

INVESTIGATION OF THE NITROGEN LASER AS A PUMP
FOR INTERFERENCE TUNED DYE LASER SPECTROSCOPY

by

Philip Lucht

RESEARCH PROJECT

Submitted to the Department of Electrical Engineering and
Computer Sciences, University of California, Berkeley, in
partial satisfaction of the requirements for the degree of
Master of Sciences, Plan II.

Approval for the Report and Comprehensive Examination:

COMMITTEE

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L. L. Gustafson

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(1) Introduction

Among those characteristics which delimit the usefulness of an absorption spectrometer, three stand out as being most significant: (1) range of tuneability, (2) spectral resolution, and (3) brilliance. A typical laboratory absorption spectrometer has a tuning range of several thousand angstroms and a spectral resolution of $1 \text{ \AA}$. The light source usually employed is a tungsten filament having a brilliance of at most $100 \text{ W/cm}^2\text{-sr}$ over the visible portion of the spectrum. Light from the tungsten source is passed through a monochromator and the brilliance of the resulting sample probe beam, which we assume to have a spectral width of $1 \text{ \AA}$, is thus about $30 \text{ mW/cm}^2\text{-sr}$.

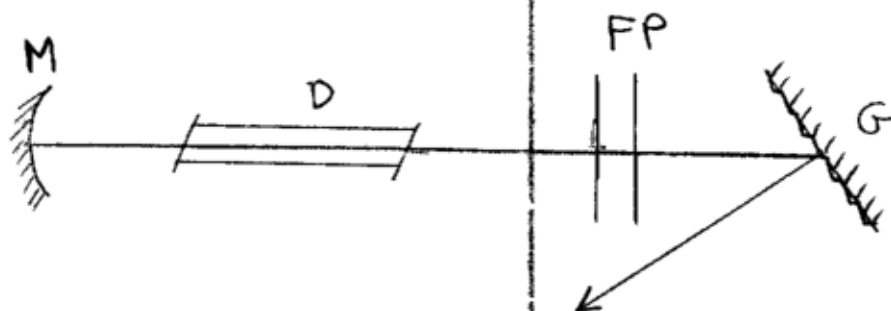
For a given light source, an improvement in spectral resolution may be bought only at the expense of brilliance, for as the spectrometer slit is made narrower, less light gets through. One is thus led to seek a more brilliant source of illumination. The brightest conventional light source known is the arc lamp. Although the emission spectrum of an arc lamp is highly irregular, fractional differential absorption techniques make it possible to use such a source for spectroscopy. Nevertheless, arc lamps provide less than an order of magnitude improvement in brilliance. For example, a PEK 110 arc lamp has a total brilliance in the visible of only $250 \text{ W/cm}^2\text{-sr}$.

It is interesting to compare these brilliance figures with those typical of lasers. As pointed out by Klein¹, a $4 \text{ mW } 6328 \text{ \AA}$ helium-neon laser with a beam of 1 mm radius and 2 mrad angular divergence has a brilliance of $10^6 \text{ W/cm}^2\text{-sr}$. Moreover, the single-mode helium-neon laser is monochromatic. Judged on the criteria of brilliance and resolution, the helium-neon laser rates very well as a potential spectrometer light source. Unfortunately, its range of tuneability is only $.01 \text{ \AA}$.

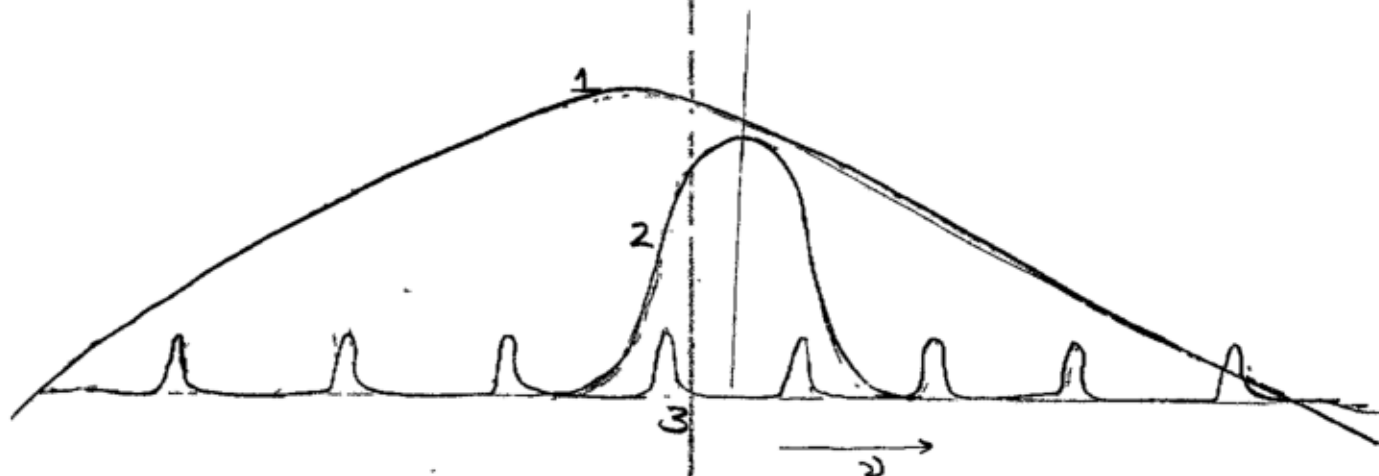
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Tuneable parametric oscillators had many problems associated with them, so it was not really until the discovery in 1966 of the tuneable organic dye laser² that broadband laser spectroscopy became a practicality. Solutions of organic dyes have been shown to exhibit gain over several hundred angstroms, as compared to .01 Å for the helium-neon mixture. In particular, the dye 4-Methylumbelliferone has a tuning range of 1760 Å.³ ~~At present~~ At present, organic dye lasers are pumped either by flashlamps, or by other lasers. Continuous argon and pulsed nitrogen lasers, for example, have both been successfully implemented as dye laser pumps. Tuning of the dye laser is accomplished by replacing one of the dye laser cavity mirrors with a rotateable diffraction grating. In this way one can generate a widely tuneable, highly brilliant spectroscopic probe beam having a spectral width of .5 Å. And by using several different dyes, the entire visible spectrum can be scanned.

An improvement in the resolving power of the dye laser spectrometer is the next logical step. In order to be able to study the fine structure in the absorption spectra of weakly absorbing compounds, chemists would find a broadly tuneable laser spectrometer with a resolution of 10 mÅ to be quite a useful tool. Such a high resolution cannot, however, be obtained with a diffraction grating alone. A Fabry-Perot interferometer must be inserted into the dye laser cavity as shown in Fig. 1(a). The interferometer must, of course, be short enough so that only one of its transmission maxima falls within the dye's gain profile, which profile is determined by the characteristics of the grating. Otherwise, laser oscillations can occur at more than one wavelength and the purpose of high resolution is defeated. This undesirable situation is depicted in Figure 1(b) where the laser can oscillate at two frequencies. If an interferometer has a



- (a) M curved mirror
D dye cell
FP Fabry-Perot etalon
G diffraction grating



- (b) 1 organic dye gain profile without grating
2 organic dye gain profile with grating
3 transmission peaks of Fabry-Perot interferometer

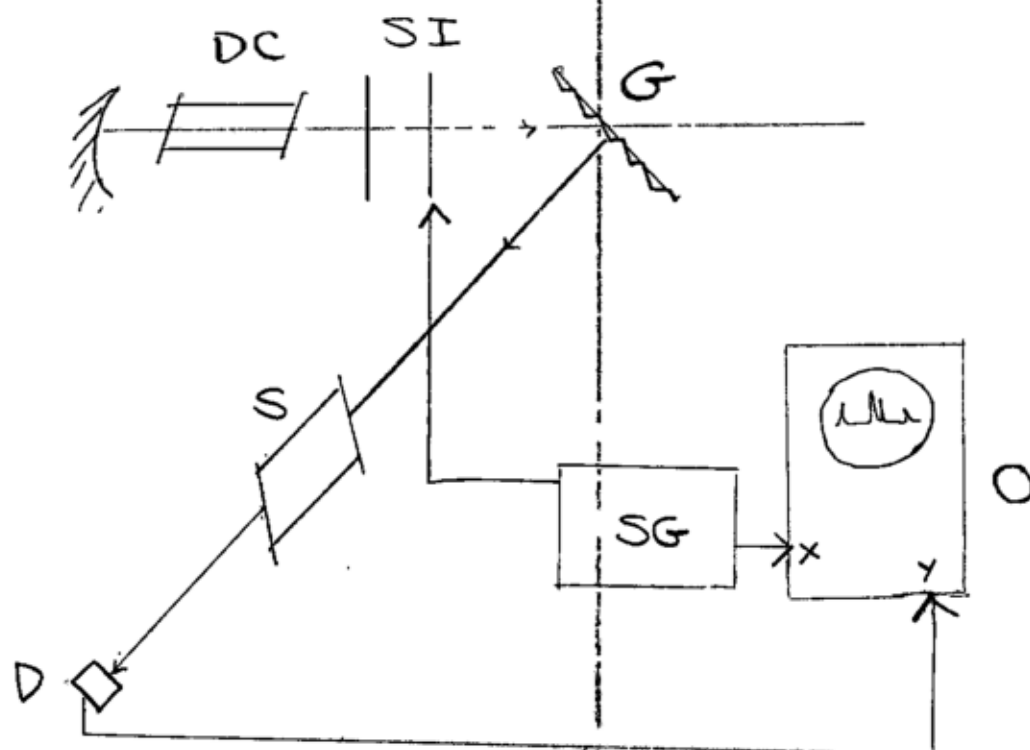
Figure 1.

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mirror separation of 5 mm, the wavelength spacing between its transmission peaks is $0.6 \text{ \AA}$ at $6000 \text{ \AA}$. Such an interferometer, then, would be adequate for the fine-tuning of a dye laser. The type of tuning system described here has been investigated by Soffer and McFarland,⁴ and by Bonch-Bruyevich et al.⁵

The inconvenience of the above tuning technique is that long range continuous tuning requires adjustment of both the grating and the interferometer. For a given setting of the diffraction grating, adjustment of the interferometer tunes the laser over a mere $0.6 \text{ \AA}$. If, on the other hand, the grating is rotated without adjustment of the interferometer, laser oscillation occurs only at discrete steps in frequency; the tuning is not continuous.

One solution to the tuning problem is to replace the Fabry-Perot device with a scanning interferometer which continuously scans back and forth across the spectral width of the diffraction grating. As the grating is rotated, the spectral window displayed on an oscilloscope connected to the detector and the scanning circuit translates through the spectrum. This scheme is sketched in Figure 2.

