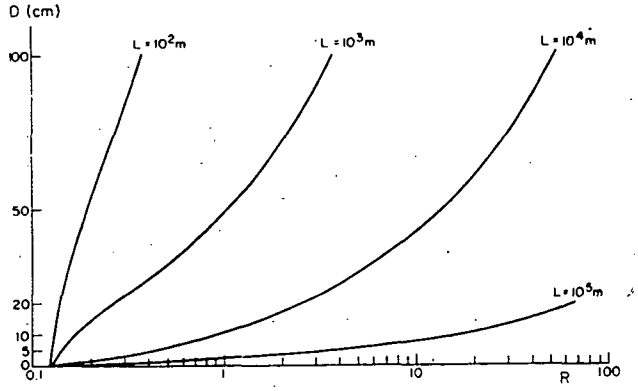

Fig. 3. R vs D for an intermediate turbulence $C_n = 3 \times 10^{-8}$ $m^{-1/3}$ at $\lambda = 1\ \mu$.

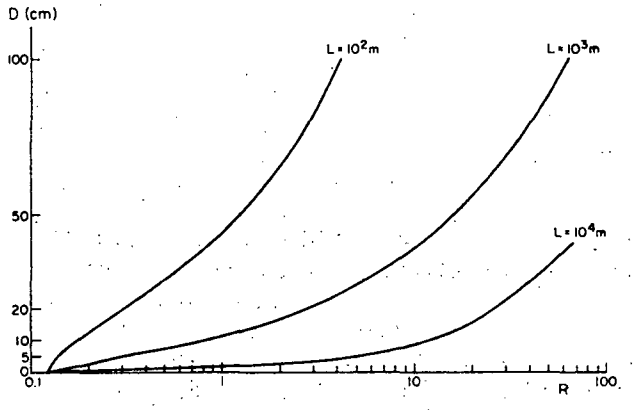

Fig. 4. R vs D for a strong turbulence $C_n = 10^{-7}$ $m^{-1/3}$ at $\lambda = 1\ \mu$.

correlation function of the index of refraction $B_n(\alpha)$ in the following way:

$$B_{u(1)}(L,\mathbf{r}_1,\mathbf{r}_2) = B_{u(1)}(L,\rho) = A_{(1)}^2 \exp\left[-2k^2L \times \left(\int_0^{L_0} B_n(\alpha)d\alpha - \int_0^{(L_0^2-\rho^2)^{\frac{1}{2}}} B_n(\alpha^2+\rho^2)^{\frac{1}{2}}\right) d\alpha\right]; \quad (20)$$

L_0 is the outer scale of the turbulence, and $\rho = |\mathbf{r}_1 - \mathbf{r}_2|$. No theory to date has proven successful in finding an exact analytical expression for the intensity correlation function $B_I(L,\mathbf{r}_1,\mathbf{r}_2)$ which is applicable beyond the limits of validity of the Rytov approximation[4] (long propagation paths, strong turbulences). For the purpose of a numerical application, we choose an expression for B_I which fits the recent experimental data of Gracheva for small and large $\sigma_1(L,0)$[4,6] as defined by Eq. (22).

$$B_{I(2)}(L,\mathbf{r}_1,\mathbf{r}_2) = B_{I(2)}(L,\rho) = A_{(2)}^4\left[2 - \frac{1}{1+\sigma_1^2(L,\rho)}\right], \quad (21)$$

where

$$\sigma_1^2(L,\rho) = 8\pi^2k^2L\int_0 J_0(K\rho) \times \left(1 - \frac{k}{K^2L}\sin\frac{K^2L}{k}\right)\phi_n(K)K\,dK; \quad (22)$$

$\phi_n(K)$ is the three-dimensional Fourier transform of the refractive index correlation function. In deriving Eq. (21), we have used the assumption which is supported by experimental evidence,[6-9] that the amplitude of an optical wave propagating through atmospheric turbulence has a log-normal probability distribution function.

With the help of Eqs. (20)–(22) and (19), we have performed a numerical calculation of R using the Kolmogoroff-Obukhov model of turbulence:[10]

$$\phi_n(K) = \begin{cases} 0.033C_n^2K^{-\frac{11}{3}} & \text{for } K < K_m = 5.48/l_0 \\ 0 & \text{for } K > K_m \end{cases}$$

and have expressed R in terms of the diameter of the detector D, the wavelength λ, the communication distance L, and the turbulence strength as represented by the structure constant C_n. The results are presented in Figs. 2–6.

In Fig. 2, the performance criterion R is plotted as a function of the detector diameter D for a communication link at 1 μ propagating up to a distance of 100 km through a weak turbulence ($C_n = 10^{-8}\ m^{-\frac{1}{3}}$). R vs D plots for the same communication system in intermedi-

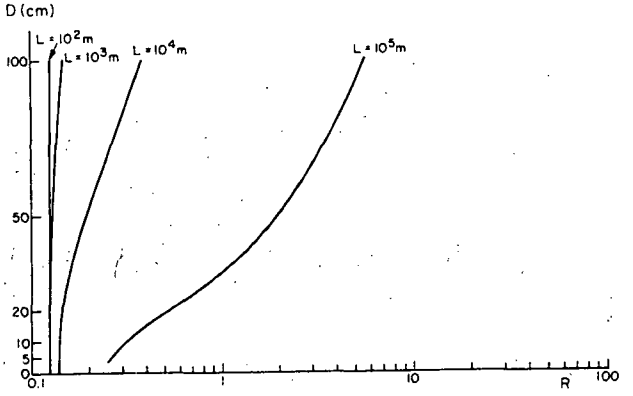

Fig. 5. R vs D for an intermediate turbulence $C_n = 3 \times 10^{-8}$ $m^{-1/3}$ at $\lambda = 10\ \mu$.