Another approach to dye laser tuning which dispenses altogether with gratings, interferometers, and even cavity mirrors is the use of distributed feedback. Whereas the feedback necessary for oscillation in a conventional laser is provided by cavity mirrors, the feedback in a distributed feedback ~~lax~~ (DFB) laser is, as the name suggests, distributed along the length of the active medium. In one scheme⁶, the distributed feedback is generated in a dye solution by the interference pattern caused by two pumping laser beams which are incident on the dye from different angles. By adjusting these angles it is possible to tune the dye laser in a continuous fashion over a large range of the spectrum. Moreover, the



DC dye cell
 SI scanning interferometer
 G diffraction grating
 S sample cell
 D detector
 O oscilloscope
 SG sweep generator

Figure 2.

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spectral width of the DFB dye laser emission can be made as small as 10 mÅ. All of these factors suggest that the interference-tuned distributed feedback (ITDFB) dye laser is ideally suited for high resolution spectroscopy.

At present, the only pumping source that has been used for an ITDFB dye laser is the frequency-doubled, pulsed ruby laser. In this report the possibility of using the pulsed nitrogen laser as an alternative pumping source is investigated. The obvious advantage of the nitrogen laser is its low cost; whereas ruby rods are fairly expensive, nitrogen is essentially free. A second advantage is that the nitrogen laser already operates in the ultraviolet portion of the spectrum where dye pumping is most efficient, so there is no need for frequency doubling hardware and its associated problems. The disadvantage of the nitrogen laser is the lack of spatial and spectral coherence in its output. As we shall see, it turns out that this incoherence severely limits the usefulness of the pulsed nitrogen laser as a pump for the ITDFB dye laser.

In the first section of this report we discuss DFB lasers and in particular, interference-tuned DFB lasers. The next section deals with those factors which determine the spectral width of the ITDFB laser, and the feasibility of using the nitrogen laser as a pump is evaluated. ~~Finally,~~ ~~in Section (5)~~ Experimental work with a nitrogen laser is presented in the fourth section. Finally, in Section (5), an explanation for the lack of coherence in the nitrogen laser output is given, and a suggestion for an improved pumping system is put forward.

(2) Description of the Interference-Tuned Distributed-Feedback Dye Laser

2.1 The Distributed Feedback Dye Laser

If a spatially periodic variation in gain or index of refraction is induced along the length of a narrow cell containing a gain medium such as an organic dye solution, it is found that, when pumped by an appropriate source, the dye oscillates in very narrow frequency bands and that these bands are much narrower than those found in the emissions of a dye laser which is allowed to superradiate. The periodic structure induced in the dye thus provides frequency-selective feedback so that cavity mirrors, the sources of feedback in conventional lasers, are entirely dispensed with. A laser in which feedback is provided in this fashion is called a distributed feedback (DFB) laser.

In an experiment carried out by H. Kogelnik and C.V. Shank of Bell Laboratories⁷ the index of refraction of a gelatin solution of the dye rhodamine 6G was permanently spatially modulated by a holographic exposure technique so that the spatial period of the modulation was 0.3 μm . When the resultant DFB structure was transversely pumped with an ultraviolet nitrogen laser at a pump density of about 10^6 watts/cm², the dye laser oscillated near 6300 Å with a spectral linewidth of less than 0.5 Å, this being the resolution limit of their spectrometer. This linewidth is to be compared with the 50 Å linewidth, centered at 5900 Å, characteristic of a superradiant rhodamine 6G laser without DFB under the same pumping conditions. Clearly the distributed feedback had a strong frequency-selective effect.

The wavelengths λ_d of the longitudinal resonances of the DFB structure may be obtained from the Bragg condition in reference to Fig. 3,

$$s = N \left(\frac{2d}{2n} \right), \quad N = 1, 2, 3 \dots \quad (2.1.1)$$

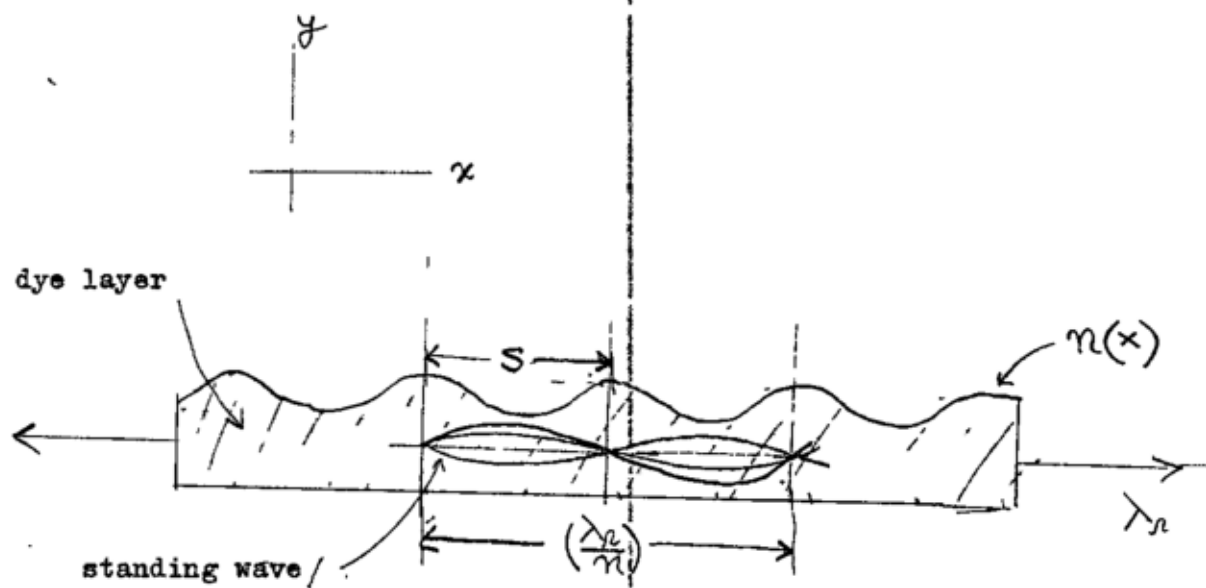


Figure 3. A standing wave fulfilling the Bragg condition (1.1) with $N=1$. $n(x)$ represents the spatial variation of the index of refraction of the dye layer having spatial period s .

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where n is the average index of refraction of the DFB medium and s is the spatial period of the DFB structure. The Bragg condition merely states that the spatial DFB period must be an integral multiple of $\lambda/2n$ the resonant ^{half} wavelength as measured in the dye ($\lambda/2n$). If s is sufficiently small, then a λ corresponding to only one value of N will fall within the dye's overall gain profile, so that only one longitudinal mode can lase. ~~Kogelnik and Shank~~ Kogelnik and Shank did find evidence of higher order transverse modes in their DFB medium resulting from the waveguide effect of periodic reflections at the gelatin interfaces. For a $14\text{ }\mu\text{m}$ gelatin thickness, the spacing of these transverse modes was about $5\text{ }\text{\AA}$. These higher order modes appeared, however, only when the dye was pumped considerably beyond its threshold. Just above threshold, the DFB dye laser output is a clean single line.

2. The Interference-Tuning Method

In the experiment described above, the spatial modulation induced in the dye was permanent and the dye laser was non-tuneable. If, however, the spatial period s was somehow made variable, tuneability could be achieved. A method which allows for adjustment of s , and hence of the resonant wavelength λ , is interference tuning.

Fig. 4 shows a thin dye layer being pumped by two ultraviolet plane waves of the same frequency (ω). These plane waves differ in amplitude, phase, and in their angle of incidence upon the dye. The electric fields of the waves are polarized in the z -direction which is out of the plane of paper in Fig 4. The combined electric field of the

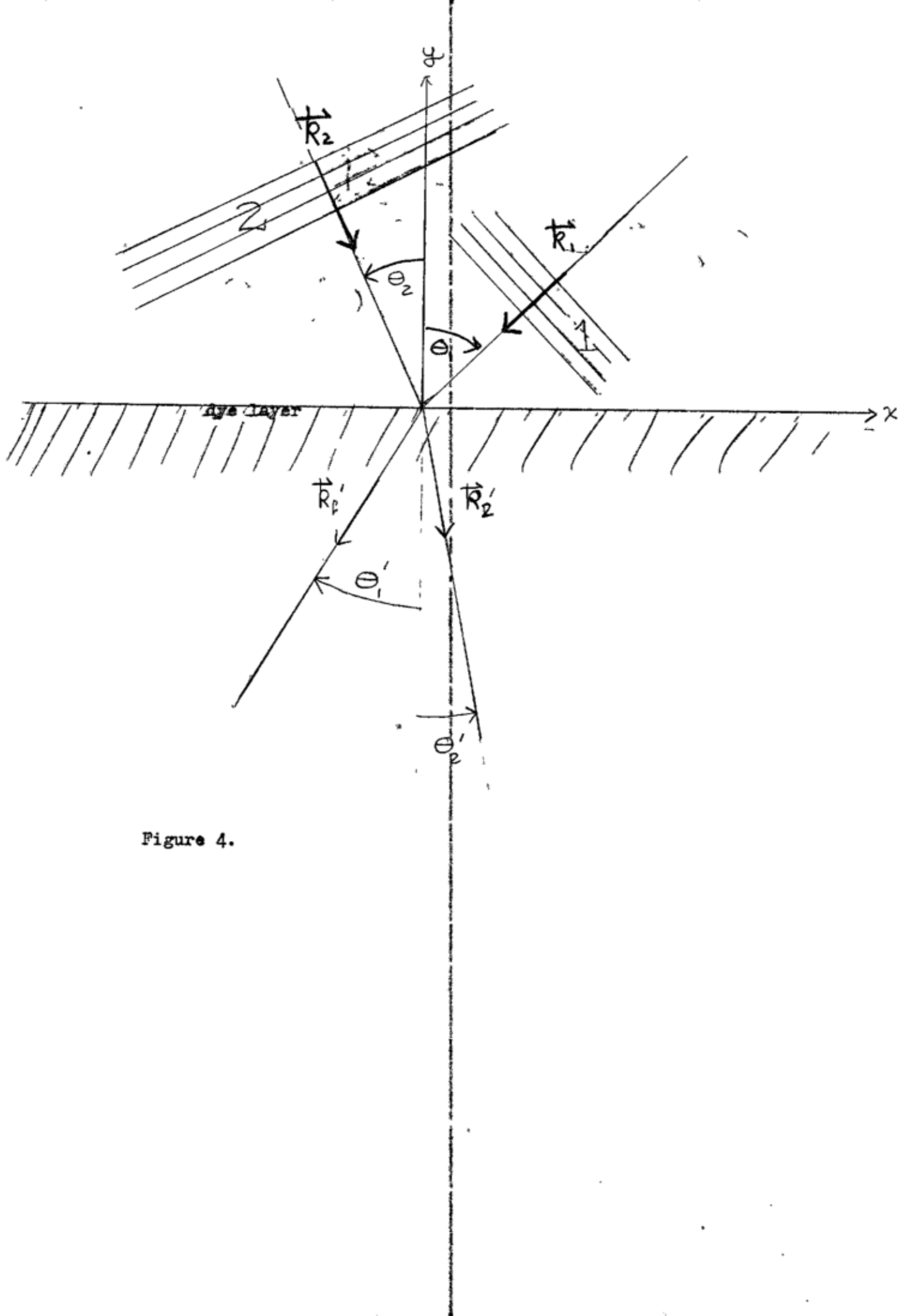


Figure 4.

two plane waves inside the dye is

$$E_z(\vec{r}, t) = A_1 \cos \beta'_1 + A_2 \cos \beta'_2 \quad (2.2.1)$$

where A_i is the amplitude of the i -th plane wave and

$$\beta'_i = \vec{k}'_i \cdot \vec{r} - \omega t + \phi_i, \quad (2.2.2)$$

where $\vec{k}'_i$ is the k -vector of the i -th wave in the dye (hence the prime), and ϕ_i is the phase.

The intensity of the electromagnetic field is given by

$$\begin{aligned} \mathcal{I}(\vec{r}, t) &= \frac{\epsilon_0}{4\pi} |\vec{E}(\vec{r}, t)|^2 \\ &= \frac{\epsilon_0}{4\pi} \left\{ A_1^2 \cos^2 \beta'_1 + A_2^2 \cos^2 \beta'_2 \right. \\ &\quad \left. + A_1 A_2 [\cos(\beta'_1 + \beta'_2) + \cos(\beta'_1 - \beta'_2)] \right\}. \end{aligned} \quad (2.2.3)$$

If this intensity is averaged over time the result is,

$$\mathcal{I}(\vec{r}) = \frac{\epsilon_0}{8\pi} \left[A_1^2 + A_2^2 + 2A_1 A_2 \cos(\Delta\beta') \right], \quad (2.2.4)$$

where

$$\begin{aligned} \Delta\beta' &= \beta'_1 - \beta'_2 = (\vec{k}'_1 - \vec{k}'_2) \cdot \vec{r} + \Delta\phi \\ \Delta\phi &= \phi_1 - \phi_2. \end{aligned} \quad (2.2.5)$$

Along the dye cell where $y = 0$ we have

$$\Delta\beta_x' = (k_{1x}' - k_{2x}')x + \Delta\phi \quad (2.2.6)$$

According to Snell's Law,

$$n \sin\Theta_1' = \sin\Theta_2 \quad (2.2.7)$$

where the Θ_1' and Θ_2 are the angles of incidence of the plane waves inside and outside the dye. Since $k' = nk$, we find that,

$$\begin{aligned} k_{1x}' &= k' \sin\Theta_1' = k \sin\Theta_1 = k_{1x} \\ k_{2x}' &= -k \sin\Theta_2 = k_{2x}. \end{aligned} \quad (2.2.8)$$

Thus, the intensity of the electromagnetic field along $y = 0$ is, from (2.2.4),

$$u(x) = \frac{\epsilon}{8\pi} [A_1^2 + A_2^2 + 2A_1 A_2 \cos(\Delta\beta_x(x))] \quad (2.2.9)$$

where

$$\Delta\beta_x(x) = k(\sin\Theta_1 + \sin\Theta_2)x + \Delta\phi. \quad (2.2.10)$$

Clearly there is a periodic variation in the intensity of the pumping field along the dye medium. Since there is a monotonic relationship between the pump intensity and the gain of the dye, the interfering plane waves cause a spatial modulation of the dye's gain. The spatial period

of the modulation is found from

$$\Delta\beta_x(s) = \Delta\beta_x(0) + 2\pi \quad (2.2.11)$$

which tells us that

$$s = \frac{\lambda_p}{(S_{\theta_1} + S_{\theta_2})^*}, \quad (2.2.12)$$

where λ_p is the pump frequency,

$$\lambda_p = \frac{2\pi}{k} = 2\pi \frac{c}{\omega}, \quad (2.2.13)$$

Combining the expression (2.2.12) for s with (2.1.1) we obtain the fundamental equation for the wavelength of the ITDFB laser,

$$\lambda_d = \frac{2n\lambda_p}{(S_{\theta_1} + S_{\theta_2})}. \quad (2.2.14)$$

Thus, the dye laser wavelength may be tuned by adjusting the angle of incidence of either plane wave. This is the method of interference tuning.

For the special case of $\theta_1 = \theta_2 = \theta^*$ we have

*The notation $\sin(x) = S_x$ and $\cos(x) = C_x$ will be used frequently.

$$\lambda_d = (n \csc \theta) \lambda_p \quad (2.2.15)$$

If, in addition, $A_1 = A_2 = A$, (2.2.9) takes on the simple form

$$\mu(x) = \frac{\epsilon A^2}{2\pi} \cos^2 \left[(k \sin \theta)x + \frac{\Delta \phi}{2} \right]. \quad (2.2.16)$$

2.3 The Results of Shank, Bjorkholm and Kogelnik.

In the early part of 1971 an interference-tuned distributed-feedback dye laser was implemented by C.V. Shank, J.E. Bjorkholm, and H. Kogelnik.⁶ The dye used was a 3×10^{-3} M solution of rhodamine 6G in ethanol. The pumping scheme is shown in Fig. 5, taken directly from their memorandum. The $3470 \text{ \AA}$ second harmonic of a pulsed ruby laser, after being passed through a cylindrical lens, was split into two nearly equal parts by a beam-splitter. These two beams were then made to cross inside a dye cell by deflection from adjustable plane mirrors. The angles of incidence of the two beams upon the dye cell were kept roughly equal. With the ruby pumping power just above the dye laser threshold of 13 kW, the ITDFB dye laser had a spectral linewidth of less than $10 \text{ m\AA}$ and was tuneable over a range of $640 \text{ \AA}$, the center of the range lying at $5900 \text{ \AA}$. The angular tuning rate was $80 \text{ \AA/degree}$ with the incidence angle θ centering at about 50° .

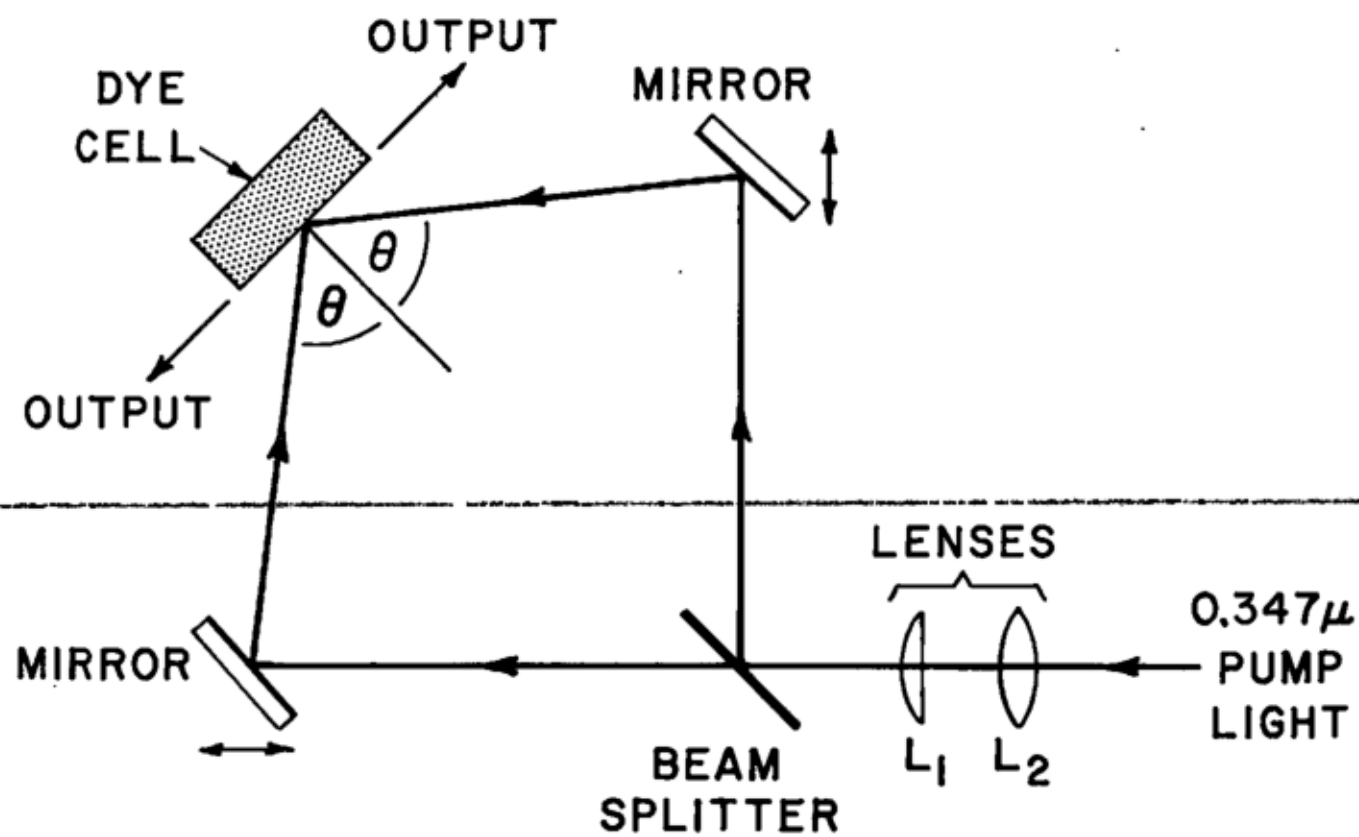


Figure 5.

FIGURE 1

(3) Determinants of the Spectral Width of an Interference-Tuned Dye Laser

In the preceding discussion of the interference tuning method we found that the time-averaged intensity along the x-axis of the two interfering plane waves shown in Fig. 4 was

$$u(x) = \frac{\epsilon}{8\pi} \left\{ A_1^2 + A_2^2 + 2A_1A_2 \cos \left[k(\sin \theta_1 + \sin \theta_2)x + \Delta \phi \right] \right\} \quad (2.299)$$

The spatial period of the gain modulation induced by this intensity variation in a dye medium lying along the x-axis was found to be,

$$s = \frac{\lambda_p}{\sin \theta_1 + \sin \theta_2} \quad (2.2.12)$$

For the idealized case of two plane waves, the spatial period s is exactly defined; so is the wavelength of the dye laser since,

$$\lambda_d = 2ns. \quad (2.1.1)$$

In a practical interference-tuned laser the plane waves are replaced with laser beams. It seems reasonable to expect that the deviations of a laser beam from a true plane wave may create a spread in the spatial modulation period s . Such a spread would manifest itself as a spectral broadening in the dye laser radiation. Since we are interested in obtaining a very narrow dye laser emission line for spectroscopic purposes, it is of great interest to know how the characteristics of the pumping laser beams influence the spectral width of the dye laser.

Those factors differentiating a practical pumping beam from a plane wave which seem to be most significant in producing a spread in s are (1) spatial divergence, (2) amplitude variation, (3) spatial incoherence, and (4) spectral spread. A plane wave, of course, being infinite in extent,

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has no spatial divergence, its amplitude is the same everywhere, and its k-vector is perfectly well-defined both in direction and magnitude (monochromatic). A plane wave thus has none of the abovementioned "problems."