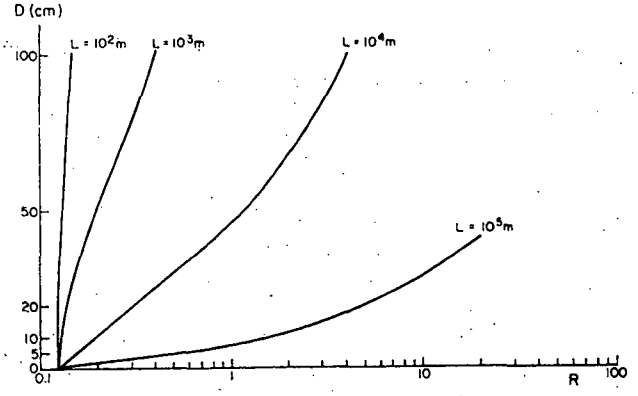

Fig. 6. R vs D for a strong turbulence $C_n = 10^{-7}$ $m^{-1/3}$ at $\lambda = 10\ \mu$.

ate ($C_n = 3 \times 10^{-8}\ m^{-\frac{1}{3}}$) and strong ($C_n = 10^{-7}\ m^{-\frac{1}{3}}$) turbulences are given in Figs. 3 and 4, respectively. It is found that R is an increasing function of the detector diameter of the propagation distance and of the turbulence strength. For a propagation distance of 10 km at 1 μ, as an example, the video communication scheme has a larger signal-to-noise ratio than the heterodyne detection scheme ($R > 1$) when $D > 40$ cm, $D > 10$ cm, and $D > 2$ cm in weak, intermediate, and strong turbulences, respectively. A video communication scheme would perform even better for longer propagation distances, stronger turbulences, and shorter wavelengths. This is due to the cancellation of the phase fluctuations between the reference and the signal parts of the beam.

In Figs. 5 and 6, we have represented R vs D for a communication link at a wavelength of 10 μ under intermediate and strong turbulences. In this case, R increases more slowly with D, L, and C_n than for $\lambda = 1\ \mu$. It is only for long communication lengths under strong turbulence ($L = 100$ km, $C_n = 10^{-7}\ m^{-\frac{1}{3}}$) that a video system would be preferable to a superheterodyne detection system. This is due to the fact that the correlation function $B_u(L,\rho)$, as expressed by Eq. (20), varies as e^{-1/λ^2} and therefore decreases more slowly with a longer wavelength. The phase fluctuations are less important at 10 μ than at 1 μ.

VI. Conclusion

We have calculated the (S/N) for (1) a superheterodyne and (2) a video optical communication system, involving propagation through the atmospheric turbulence. It has been found that for long propagation distances in a weak turbulence, or for a propagation distance of a few kilometers in an intermediate turbulence, scheme (2) operating at 1 μ has a larger signal-to-noise ratio than scheme (1). However, a heterodyne system operating at 10 μ is less sensitive to the random phase fluctuations introduced by the atmosphere than a system at 1 μ.

As far as the effects of atmospheric turbulence are concerned, at 1 μ and smaller wavelengths a video system is preferable to a superheterodyne system, while at 10 μ and longer wavelengths, the inverse is true. These theoretical results are in agreement with some recent experimental results.[11]

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Analysis of Mode Locking and Ultrashort Laser Pulses With a Nonlinear Refractive Index

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Abstract—A new method for locking the longitudinal modes of a laser resonator and generating ultrashort pulses of light is described. The cavity modes are coupled together when a medium with a refractive index nonlinearity is placed in the cavity.

A theoretical study is presented which analyzes the mode structure of a laser resonator containing a cell filled with an anisotropic molecular liquid. It is found that under certain conditions the energy exchange between the modes gives rise to a mode-locked spectrum and to the attendant generation of ultrashort pulses of light ($\sim 10^{-11}$ second for a ruby laser, 10^{-12} second for a Nd^{3+}: glass laser).

Introduction

THE OUTPUT electric field of a laser is equal to the sum of the electric fields of the individual modes of the cavity that are amplified by the laser medium, i.e., whose frequencies lie within the gain linewidth $\Delta\nu_G$ of the amplifying transition.

In the normal mode of oscillation of a laser (no perturbation inside the cavity), the phases of the modes are random and uncorrelated, and the output intensity is fluctuating randomly in time around its mean value $N\bar{I}$, where N is the number of oscillating cavity modes and $\bar{I}$ is the average mode intensity.

It has been shown [1]–[3] that if the losses of the laser cavity are modulated at a frequency equal to the intermode spacing frequency $c/2L$ (L is the length of the cavity), mode locking results and the output of the laser consists of a continuous train of pulses that have the following properties.

1) The pulsewidth is equal to the reciprocal of the gain linewidth $1/\Delta\nu_G$.
2) The pulses are separated in time by the double transit time of the light inside the cavity $2L/c$.
3) The peak power is equal to N times the average power of the laser where N is the number of coupled modes.

Using internal modulators, ultrashort pulses have been obtained in continuous-wave gas lasers [2] (with a width of 2.5×10^{-9} second) and solid-state lasers [3] (8×10^{-11} second) with a pulsewidth approaching the theoretical value $1/\Delta\nu_G$. Internal modulators have also been used to generate ultrashort pulses in pulsed solid-state lasers where the duration of the pulsing ($\sim 1\ \mu s$) is larger than the modulating period [4], [5]. The observed pulsewidths were 2×10^{-9} second for ruby and 0.5×10^{-9} second for Nd:glass, while the theoretical values are, respectively, 10^{-11} second and 4×10^{-13} second, indicating that the whole linewidth is either not fully mode locked, or that the frequency is swept.