1. Spatial Divergence

The dye cell employed in an ITDFB dye laser must have some finite length. If the pumping beams have a spatial divergence, the angles θ_1 and θ_2 of Fig. 4 will vary along the length of the cell, and this variation will result in a spectral broadening of the dye laser line.

Let us assume, for example, that the pumping beams originate from point sources symmetrically located with respect to the dye cell as shown in Fig. 6a where the length of the dye cell is $2l$. The fractional variation in s over the length of the cell may be calculated as follows. From (2.1.1) we have

$$s = \frac{\lambda p}{f(x)} \quad (3.1.1)$$

where

$$f(x) = \sin[\theta_1(x)] + \sin[\theta_2(x)] \quad (3.1.2)$$

and $\theta_1(x)$ and $\theta_2(x)$ are the angles shown in Fig. 6b. The fractional variation in s is,

$$\frac{\Delta s}{s} \cong \frac{\Delta f}{f} = \frac{f_{\max} - f_{\min}}{f} \quad (3.1.3)$$

Since $f(x)$ has the form shown in Fig. 6c, we may write,

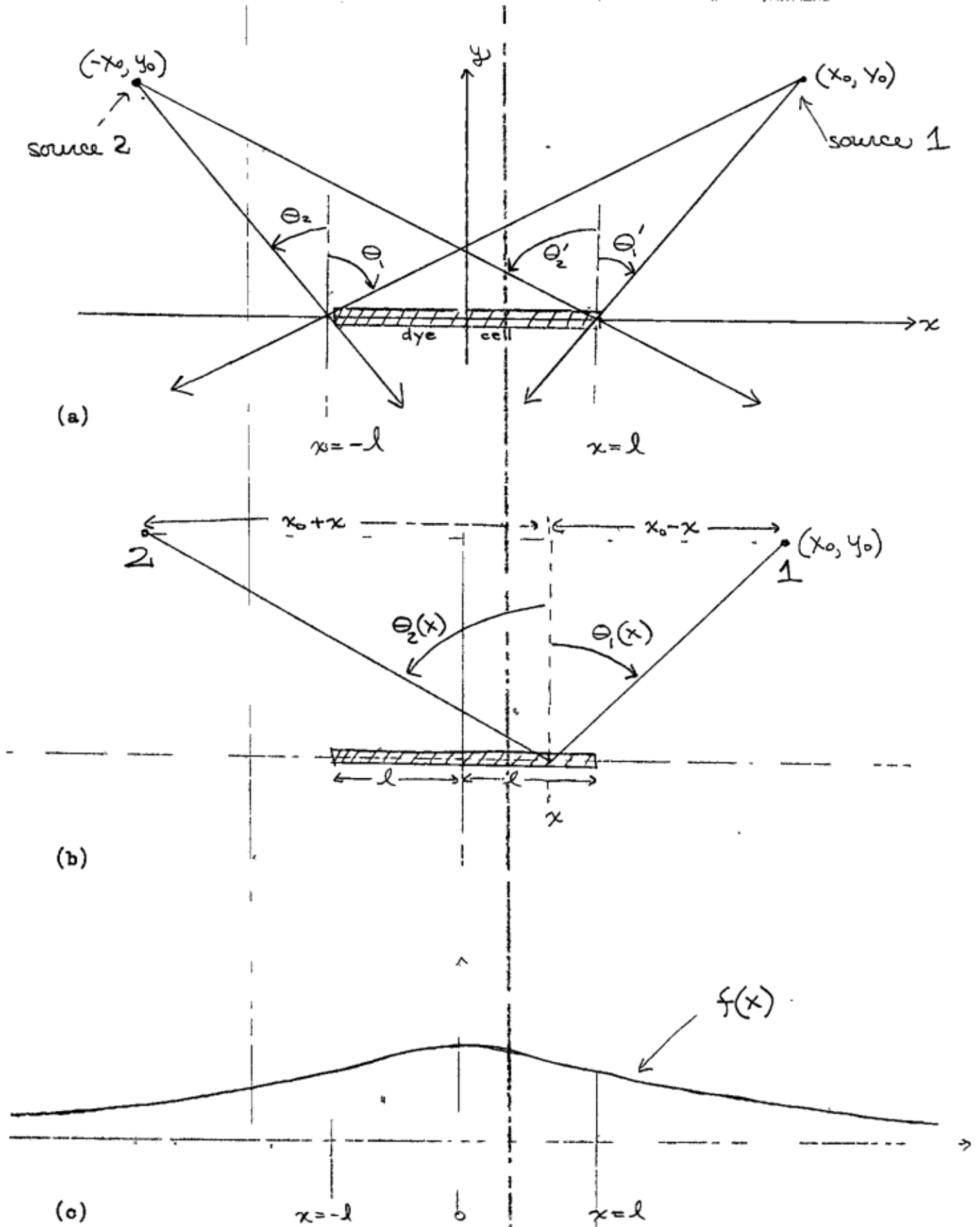


Figure 6.

$$\frac{\Delta S}{S} = \frac{f(0) - f(l)}{f} \quad (3.1.4)$$

From the geometry of Fig. 5b it is apparent that

$$f(x) = \frac{x_0 + x}{\sqrt{(x_0 + x)^2 + y_0^2}} + \frac{x_0 - x}{\sqrt{(x_0 - x)^2 + y_0^2}} \quad (3.1.5)$$

where (x_0, y_0) and $(-x_0, y_0)$ are the coordinates of the point pumping sources and x is position along the cell. By appropriate power series expansions in which it is assumed only that l is small compared to both x_0 and y_0 it can be shown that

$$\frac{f(0) - f(l)}{f} = \frac{l^2}{r_0^2} \left[1 - \frac{3}{4} \frac{x_0^2}{r_0^2} \right] \quad (3.1.6)$$

where $r_0^2 = x_0^2 + y_0^2$. If the point sources lie along the y -axis so that $x_0 = 0$, the fractional spread in s has a maximum value of

$$\frac{\Delta S}{S} = \frac{l^2}{r_0^2} \quad (3.1.7)$$

If, on the other hand, the point sources lie close to the x -axis, the fractional spread takes on its minimum value,

$$\frac{\Delta S}{S} = \frac{1}{4} \frac{l^2}{r_0^2} \quad (3.1.8)$$

If point sources were actually used for pumping an ITDFB laser, typical dimensions might be $l = 1$ cm and $r_0 = 1$ m so $\frac{\Delta S}{S} \approx 10^{-4}$. At a wavelength of $6000 \text{ \AA}$, a spectral width of $0.6 \text{ \AA}$ is implied.

Laser beams lie somewhere in the domain between plane waves and point sources. If a laser source of beam divergence α radians were placed at (x_0, y_0) as shown in Fig. 7, the effective or equivalent point source would lie much farther away than the laser itself. The effective r_0 in (3.1.7) (~~3.1.7~~) would be,

$$r_0 \cong 2l \left(\frac{\cos \theta}{\sin \alpha} \right) \quad (3.1.9)$$

and the fractional spread in the dye laser line would be, at most,

$$\frac{\Delta \lambda}{\lambda} = \frac{\Delta S}{S} \approx \frac{l^2}{r_0^2} = \frac{1}{4} \frac{\sin^2 \alpha}{\cos^2 \theta} \quad (3.1.10)$$

Here we have the fractional spectral width as a function of pump beam divergence. If we are building a spectrometer, this equation tells us the maximum divergence tolerable for a given resolution. The achievement of

a 10 mÅ spread at 6000 Å requires a pump beam divergence of less

then

$$\alpha \approx \sin \alpha = 2 \cos \theta \sqrt{\frac{\Delta \lambda}{\lambda}} = (1.4) \times \sqrt{1.7 \times 10^{-6}} \approx 2 \text{ mrad.} \quad (3.1.11)$$

where we assume that $\theta \approx 45^\circ$.

The divergence specification of 2 mrad is not too difficult to meet with a gaussian beam from, say, an argon laser. The far field divergence of a gaussian beam is determined by ^{the} ratio of the wavelength to the radius of the beam waist, w_0 :

$$\alpha = \frac{\lambda}{\pi w_0} \quad (3.1.12)$$

By transforming an argon beam with lenses, a waist radius of 1 cm could

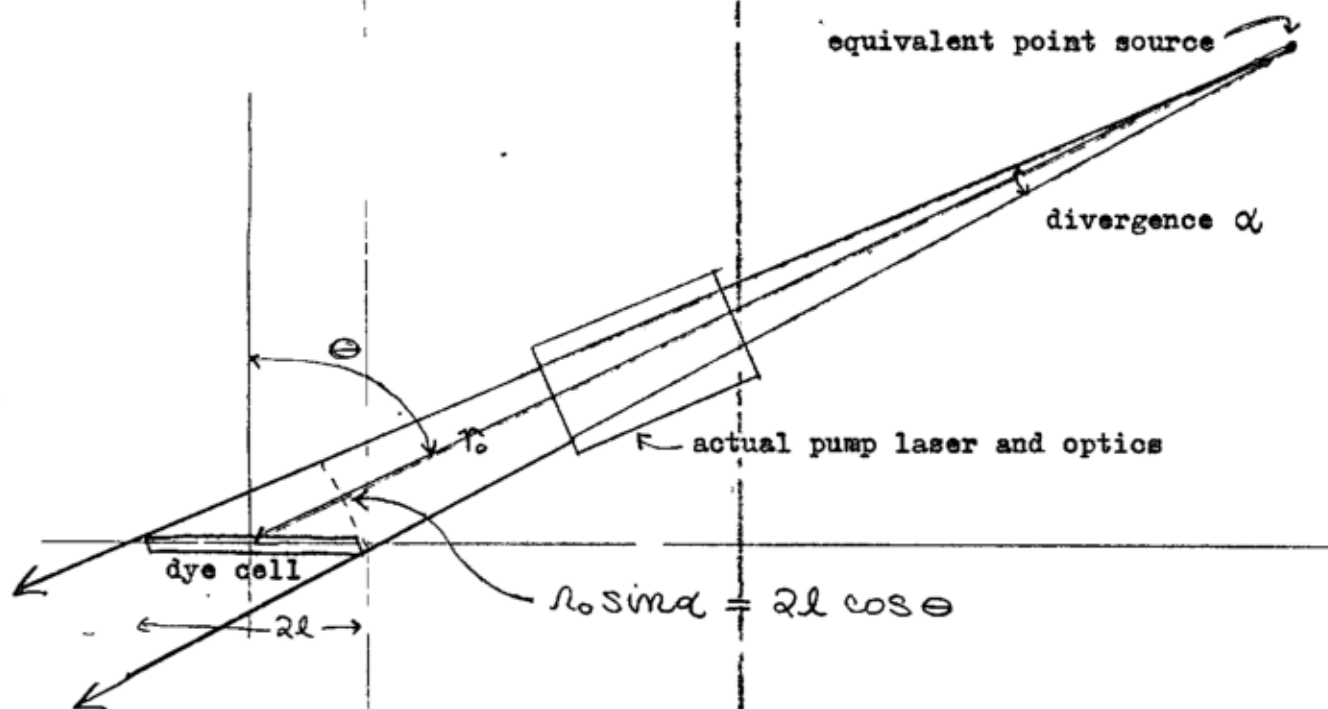


Figure 7.

easily be achieved, in which case we would have,

$$\alpha = \frac{4880 \times 10^{-8}}{3.14 \times 1} = .015 \text{ mrad,}$$

a value well within the 2 mrad limit.

Obtaining a 2 mrad divergence from a superradiant ~~nitrogen~~ laser such as the nitrogen laser is somewhat more difficult, though not impossible. As schematized in Fig 8a, the angular divergence of the superradiant beam of a nitrogen laser with a plane mirror at one end is roughly,

$$\alpha = \frac{w}{2l} \quad (3.1.13)$$

Typically, $l = 1 \text{ m}$ and $w = 1 \text{ cm}$ so $\alpha = 5 \text{ mrad}$.

Since the superradiant nitrogen laser output is spatially incoherent, it cannot be collimated into a low divergence beam as is possible with a gaussian beam or the radiation from a point source. This fact is illustrated in Fig. 8b. There, the lens succeeds in collimating the rays A, B and C, but rays D and E, which were parallel inside the laser, have a divergence after collimation which is of the same order as the divergence of the original beam.

Nevertheless, a longer nitrogen laser with a smaller cross section and just enough gain to superradiate could conceivably meet the 2 mrad criterion. However, there are much more stringent conditions on the pumping beam and these are discussed in the next sections.

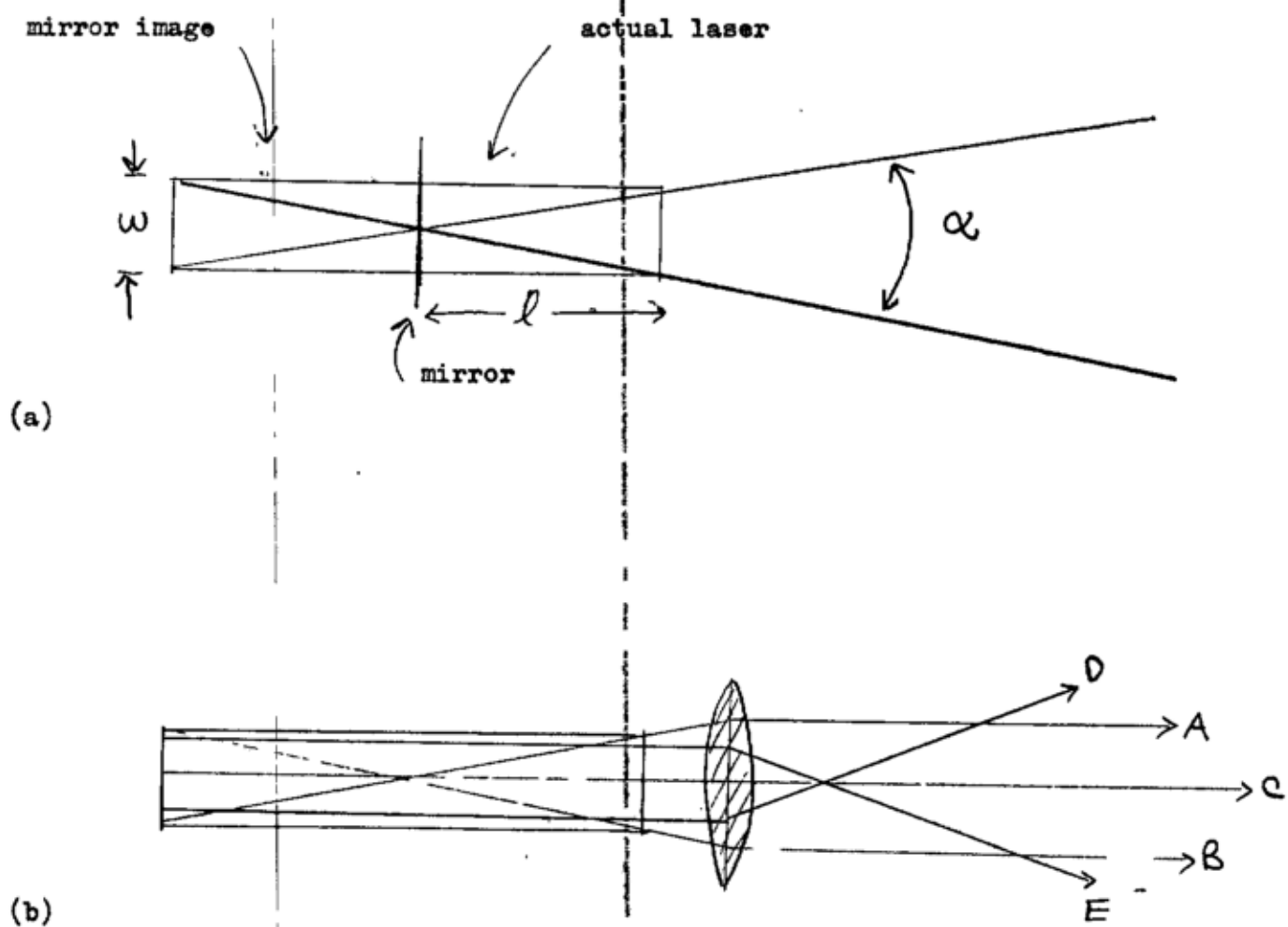


Figure 8.

2. Amplitude Variation

For simplicity, let us assume that the pumping beams used to pump the ITDFB laser are approximately gaussian. The amplitude of the electric field at any point in a gaussian beam is, by definition, a gaussian function of the distance of that point from the beam axis. As a further simplification, let us approximate the gaussian function by the single lobe of a cosine function of appropriate spatial period k_0 . This approximation is shown in Fig. 9b.*

We may enter this approximated amplitude variation into our previous equations by replacing A_1 and A_2 of Equation (2.2.1) with

$$\begin{aligned} A_1 &= A_1(\vec{r}) = a_1 \cos(\vec{k}_{01} \cdot \vec{r}) \\ A_2 &= A_2(\vec{r}) = a_2 \cos(\vec{k}_{02} \cdot \vec{r}) \end{aligned} \quad (3.2.1)$$

where the $\vec{k}_{0i}$ point in the direction of the amplitude variation of the i -th beam. Along the x -axis where the dye cell lies we have,

$$\begin{aligned} \vec{k}_{01} \cdot \vec{r} &= (k_0 \cos \theta_1) x \\ \vec{k}_{02} \cdot \vec{r} &= (k_0 \cos \theta_2) x \end{aligned} \quad (3.2.2)$$

The electric field given by substituting these new A_i into (2.2.1) may be squared and time-averaged as before; however, it is more convenient to use the intensity equation (2.2.9) directly, substituting for the A_i . The resulting time averaged intensity of the interfering beams is given by, after expanding all the cosine products,

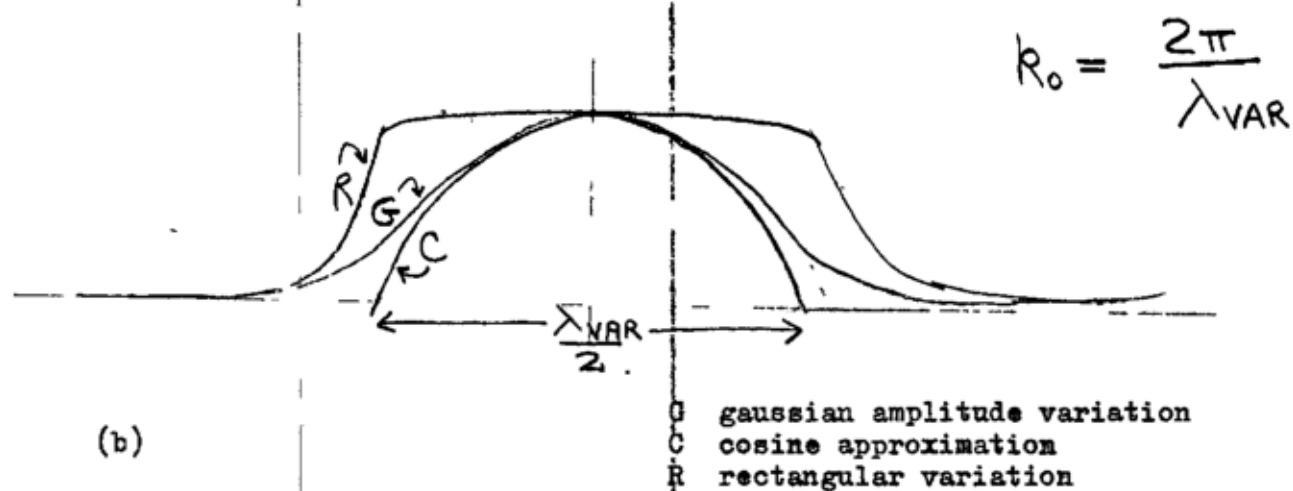
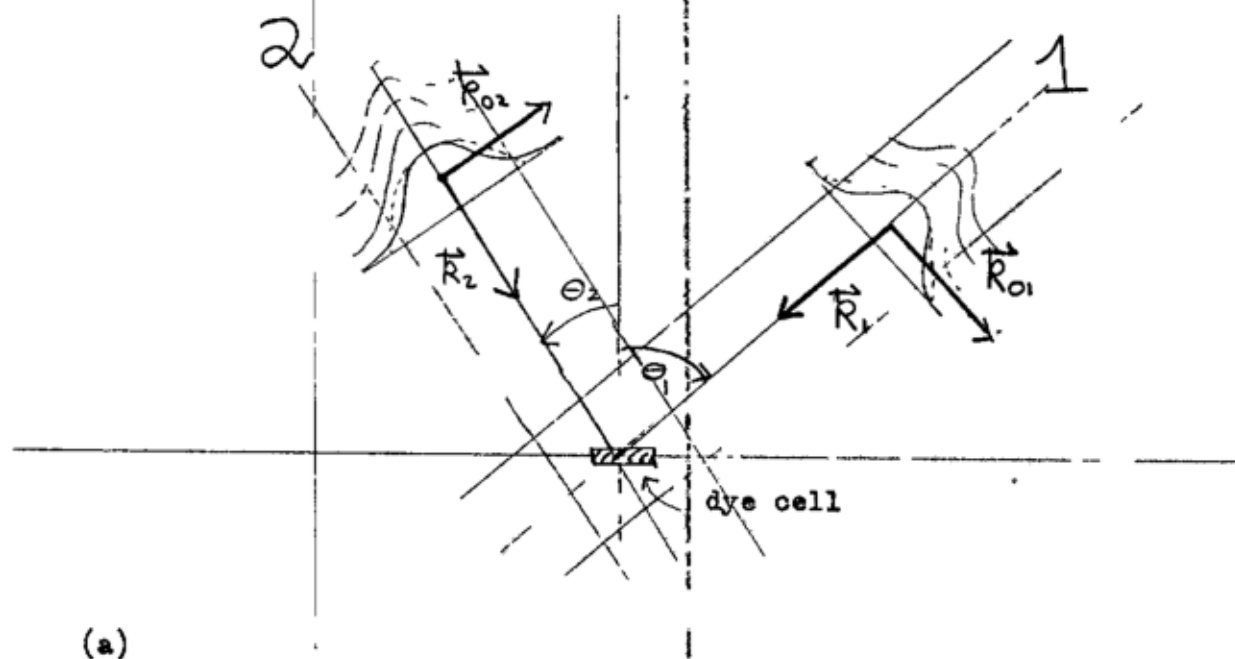