An increase in the output power of solid-state lasers has been obtained by the technique of Q switching [6]. The output of a non-mode-locked Q-switched solid-state laser consists typically of a pulse of $10\text{–}50 \times 10^{-9}$ second with a peak power of up to a few hundred megawatts. In these lasers, mode locking has been obtained by inserting a saturable absorber inside the cavity [7], [8]. A saturable absorber is an element whose optical transmission is an increasing function of the intensity of the incident beam. Pulses whose duration is $\sim 10^{-11}$ second as short in ruby lasers and $\sim 10^{-12}$ second in Nd:glass lasers with peak intensities in excess of 10^9 watts have been observed by using this technique.

In this paper we analyze a new method [9], [10] for generating high-intensity picosecond pulses in Q-switched solid-state lasers.

We show theoretically that the introduction of a refractive index nonlinearity inside a laser resonator gives rise to a mode-locked spectrum, characteristic of the ultrashort pulse mode of oscillation. The nonlinearities we consider are provided by liquids consisting of anisotropic uniaxial molecules. These molecules, of which nitrobenzene and CS_2 are two representative examples, have different polarizabilities along their axis of symmetry and along any other axis perpendicular to it. We call these polarizabilities $\alpha_\parallel$ and $\alpha_\perp$ respectively. A linearly polarized electric field applied to such a liquid induces a nonlinear polarization in the medium that is proportional to the difference $(\alpha_\parallel - \alpha_\perp)$ and to the cube of the electric field and therefore produces a change in the dielectric constant of the medium proportional to the square of the electric field. When a liquid with anisotropic molecules is placed inside a laser resonator where the optical electric fields are large enough to produce an appreciable change of the dielectric constant, it couples the longitudinal modes of the resonator together in the following way. Let us assume that three modes of the resonator (0), (+1), and (−1) oscillate with their respective frequencies ω_0, $\omega_0 + \Omega$, $\omega_0 - \Omega$. Ω is the radian intermode frequency $\Omega = \pi c/L$.

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The two modes (0) and (+1), for example, induce a change in the dielectric constant of the liquid $\Delta\epsilon \propto E_0E_1$, where E_0 and E_1 are the electric fields of the two modes. Thus $\Delta\epsilon$ has a component oscillating at the frequency $(\omega_0 + \Omega) - \omega_0 = \Omega$. The mode (-1), incident upon the liquid, "sees" a modulation of the dielectric constant at frequency Ω, which causes the generation of sideband frequencies at $(\omega_0 - \Omega) + m\Omega$, $m = \pm 1, \pm 2, \cdots$. As an example, the sideband at $(\omega_0 - \Omega) + \Omega = \omega_0$ coincides in frequency with mode (0). The effect of the nonlinearity $\Delta\epsilon \propto E^2$ is thus seen to be one of coupling modes together, i.e., introducing unique relationships between the amplitudes and the phases of the modes.

We present below a detailed theoretical analysis of the intuitive picture described in the last paragraph. The results of an experimental investigation have been reported elsewhere [9], [10].

The third-order nonlinear polarization induced in the laser cavity by the anisotropic molecular liquid is expressed in terms of the parameters of the liquid as a triple summation over the cavity modes. Maxwell's equations with the nonlinear polarization acting as a driving term are then used to find a differential equation obeyed by the modes' amplitudes. A steady-state self-consistent solution is found. This solution applies to practical experimental situations only if sufficient energy exchange takes place between the modes in a time shorter than the duration of a Q-switched pulse. The energy exchange time constant T_0 is calculated in terms of the parameters of the liquid and of a given laser system. We find that under reasonable experimental situations T_0 can be made short enough so that mode locking can take place during the duration $\sim$ 10–30 $\times$ 10^{-9} second of a typical Q-switched laser pulse.

Nonlinear Polarization Induced in the Anisotropic Molecular Liquid

The dipole moment of an anisotropic molecule induced by a linearly ($||z$) polarized electric field E along its own direction of polarization is

$$\mu_z = E(\alpha_\parallel - \alpha_\perp)\cos^2\theta + \alpha_\perp E \tag{1}$$

where θ is the angle between the direction of the electric field and the axis of symmetry of the molecule (see Fig. 1).

The average induced dipole moment of one anisotropic molecule is found by replacing $\cos^2\theta$ by its statistical average $\langle\cos^2\theta\rangle$ taken over the ensemble of molecules

$$\langle\mu_z\rangle = E(\alpha_\parallel - \alpha_\perp)\langle\cos^2\theta\rangle + \alpha_\perp E. \tag{2}$$

In the absence of any electric field, all the orientations of the molecular axis of symmetry are equally probable and $\langle\cos^2\theta\rangle = \frac{1}{3}$. In the presence of a strong electric field, the molecules tend to align with their axes parallel to the field direction and $\langle\cos^2\theta\rangle$ is different from $\frac{1}{3}$. We write

$$\langle\cos^2\theta\rangle = s + \tfrac{1}{3}. \tag{3}$$

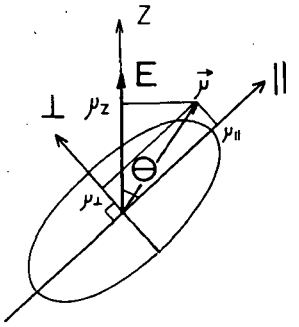

Fig. 1. Orientation of an anisotropic molecule in an electric field.