Figure 9.

$$u(x) = \frac{\epsilon}{8\pi} \left[a_1^2 \cos^2(k_0 C_{\theta_1} x) + a_2^2 \cos^2(k_0 C_{\theta_2} x) \right. \\ \left. + \frac{1}{2} a_1 a_2 \left\{ \cos(\Delta\beta_x + k_0(C_{\theta_1} + C_{\theta_2})x) \right. \right. \\ \left. + \cos(\Delta\beta_x + k_0(C_{\theta_1} - C_{\theta_2})x) \right. \\ \left. + \cos(\Delta\beta_x - k_0(C_{\theta_1} + C_{\theta_2})x) \right. \\ \left. + \cos(\Delta\beta_x - k_0(C_{\theta_1} - C_{\theta_2})x) \right\} \right] \quad (3.2.3)$$

where, from (2.2.10),

$$\Delta\beta_x = k(S_{\theta_1} + S_{\theta_2})x + \Delta\phi. \quad (3.2.4)$$

The result shows that there are four significant* spatial modulation components in the dye all having the same amplitude. The four spatial periods are given by permuting the signs in the following expression,

$$S_{\pm,\pm} = \frac{2\pi}{k(S_{\theta_1} + S_{\theta_2}) \pm k_0(C_{\theta_1} \pm C_{\theta_2})}. \quad (3.2.5)$$

The fractional separation of the two farthest separated periods is

$$\frac{\Delta S}{S} = \frac{(S_{-+}) - (S_{++})}{S} \approx 2 \frac{k_0}{k} \left(\frac{C_{\theta_1} + C_{\theta_2}}{S_{\theta_1} + S_{\theta_2}} \right). \quad (3.2.6)$$

*The spatial frequencies (wave numbers) of the first two terms are too low to matter; the dye could never lase at a wavelength the order of k_0 .

Taking both angles to be 45° , we arrive at an expression for the spectral broadening of an ITDFB laser due to amplitude variations in the pumping beams:

$$\frac{\Delta\lambda}{\lambda} = \frac{\Delta S}{S} = 2 \frac{\lambda_p}{\lambda_{VAR}}$$

In order to achieve a spectral width of $10 \text{ m}\text{\AA}$ at $6000 \text{ \AA}$ we need a value of λ_{VAR} greater than

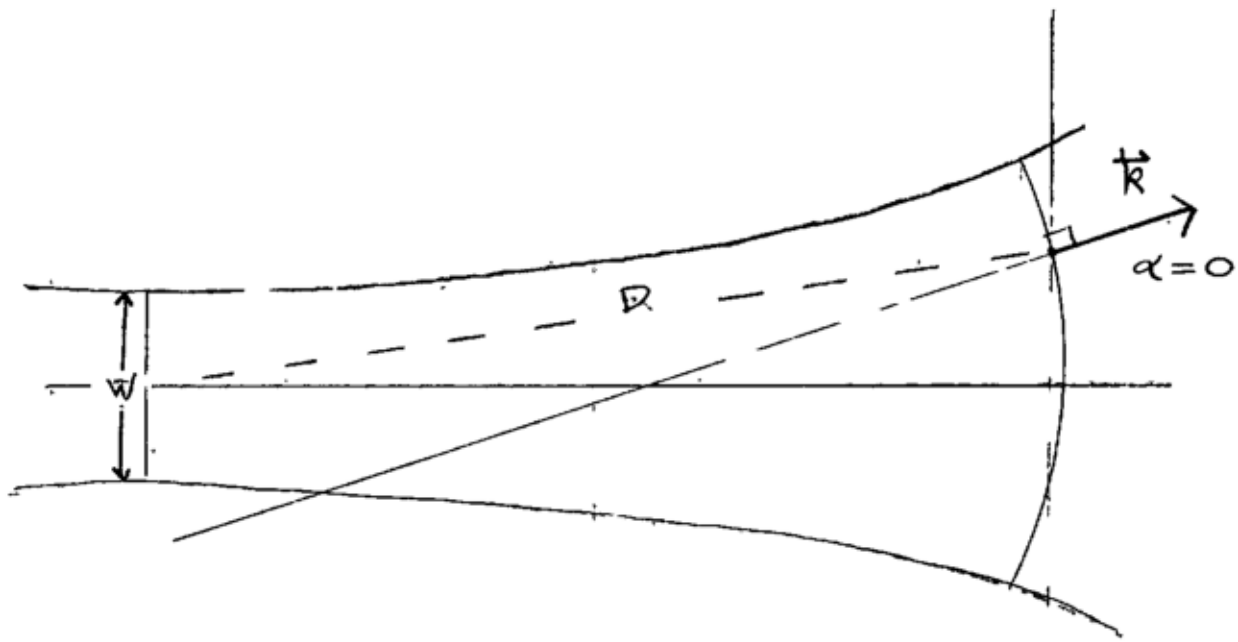
$$\lambda_{VAR} = 2\lambda_p \left(\frac{\Delta\lambda}{\lambda} \right)^{-1} = 2(3 \times 10^5) 10^6 = 60 \text{ cm.}$$

With the use of cylindrical lenses, the variation in the pumping beams across the dye cell may be made to look much like the rectangular variation shown in Fig. 9b. The effective λ_{VAR} of the cosine which approximates this variation over the cell may easily be made greater than 60 cm, so we conclude that amplitude variations of this nature, though important, are not critical. The criterion discussed in the next section, it turns out, is much more critical for the nitrogen laser pump.

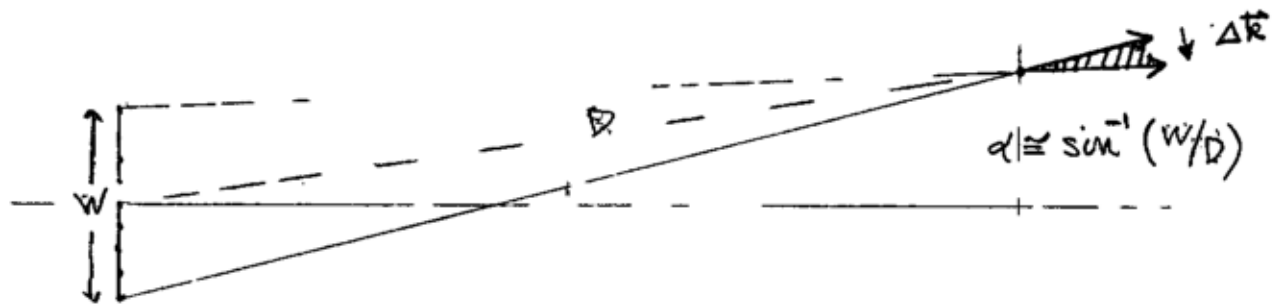
3. Spatial Incoherence

Spatial incoherence describes any situation in which there is a lack of phase correlation between the fields at two points in space. One type of spatial incoherence which has a serious broadening effect on the spectral width of the ITDFB laser is what might be called k -vector spread. The problem of k -vector spread in a pump beam is related to the problem of divergence described above, but it is not the same thing, as will be shown.

At any point in a plane wave, the k -vector is perfectly defined. The same is true in a gaussian beam, for at each point in the beam $\vec{k}$ is exactly normal to the curved phase front and the k -vector spread is zero at that point. For clarification of this point, compare the k -vector spread of an incoherent source with that of a gaussian beam of identical geometry as shown in Fig. 10. With the incoherent source, there is a k -vector spread of $\sin^{-1}(W/D)$ radians, whereas the coherent source has ~~zero spread~~.



(a) coherent gaussian beam



(b) incoherent source

Figure 10.

The effect of a k-vector spread on the ITDFB laser can be estimated with the aid of Fig. 11. Each of the original beams shown in Fig. 4 is replaced with two beams of the same phase, but half the amplitude of the original beam. The k vectors of the new beams are displaced by a small amount $\Delta \vec{k}$ from the k vectors of Fig. 4. As $\Delta \vec{k} \rightarrow 0$, the situation depicted in Fig. 4 and described in Section 2.2 is approached exactly. As before, all electric field vectors point along the z-axis, out of the plane of paper. The combined electric field of the four beams shown in Fig. 11(a) is

$$E_z(\vec{r}, t) = \frac{A_1}{2} \cos(\beta_1 + \Delta \vec{k} \cdot \vec{r}) + \frac{A_1}{2} \cos(\beta_1 - \Delta \vec{k} \cdot \vec{r}) \\ + \frac{A_2}{2} \cos(\beta_2 + \Delta \vec{k} \cdot \vec{r}) + \frac{A_2}{2} \cos(\beta_2 - \Delta \vec{k} \cdot \vec{r}), \quad (3.3.1)$$

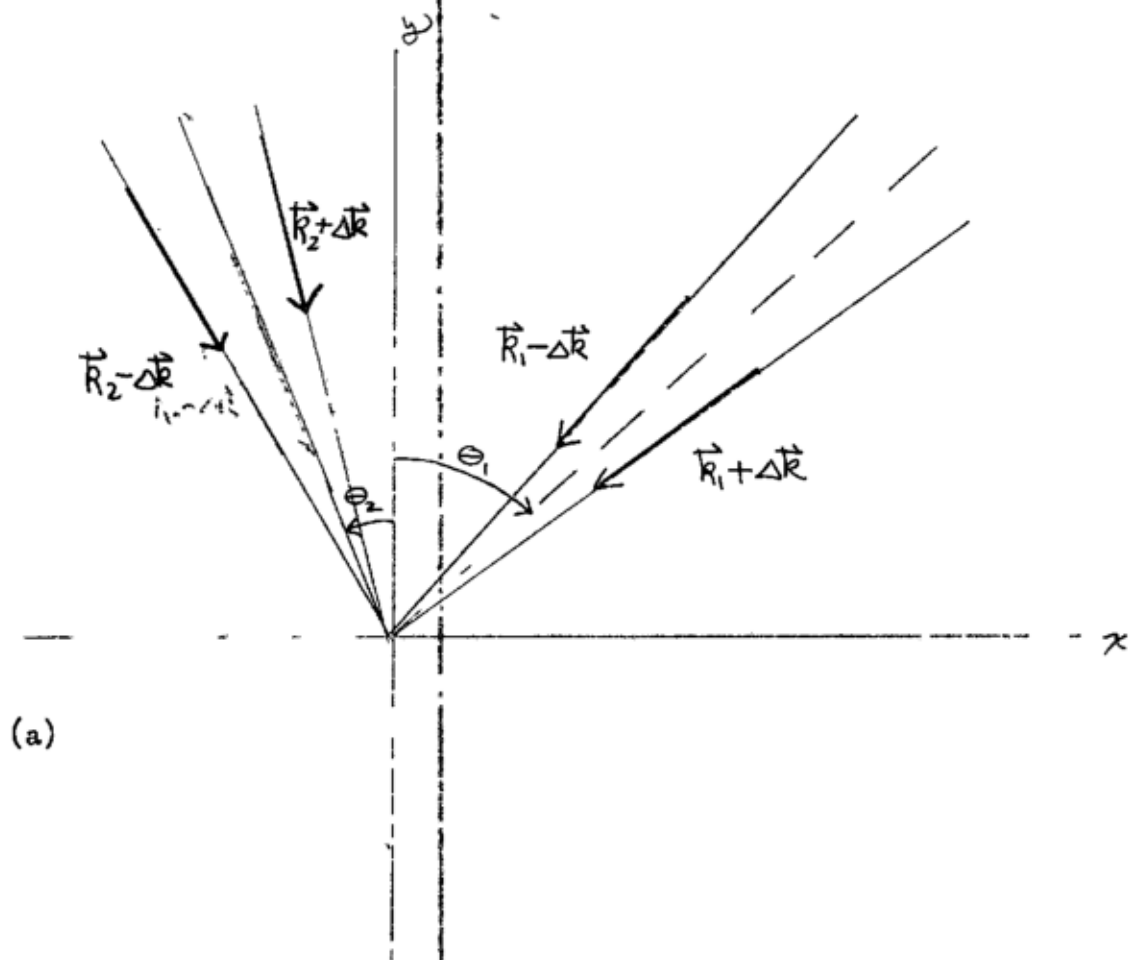
where

$$\beta_i = \vec{k}_i \cdot \vec{r} - \omega t + \phi_i. \quad (3.3.2)$$

Algebraic manipulation yields

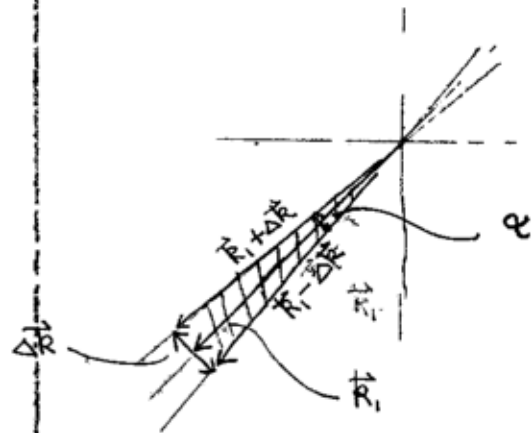
$$E_z(\vec{r}, t) = \cos(\Delta \vec{k} \cdot \vec{r}) [A_1 \cos \beta_1 + A_2 \cos \beta_2]. \quad (3.3.3)$$

The resultant time averaged field intensity becomes,



(a)

Figure 11.



(b)

$$u(\vec{r}) = \frac{\epsilon}{8\pi} \left[(A_1^2 + A_2^2) \cos^2(\Delta\vec{k} \cdot \vec{r}) + A_1 A_2 \times \right. \\ \left. \left\{ \cos(\Delta\beta) + \frac{1}{2} \cos(\Delta\beta + 2\Delta\vec{k} \cdot \vec{r}) + \frac{1}{2} \cos(\Delta\beta - 2\Delta\vec{k} \cdot \vec{r}) \right\} \right] \quad (3.3.4)$$

where

$$\Delta\beta = \beta_1 - \beta_2 = (\vec{r}_1 - \vec{r}_2) \cdot \vec{r} + \Delta\phi. \quad (3.3.5)$$

The intensity shows three significant terms. From Fig. 1-1 it is evident that,

$$\begin{aligned} R_{1x} &= k \sin \theta_1 \\ R_{2x} &= -k \sin \theta_2 \\ (\Delta k)_x &= (\Delta k) \cos \theta_1. \end{aligned} \quad (3.3.6)$$

Thus, evaluated along the x-axis, the three significant terms are

$$u(x) = \frac{\epsilon A_1 A_2}{8\pi} \left\{ \cos \Delta\beta_x + \frac{1}{2} \cos(\Delta\beta_x + 2\Delta k_x x) + \frac{1}{2} \cos(\Delta\beta_x - 2\Delta k_x x) \right\}. \quad (3.3.7)$$

The separation in the spatial periods of the two spatial sidebands is

$$\Delta S \cong \frac{8\pi(\Delta k)_x}{k^2 (S_{\theta_1} + S_{\theta_2})^2} \quad (3.3.8)$$

and the total fractional spread in s becomes,

$$\frac{\Delta S}{S} = 4 \frac{\Delta k}{k} \left(\frac{C_{01}}{S_{01} + S_{02}} \right). \quad (3.3.9)$$

If we take both angles to be 45° , the estimate for the spectral width of the ITDFB laser line due to k-vector spread is

$$\frac{\Delta \lambda}{\lambda} = \frac{\Delta S}{S} = \frac{2 \Delta k}{k} = \sin \alpha \approx \alpha \quad (3.3.10)$$

where α is the angle of the k-vector spread shown in Fig. 11(b).

Using the criterion of $10 \text{ m}\lambda$ or $6000 \text{ \AA}$, the required value for α is about 10^{-6} radians. For a pure gaussian beam, as noted above, the k-vector spread is zero radians so there is no problem. For a superadiant nitrogen laser of the type shown in Fig. 8 and redrawn in Fig. 12, 10^{-6} radians of k-vector spread is an extremely stringent specification. As shown in Fig. 12, the k-vector spread of the nitrogen laser is

$$\alpha \approx \frac{W}{D} \quad (3.3.11)$$

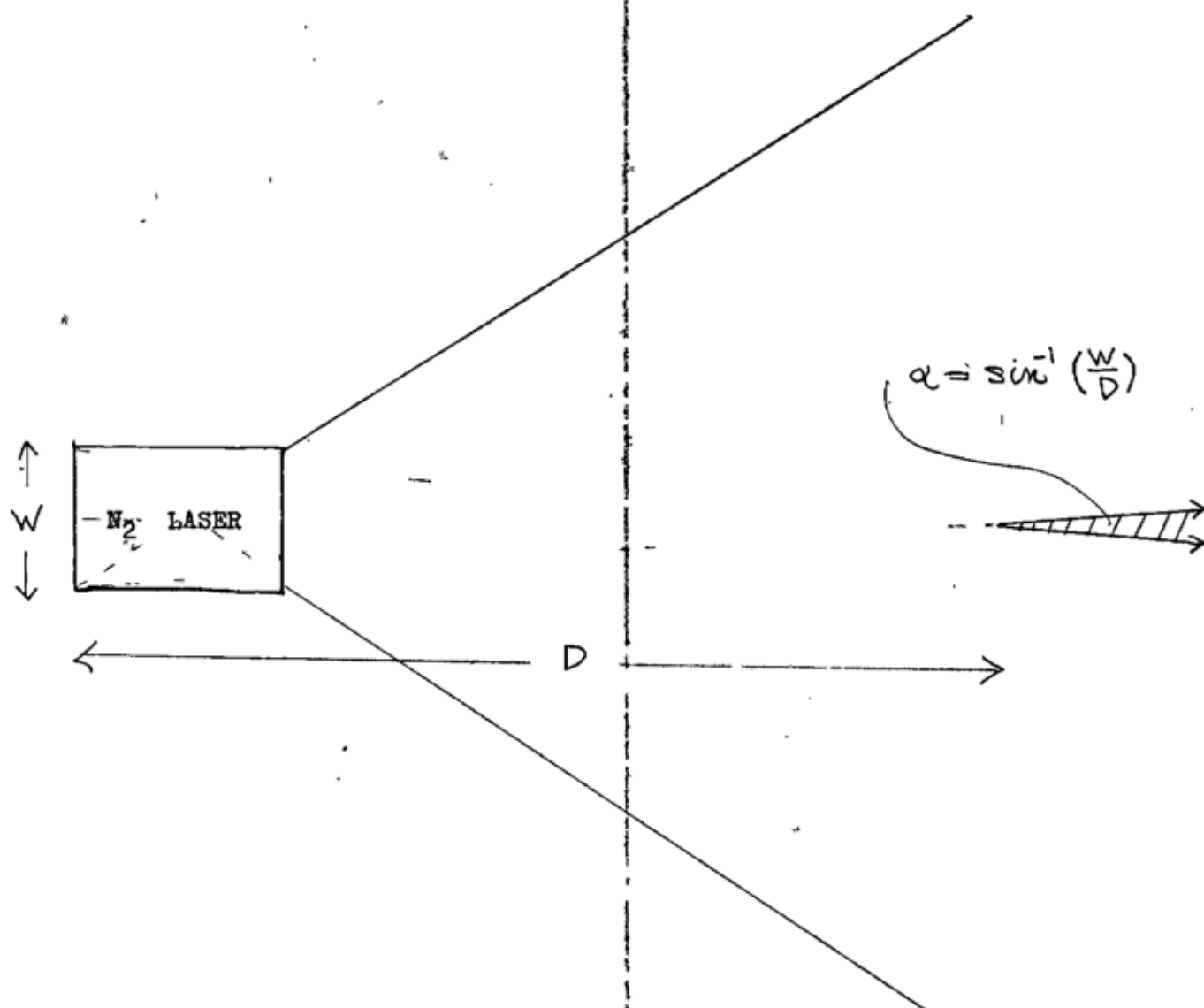


Figure 12. The k-vector spread of a nitrogen laser.

where W is the channel width and D is the distance from the laser to the point of interest. Since a channel width of at least 1 cm is necessary to generate sufficient pumping power in the nitrogen laser, the value of D needed to meet the 10^{-6} radian requirement is an impractical 10 km. Alternatively, using a reasonable value for D of 3 m, we find that θ is 3 mrad and the spectral broadening of the dye line at $6000 \text{ \AA}$ is $18 \text{ \AA}$, one third the total gain width of rhodamine 6G.