The quantity s determines the average deviation of the orientation of anisotropic molecules from a purely random orientation. s is the first diagonal element of the anisotropy tensor [11]. From (2) and (3) we obtain

$$\langle\mu_z\rangle = E(\alpha_\parallel - \alpha_\perp)s + \frac{E}{3}(\alpha_\parallel + 2\alpha_\perp). \tag{4}$$

The anisotropy tensor element s can be shown to obey the following differential equation [11]:

$$\frac{ds}{dt} + \frac{s}{\tau} = \frac{2\lambda}{3}E^2 \tag{5}$$

where E is the linearly polarized electric field. τ is the time constant with which the molecules regain their random orientation after the electric field has been turned off. It is often called the Debye relaxation time [12] or the orientational relaxation time. λ can be shown by a simple thermal equilibrium argument [11] to be given by

$$\lambda = \frac{1}{15}\frac{(\alpha_\parallel - \alpha_\perp)}{kT\tau}.$$

We look for a solution of the differential equation (5) in the case where the electric field is the sum of the electric fields of the modes of a laser resonator. For this purpose we find it advantageous to introduce, first, the normal mode formalism for describing the optical resonator field.

Normal Mode Formalism

In order to describe the mode spectrum of the laser resonator, we introduce a set of orthonormal electric and magnetic vector functions $\boldsymbol{E}_n(r)$ and $\boldsymbol{H}_n(r)$ as defined by Slater [13], [14]. They are related by the following relationships:

$$k_n\boldsymbol{E}_n(r) = \nabla \times \boldsymbol{H}_n(r),\ k_n\boldsymbol{H}_n(r) = \nabla \times \boldsymbol{E}_n(r) \tag{6}$$

$$\nabla\cdot\boldsymbol{E}_n(r) = \nabla\cdot\boldsymbol{H}_n(r) = 0 \tag{7}$$

where k_n is a constant and n is the index mode number. According to (6) and (7), they satisfy the following differential equations:

$$(\nabla^2 + k_n^2)\boldsymbol{E}_n(r) = 0, \qquad (\nabla^2 + k_n^2)\boldsymbol{H}_n(r) = 0 \tag{8}$$

and they are defined so as to obey the normalization conditions

$$\int_V \boldsymbol{E}_n(r)\cdot\boldsymbol{E}_m(r)\,dr = \delta_{nm},\quad \int_V \boldsymbol{H}_n(r)\cdot\boldsymbol{H}_m(r)\,dr = \delta_{nm}. \tag{9}$$

The above integrations are performed over the total volume of the cavity.

We express the total electric field $E(r, t)$ (here we go over to a scalar notation appropriate for TEM-like optical resonator modes), and the total magnetic field $H(r, t)$ inside the cavity as

$$E(r, t) = -\sum_n \frac{1}{\sqrt{\epsilon_0}} p_n(t)E_n(r) \tag{10}$$

$$H(r, t) = \sum_n \frac{1}{\sqrt{\mu_0}} \omega_n q_n(t)H_n(r) \tag{11}$$

where ω_n is defined by

$$k_n = \omega_n \sqrt{\mu_0\epsilon_0} \tag{12}$$

and where $E_n(r)$ and $H_n(r)$ are the electric and magnetic scalar functions defined above. The summation is performed over the total number of modes of the cavity. ϵ_0 and μ_0 are the dielectric constant and the permeability of the medium filling the cavity. $p_n(t)$ and $q_n(t)$ are unknown functions of time describing the amplitudes and phases of the individual longitudinal cavity modes.

Expression of the Anisotropy Tensor Element s in a Laser Cavity

With the help of the formalism presented above we rewrite the differential equation (5) as follows:

$$\frac{ds}{dt} + \frac{s}{\tau} = \frac{2\lambda}{3} \sum_a \sum_b \frac{1}{\epsilon_0} p_a(t)p_b(t)E_a(r)E_b(r). \tag{13}$$

We assume a solution for $p_n(t)$ in the form

$$p_n(t) = i\sqrt{\frac{\omega_n}{2}}(D_n^*(t)e^{i\omega_n t} - D_n(t)e^{-i\omega_n t}) \tag{14}$$

where ω_n is the optical frequency of the oscillating mode n and $D_n(t)$ and $D_n^*(t)$ are slowly varying functions of time compared to $e^{i\omega_n t}$. $D_n^*(t)$ is the complex conjugate of $D_n(t)$. The phase and amplitude evolution of mode n is thus contained completely in $D_n^*(t)$. $p_a(t)$ and $p_b(t)$ are replaced in (13) by (14). The product $p_a(t)p_b(t)$ is made up of two frequency components, one at frequency $\omega_a + \omega_b$ and the other at frequency $\omega_a - \omega_b$. Both ω_a and ω_b are optical frequencies $\sim 10^{15}$ rad/s, so $\omega_a + \omega_b$ is an optical frequency. Since the orientational relaxation time of the liquid is of the order of 10^{-10} to 10^{-12} second, the molecules cannot respond to fields at optical frequencies. The only term to which the molecules can respond is the term at frequency $\omega_a - \omega_b$, which ranges from $\omega_a - \omega_b = 0$ to $\omega_a - \omega_b \sim 2\pi\,\Delta\nu_G$ where $\Delta\nu_G$ is the gain linewidth of the laser medium. Then

$$\frac{ds}{dt} + \frac{s}{\tau} = \frac{2\lambda}{3} \sum_a \sum_b E_a(r)E_b(r) \frac{\sqrt{\omega_a\omega_b}}{2\epsilon_0} \cdot(D_a^*(t)D_b(t) \exp [i(\omega_a - \omega_b)t] + \text{c.c.}). \tag{15}$$

We look for a solution for s in the following form:

$$s = \sum_a \sum_b s_{ab}^*(t) \exp [i(\omega_a - \omega_b)t] + \text{c.c.} \tag{16}$$

where $s_{ab}^*(t)$ is a slowly varying function of time compared to $\exp [i(\omega_a - \omega_b)t]$ when $a \neq b$. Therefore for $a \neq b$ we neglect $ds_{ab}^*(t)/dt$ with respect to $(\omega_a - \omega_b)s_{ab}^*(t)$. By substituting (16) for s in (15) we find the following expression for $s_{ab}^*(t)$:

$$s_{ab}^*(t) = \frac{\lambda\tau}{3\epsilon_0} E_a(r)E_b(r) \sqrt{\omega_a\omega_b} \frac{D_a^*(t)D_b(t)}{1 + i(\omega_a - \omega_b)\tau}. \tag{17}$$

Using this last result in (16) leads to

$$s = \frac{\lambda\tau}{3\epsilon_0} \sum_a \sum_b \sqrt{\omega_a\omega_b} \frac{D_a^*(t)D_b(t)}{1 + i(\omega_a - \omega_b)\tau} \cdot \exp [i(\omega_a - \omega_b)t]E_a(r)E_b(r) + \text{c.c.} \tag{18}$$

We thus find that the average molecular orientation oscillates at integral multiples of the intermode frequency (since $\omega_a - \omega_b = m\pi c/L$, $m = \pm 1, \pm 2, \cdots$). The amplitude of each frequency component is proportional to the product of two laser fields $D_a^*\,D_b$ and lags by $\phi_{ab} = \tan^{-1}[(\omega_a - \omega_b)\tau]$.

Total Nonlinear Polarization

The total average polarization P induced in the anisotropic molecular liquid per unit volume is $P = N_0\langle\mu_z\rangle$ where N_0 is the number of molecules per unit volume and $\langle\mu_z\rangle$ is given by (4). In MKS units the displacement vector $\boldsymbol{D}$ is expressed as $\boldsymbol{D} = \epsilon_0\boldsymbol{E} + \boldsymbol{P}$, which can be written as $\boldsymbol{D} = \epsilon_L\boldsymbol{E} + \boldsymbol{P}_{\text{NL}}$ where ϵ_L is the dielectric constant of the liquid and $\boldsymbol{P}_{\text{NL}}$ is the total nonlinear polarization induced in the liquid. According to (4) the scalar polarization field is

$$P_{\text{NL}} = N_0(\alpha_\parallel - \alpha_\perp)sE. \tag{19}$$

P_{NL} can be expressed in terms of the cavity mode wave functions $D_n(t)E_n(r)$ with the help of (10), (14), and (18):

$$P_{NL} = -3i\epsilon_2 \sum_a \sum_b \sum_c \sqrt{\frac{\omega_a\omega_b\omega_c}{8\epsilon_0^3}} E_a(r)E_b(r)E_c(r) \times \frac{D_a^*(t)D_b(t)D_c^*(t)}{1 + i(\omega_a - \omega_b)\tau} \exp [i(\omega_a - \omega_b + \omega_c)t] + \text{c.c.} \tag{20}$$

where ϵ_2 is the optical Kerr constant expressed in MKS units

$$\epsilon_2 = \frac{4}{135} N_0 \frac{(\alpha_\parallel - \alpha_\perp)^2}{kT}. \tag{21}$$

Solution of Maxwell Equations With a Third-Order Nonlinear Polarization

In order to find an expression for the electric field in a laser resonator in the presence of an anisotropic molecular liquid, a nonlinear polarization driving term is included in the Maxwell equations:

$$\nabla \times \boldsymbol{E} = -\mu_0 \frac{\partial \boldsymbol{H}}{\partial t} \tag{22}$$

$$\nabla \times \boldsymbol{H} = \sigma \boldsymbol{E} + \frac{\partial}{\partial t}(\epsilon_0 \boldsymbol{E} + \boldsymbol{P}_{\mathrm{NL}}) \tag{23}$$

where σ is the electrical conductivity of the medium. For simplicity, we have assumed that the linear dielectric constant of the liquid is equal to 1.

By this procedure we first find a differential equation obeyed by the mode amplitudes $D_n^*(t)$.

Differential Equation for the Mode Amplitude $D_n(t)$

We replace the electric field and magnetic field by their expressions (10) and (11) in the first of the Maxwell equations (22). Both sides of the equation are then multiplied by $\boldsymbol{H}_m(r)$ and integrated over the volume of the cavity. With the help of (9) and (12), we find

$$p_n(t) = q_n'(t). \tag{24}$$

The first Maxwell equation (22) has provided a relationship between $p_n(t)$ and $q_n(t)$. According to (24) and (14) we can now express $q_n(t)$ and then the magnetic field $H(r, t)$ in terms of the complex amplitude $D_n^*(t)$ of the nth cavity mode:

$$q_n(t) = \frac{1}{\sqrt{2\omega_n}}(D_n^*(t)e^{i\omega_n t} + D_n(t)e^{-i\omega_n t}). \tag{25}$$

It has been assumed in deriving expression (25) that $D_n(t)$ is a very slowly varying function of time compared to $e^{i\omega_n t}$, i.e.,

$$\left|\frac{dD_n(t)}{dt}\right| \ll \omega_n |D_n(t)| .$$

A second relationship between $p_n(t)$ and $q_n(t)$ is obtained by using (23) in which E, H, and P_{NL} are replaced by their expressions (10), (11), and (20). Both sides of the equation are multiplied by $E_m(r)$ and integrated over the volume of the cavity. The relationships (6) and (9) are used to find the differential equation

$$\omega_n^2 q_n(t) + \frac{\sigma}{\epsilon_0} p_n(t) + p_n'(t) = \frac{3\epsilon_2}{2\sqrt{2}\,\epsilon_0^2} \sum_a \sum_b \sum_c \sqrt{\omega_a\omega_b\omega_c}\, S_{nabc} \times (\omega_a - \omega_b + \omega_c) \left(\frac{D_a^*(t) D_b(t) D_c^*(t)}{1 + i(\omega_a - \omega_b)\tau} \cdot \exp\left[i(\omega_a - \omega_b + \omega_c)t\right] + \text{c.c.}\right) \tag{26}$$

where S_{nabc} as defined by the relationship

$$S_{nabc} = \int_{\substack{\text{volume of the} \\ \text{anisotropic liquid}}} E_n(r)E_a(r)E_b(r)E_c(r)\, dV \tag{27}$$

is a factor that depends upon the dimensions and the position of the anisotropic liquid inside the laser cavity.

The equation $p_n(t) = q_n'(t)$ has allowed us to express $q_n(t)$ in terms of $D_n^*(t)$ and $D_n(t)$. We then replace $p_n(t)$ and $q_n(t)$ by their expressions (14) and (25) and find a differential equation for $D_n^*(t)$ instead of $p_n(t)$:

$$\frac{dD_n^*(t)}{dt} + \frac{\sigma}{\epsilon_0} D_n^*(t) = -\frac{3i\epsilon_2}{2\epsilon_0^2} \sqrt{\omega_n} \sum_{a,b,c} \sqrt{\omega_a\omega_b\omega_c}\, S_{nabc} \cdot \frac{D_a^*(t) D_b(t) D_c^*(t)}{1 + i(\omega_a - \omega_b)\tau} \tag{28}$$

where only the modes a, b, and c such that

$$\omega_a - \omega_b + \omega_c = \omega_n \tag{29}$$

can provide synchronous driving of the oscillation at ω_n.

The frequency of the nth mode is defined as

$$\omega_n = n\Omega \tag{30}$$

where $\Omega = \pi c/L$ and n is a very large number equal to the number of half-wavelengths contained in the length of the resonator. With the definition (30), the condition (29) becomes $a - b + c = n$. The triple summation over a,b,c in (28) is replaced by a double summation over m and p such that $a = n + m$, $b = n + m + p$, and $c = n + p$. The above substitution leads to the differential equation

$$\frac{dD_n^*}{dt} = -\frac{3i\epsilon_2\omega_n}{2\epsilon_0^2} \sum_p \frac{D_{n+p}^*}{(1 - ip\Omega\tau)} \cdot \sum_m \omega_{n+m} D_{n+m+p} D_{n+m}^* S_{n,m,p} \tag{31}$$

where we have assumed that the gain provided by the laser medium is equal to the losses of the cavity (reflection losses at the mirror and diffraction losses), so that the effective conductivity σ is taken as zero.

The nth cavity mode has the spatial dependence:

$$E_n(r) = \sqrt{\frac{2}{AL}} \sin\frac{n\pi x}{L} \tag{32}$$

where A is the cross section of the beam. The factor $S_{n,m,p}$ is calculated explicitly, using (27), when a liquid cell of length $2l$ is inserted in an optical cavity of length L at a distance L_0 from a mirror that is taken as the origin of the coordinates (see Fig. 2):

$$S_{n,m,p} = \frac{1}{V}\left(\frac{l}{L} + \frac{1}{2p\pi}\sin\frac{2\pi pl}{L}\cos\frac{2\pi pL_0}{L} + \frac{1}{2m\pi}\sin\frac{2m\pi l}{L}\cos\frac{2\pi mL_0}{L}\right). \tag{33}$$

Solution of the Differential Equation for $D_n^*(t)$

We find a solution of the differential equation (31) in the case when the liquid cell fills half of the laser cavity $L_0 = l = L/4$. According to (31) and (33),

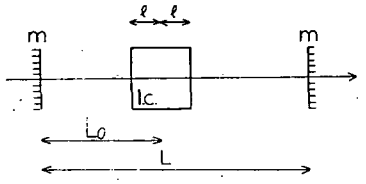

Fig. 2. Position of the liquid cell (lc) inside the cavity formed by the two mirrors (m).

$$\frac{dD_n^*}{dt} = -\frac{3i\epsilon_2\omega_n}{2\epsilon_0^2 V}\frac{l}{L}\left(\sum_p \frac{D_{n+p}^*}{(1-ip\Omega\tau)}\sum_m \omega_{n+m} D_{n+m+p} D_{n+m}^* + D_n^* \sum_m \omega_{n+m} D_{n+m} D_{n+m}^* + D_n^* \sum_p \omega_{n+p}\frac{D_{n+p} D_{n+p}^*}{(1-ip\Omega\tau)}\right). \quad (34)$$

In order to specify the values of p and m over which the summations in (34) are performed, we discuss in some detail the mechanism of power exchange between modes. The above equation describes the coupling between four modes labeled n, $n+p$, $n+m+p$, and $n+m$. The susceptibility for this coupling is $\chi = \epsilon_2/(1-ip\Omega\tau)$. We can write the susceptibility χ as a real part χ' plus an imaginary part $+i\chi''$. The transfer of energy between the modes n and $n+p$ is proportional to $\chi'' = \epsilon_2(p\Omega\tau)/[1+(p\Omega\tau)^2]$.

The rate at which energy is transferred into one mode via another mode separated in frequency by $p\Omega$ is thus proportional to the function $f(p/m_0) = (p/m_0)/[1+(p/m_0)^2]$ where $m_0 = 1/\Omega\tau$. This function has a maximum for $p = m_0$ and decreases to zero for $p > m_0$. We neglect the interaction of the nth mode with the $n+p$th mode when $p > 2m_0$; the summation over p is then limited to $-2m_0$ and $2m_0$. We label N_1 and N_2 the lowest and highest frequency modes of the gain linewidth; the total number of modes is $N = N_2 - N_1$. We look for a solution of (34) in the form

$$D_n^*(t) = \beta(t)e^{in\Phi(t)}, \qquad D_n(t) = \beta(t)e^{-in\Phi(t)} \quad (35)$$

where $\beta(t)$ and $\Phi(t)$ are real functions of time. In this assumed form the cavity modes have the same amplitude $\beta(t)$ and the phase difference between two adjacent modes is the same for all the modes and is equal to $\Phi(t)$. We substitute for D_n^* and D_n in (34) their expressions (35), and perform the summations with the limits described above to find

$$\left(\frac{d\beta(t)}{dt} + in\Phi'(t)\beta(t)\right)e^{in\Phi(t)} = -\frac{3i\epsilon_2\omega_n}{2\epsilon_0^2 V}\frac{l}{L}e^{in\Phi(t)} \times \left(\beta\sum_{p=-2m_0}^{2m_0}\left(\frac{1}{1-ip\Omega\tau}\right)\sum_m \omega_{n+m}\beta^2 + \beta\mathcal{E}_T + \beta\sum_{p=-2m_0}^{2m_0}\frac{\omega_{m+p}\beta^2}{(1-ip\Omega\tau)}\right). \quad (36)$$

In (36) we have used the fact that the total electromagnetic energy $\mathcal{E}_T$ stored in the cavity can be expressed according to (9)–(11), (14), (25), and (35) as

$$\mathcal{E}_T = \frac{1}{2}\int_{\substack{\text{volume of}\\ \text{the cavity}}}(E^2+H^2)\,dv = \sum_m \omega_{n+m} D_{n+m} D_{n+m}^* = \sum_m \omega_{n+m}\beta^2.$$

The last term in the parentheses on the right-hand side of (36) is roughly equal to $1/N$ times the sum of the first two terms. Since N is very large, it can be neglected and we write

$$\frac{d\beta(t)}{dt} + in\Phi'(t)\beta(t) = -\frac{3i\epsilon_2\omega_n}{\epsilon_0^2 V}\frac{l}{L}\mathcal{E}_T\beta\sum_{p=0}^{2m_0}\left(\frac{1}{1+p^2\Omega^2\tau^2}\right). \quad (37)$$

The right-hand side of (37) is purely imaginary. Therefore $d\beta(t)/dt = 0$ and $\beta(t) \equiv \beta =$ constant. The amplitudes of the cavity modes are constant in time and a solution of (37) is the following:

$$D_n^*(t)e^{i\omega_n t} = \beta e^{i\omega_n t}\left(1 - \frac{3\epsilon_2}{\epsilon_0^2 V}\frac{l}{L}\mathcal{E}_T\sum_{p=0}^{2m_0}\left(\frac{1}{1+p^2\Omega^2\tau^2}\right)\right)t. \quad (38)$$

Therefore the presence of an anisotropic molecular liquid inside a laser resonator gives rise to a mode-locked spectrum of equal amplitudes and zero phases. The term inside the parenthesis in the exponential represents frequency pulling. It is easy to show that a solution in the form of (38) results even for an arbitrary placement of the cell within the resonator. The time envelope $E(t) \propto (\sum_n D_n^*(t)\exp(i\omega_n t) + \text{c.c.})$, where $D_n^*(t)$ is given by (38), consists of a train of ultrashort laser pulses of very high intensity. These pulses are separated in time by the double transit time of the light inside the cavity $2L/c$ and approach a duration $T \sim (\Delta\nu_G)^{-1}$ where $\Delta\nu_G$ is the gain linewidth of the amplifying transition [1]. The resonance frequency of the nth mode is slightly pulled from its initial value ω_n by an amount proportional to the stored energy $\mathcal{E}_T$.

Energy Circulation Time Constant

As a measure of the strength of the mode coupling due to the refractive index nonlinearity, we define a circulation time T_0 as the exponential time constant for the circulation of the energy in one mode due to its interactions with all others, once phase locking is achieved. If $\mathcal{E}_n$ is the energy of the nth mode, then

$$\frac{1}{T_0} = \frac{1}{\mathcal{E}_n}\frac{d\mathcal{E}_n}{dt}. \quad (39)$$

Calculation of T_0

The energy of the nth mode is $\mathcal{E}_n = \omega_n D_n^* D_n = \omega_n\beta^2$ where D_n^* and D_n have been replaced by their expressions (35). Therefore, according to (39)

$$\frac{1}{T_0} = 2\frac{\beta'}{\beta}. \tag{40}$$

We have found earlier that $\beta' = 0$ and $\beta =$ constant. This is true because in the steady state as expressed by (38) and, assuming a very large number of modes, as much energy flows into the nth mode via the higher frequency modes as flows out of it via the lower frequency modes. We want to calculate the rate at which energy is flowing *into* the nth mode. Therefore, in order to find β', we keep in the summation of p of expression (36), only the terms where p is positive.

Only the real part of the right-hand side of (36) gives rise to power exchange between the modes. Equating $\beta'(t)$ to the real part of the right-hand side of (36) and using the definition (40) yields

$$\frac{1}{T_0} = \frac{3\epsilon_2\omega_n}{\epsilon_0^2}\frac{l}{L}\frac{\mathcal{E}_T}{V}\sum_{p=1}^{2m_0}\frac{p\Omega\tau}{1+(p\Omega\tau)^2}.$$

The term

$$\sum_{p=1}^{2m_0}\frac{p\Omega\tau}{1+(p\Omega\tau)^2},$$

accounting for the number of modes interacting with any one mode, is approximately equal to $1/\Omega\tau$. Therefore

$$\frac{1}{T_0} = \frac{3}{2}\frac{\epsilon_2\omega_n}{\epsilon_0^2}\frac{l}{L}\frac{\mathcal{E}_T}{V}\frac{1}{\Omega\tau}. \tag{41}$$

The rate at which energy is exchanged between modes is proportional to the electromagnetic energy stored per unit volume in the cavity $\mathcal{E}_T/V$, to the ratio of the length of the liquid cell to the length of the cavity l/L, to the optical Kerr constant ϵ_2, and inversely proportional to $\Omega\tau$. ϵ_2 in (41) is given in MKS units. Using the Kerr constant B_0[15] in esu units, $1/T_0$ is expressed in the following way:

$$\frac{1}{T_0} = 12\pi Cn\frac{\lambda_A}{\lambda}B_0\frac{l}{L}\frac{\mathcal{E}_T}{V}\frac{1}{\Omega\tau} \tag{42}$$

where λ is the wavelength of the solid-state laser and λ_A is the wavelength of the argon laser used in [15] for measuring the optical Kerr constants, $\lambda_A = 4880$ Å. For the following experimental situation—a 5-cm cell containing nitrobenzene ($B_0 = 2.9 \times 10^{-7}$ esu, $\tau_{300°K} = 5 \times 10^{-11}$ second) and a laser cavity 1 meter long with a beam cross section of 1 cm^2 and a total energy of 0.1 joule—we find a circulation time T_0 of the order of 1 ns and, therefore, sufficient energy exchange between cavity modes is expected to take place to produce efficient mode coupling within the duration of typical Q-switched laser pulses ($> 10^{-8}$ second).

Discussion and Conclusion

The important parameters of the anisotropic molecular liquid are its optical Kerr constant ϵ_2 and its orientational relaxation time τ. The rate of energy exchange as expressed by (41) is proportional to ϵ_2 and inversely proportional to $\Omega\tau$; therefore, for stronger mode coupling $\Omega\tau$ should be as small as possible. However, there is a lower limit to the possible values of $\Omega\tau$. This is explained by examining the physical significance of this parameter.

The rate at which energy is exchanged between the nth mode and the $(n + p)$th mode, for example, is proportional to the quantity $p\Omega\tau/[1 + (p\Omega\tau)^2]$, which is equal to the imaginary part of the nonlinear susceptibility of an anisotropic molecule in an electric field with radian beat frequency $p\Omega$. The maximum of this quantity occurs at $p = 1/\Omega\tau$.

If $\Omega\tau \gg 1$, the molecular orientation does not respond to optical envelope variations at frequency Ω or higher and the amount of refractive index nonlinearity is too small to couple the modes together.

If $\Omega\tau = 1$, the nth mode exchanges energy principally with adjacent modes. The rate at which it receives energy from higher frequency modes is maximum for the $(n+1)$th mode and decreases rapidly for the $(n + 2)$, $(n + 3)$ modes, and so on. In order to couple more modes faster, the relaxation time τ of the molecule has to be made shorter so that the molecules respond to more frequency components. But there is a limit to how short $\Omega\tau$ should be. If $\Omega\tau \ll 1/N$, where N is the total number of oscillating modes, the nth mode is coupled most effectively with modes $n + p$ and $n - p$ where $p \gg N$, so that these modes are outside the gain linewidth. In that case most of the energy is coupled outside the gain linewidth and lost without giving rise to any appreciable coupling between the oscillating modes. In Fig. 3 the function $f(p\Omega\tau) = p\Omega\tau/1 + (p^2\Omega^2\tau^2)$, which is a measure of the mode-coupling strength, is presented for $\Omega\tau = 1$, $1/N < \Omega\tau < 1$ and $\Omega\tau < 1/N$. For producing strong mode coupling, the relaxation time must be chosen so that

$$\tau \sim \frac{2}{N\Omega} = \frac{1}{\pi\Delta\nu_G}$$

and is thus determined by the laser transition. For example, in a ruby laser with a $\Delta\nu_G \approx 2$ cm^{-1}, τ should be $\approx 10^{-11}$ second.

The relaxation time τ is given by the expression [12] $\tau = \eta V/kT$ where

η = the viscosity of the liquid
V = the volume of one molecule
T = the temperature of the liquid.

The viscosity η is a decreasing function of temperature [16]. It varies as $Ae^{B/T}$ where A and B are two, empirically found, constants characteristic of the liquid. The relaxation time τ is thus a decreasing function of the temperature. This dependence has been experimentally verified for nitrobenzene by Rank *et al.* [17] who measured the frequency shift of the stimulated Rayleigh line as a function of temperature from $T = 12$ to 117°C.

The relaxation time of the anisotropic molecular liquid

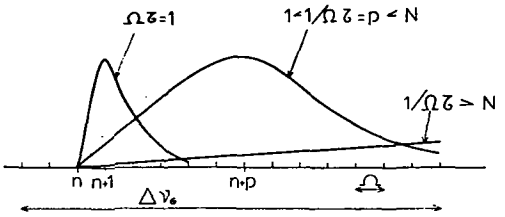

Fig. 3. The strength of the mode coupling, as represented by the function $p\Omega\tau/[1 + (p\Omega\tau)^2]$, is a function of $\Omega\tau$.

can be adjusted by controlling its temperature. For the optimum value of τ given above, nitrobenzene with a room temperature relaxation time of 50×10^{-12} second requires heating, while carbon disulfide with a room temperature relaxation time of 10^{-12} second requires cooling.

The dependence of mode locking on the proper value of the molecular orientation time τ and its manipulation by temperature has been verified experimentally [9], [10].

One important problem that we have not considered is the following. We have shown that a nonlinear refractive index can give rise to a steady-state mode locking via physical exchange of energy between the longitudinal modes of the laser. What we have not shown, however, is that starting from a nonlocked mode spectrum (i.e., arbitrary phases) the effect of the nonlinearity is to force the phases to lock. That this may be the case is suggested by the model of pulse steepening in a medium in which the index of refraction increases with the wave intensity [18]. This point is left open and it is hoped that a numerical transient analysis now in progress will help clarify it.

In conclusion, an analysis has been presented that treats the problem of multimode laser oscillation in the presence of a nonlinear dielectric medium. The steady-state laser field corresponds to mode-locked oscillation of the type giving rise to ultrashort pulses.

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