3.4 Spectral Width

The spectral width of the output of a continuous single mode laser is zero. In a cavity laser of the type that might be used to pump an ITDFB laser, several longitudinal modes oscillate at once so the laser emission consists of discrete frequencies separated by the well-known spacing $c/2\ell$. A one meter argon ion laser, for example, has a bandwidth of about 3 GHz and a longitudinal mode spacing of 150 MHz. Thus, there can be up to 20 or so lines in the argon laser spectrum evenly spread over the 3 GHz bandwidth. The spacing between the farthest separated argon lines is thus

$$\begin{aligned} \Delta\lambda_p(\text{m\AA}) &= \frac{1}{3} \Delta\nu(\text{GHz}) [\lambda(\text{k\AA})]^2 \\ &= \frac{1}{3} \times 3 \times (4.88)^2 \\ &\approx 25 \text{ m\AA} \end{aligned} \quad (3.4.1)$$

The pulsed nitrogen laser also has several discrete components in its emission spectrum, but for a very different reason. The superradiant nitrogen laser oscillates on several of the rotational lines of the nitrogen molecule, which lines span a range of $\Delta\lambda_p \approx 0.6 \text{ \AA}$.

If a multiline laser is used as an ITDFB laser pump, we want to know the effect of these multiple lines on the overall spectral width of the ITDFB laser. For a simple analysis, let us assume that the pumping

laser has only two lines of equal amplitude. The situation is adequately illustrated in Fig. 4 except that now each "beam" is made up of two plane waves separated in frequency by $2\Delta\omega$ where

$$2\Delta\omega = \frac{\omega}{\lambda_p} \Delta\lambda_p = 2\pi c \frac{\Delta\lambda_p}{(\lambda_p)^2} \quad (3.4.2)$$

The total electric field of the four interfering plane waves is, as usual, in the z -direction, and has a magnitude given by

$$\begin{aligned} E_z(\vec{r}, t) = & \frac{1}{2} A_1 \cos[\gamma_1 - (\omega + \Delta\omega)t] \\ & + \frac{1}{2} A_1 \cos[\gamma_1 - (\omega - \Delta\omega)t] \\ & + \frac{1}{2} A_2 \cos[\gamma_2 - (\omega + \Delta\omega)t] \\ & + \frac{1}{2} A_2 \cos[\gamma_2 - (\omega - \Delta\omega)t] \end{aligned} \quad (3.4.3)$$

where

$$\gamma_i = \vec{k}_i \cdot \vec{r} + \phi_i \quad (3.4.4)$$

Algebraic manipulation yields

$$E_z(\vec{r}, t) = \cos(\Delta\omega t) [A_1 \cos(\gamma_1 - \omega t) + A_2 \cos(\gamma_2 - \omega t)] \quad (3.4.5)$$

The intensity of the field is

$$u(\vec{r}, t) = \frac{\epsilon}{8\pi} \cos^2(\Delta\omega t) [A_1^2 + A_2^2 + 2A_1 A_2 \times \{\cos(\gamma_1 + \gamma_2 - 2\omega t) + \cos(\gamma_1 - \gamma_2)\}] \quad (3.4.6)$$

If the optical portion of the intensity is time averaged, the result is,

$$u(\vec{r}, t) = \frac{E}{8\pi\epsilon_0} \cos^2(\Delta\omega t) \left\{ A_1^2 + A_2^2 + 2A_1 A_2 \cos(\Delta\beta) \right\}. \quad (3.4.7)$$

where $\Delta\beta$ is given by (3.3.5). The effect, then, of a non-zero $\Delta\omega$ is a temporal amplitude modulation at frequency $(2\Delta\omega)$ of the spatial intensity distribution we found earlier in (2.2.4). No spread in the DFB spatial period is produced. s remains fixed and is determined by (2.2.12).

The periodic spatial pattern induced in the dye will rise and fall as illustrated in Fig. 13. As the average pump intensity periodically falls below the dye lasing threshold, the dye laser output falls to zero. Thus, the dye's radiation is amplitude modulated. The percentage of modulation is greater than 100%, the exact amount depending on the specific dye threshold. If we assume that the dye laser is "cut off" for half the time, then the duration of a dye laser burst as shown in Fig. 13 is given by

$$t_b = \frac{\tau}{2} = \frac{\pi}{2\Delta\omega} = \frac{\lambda_p^2}{2c \Delta\lambda_p}, \quad (3.4.8)$$

where we made use of (3.4.2), and τ is the modulation period of the dye output (see Fig. 13). In the case of the nitrogen pump,

$$t_b = \frac{(3 \times 10^{-5})^2}{2(3 \times 10^{10})(.6 \times 10^{-3})} \approx 2 \text{ picoseconds} \quad (3.4.9)$$

This duration, incidentally, is long compared to the optical period of 10^{-15} sec and short compared to the nitrogen pulse duration of 10^{-8} sec.

The frequency spread of the dye pulse implied by t_b is,

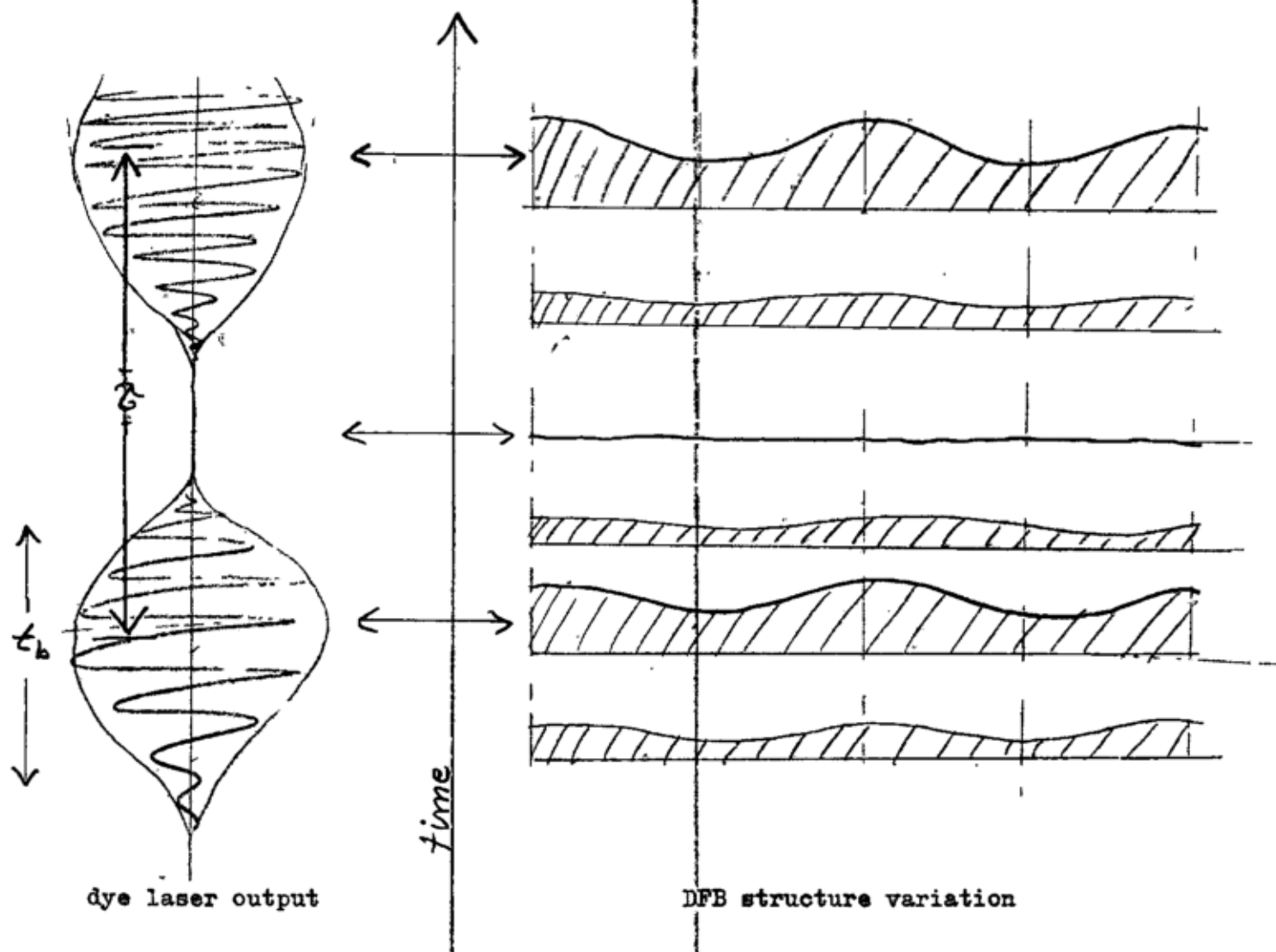


Figure 13. Correspondence of the amplitude modulated DFB dye laser output to the temporal variation in the DFB structure caused by dual line pumping.

$$\Delta\omega_d \cong \frac{2\pi}{t_b} = 4\Delta\omega \quad (3.4.10)$$

Hence, the fractional spectral width of the ITDFB dye laser emission due to a simple case of multiline pumping is

$$\frac{\Delta\lambda_d}{\lambda_d} = \frac{\Delta\omega_d}{\omega_d} = 4 \frac{\Delta\omega}{\omega_d} \quad (3.4.11)$$

Substituting from (3.4.2) and using the relation

$$\omega_d = \frac{2\pi c}{\lambda_d}$$

we obtain the result

$$\Delta\lambda_d = 2 \Delta\lambda_p \times \left(\frac{\lambda_d}{\lambda_p} \right)^2 \quad (3.4.12)$$

For the multimode argon pump laser described earlier, $\lambda_d \approx \lambda_p$ and the dye linewidth becomes,

$$\Delta\lambda_d = 2 \Delta\lambda_p \approx 50 \text{ mÅ}$$

In the case of the nitrogen laser, things are much worse. Since $\lambda_d \approx 2\lambda_p$ we have,

$$\Delta\lambda_d = 8 \Delta\lambda_p \approx 5 \text{ Å} \quad (3.4.13)$$

The numerical factor in (3.4.13) comes from the ratio $\left(\frac{\lambda_d}{\lambda_p} \right)^2 \approx 4$ and from

the assumption that $\frac{\tau_c}{\tau_b} = 2$. It cannot be reduced much below 8. Thus, in order to achieve our desired 10 mÅ dye laser linewidth, the spectral width of the nitrogen laser must be held to about 1.2 Å.

Even if the nitrogen laser is restricted to oscillate on only one of its rotational lines, it still has a finite spectral width on account of the 10 nsec pulse duration. That is,

$$\Delta\nu = \frac{1}{\tau_{\text{pulse}}} = 0.1 \text{ GHz}$$

and

$$\begin{aligned} \Delta\lambda_p(\text{mÅ}) &= \frac{1}{3} \Delta\nu(\text{GHz}) [\lambda(\text{kÅ})]^2 \\ &= \frac{1}{3} (1) (3.371)^2 \\ &\approx 0.4 \text{ mÅ}. \end{aligned} \quad (3.4.14)$$

Thus, from the point of view of single-line operation, the nitrogen laser clears the wire by a factor of three.

5. Summary of the Spectral Broadening Factors

In Sections 3.1 through 3.4 the spectral broadening of the ITDFB dye laser line was calculated for each of four pathological characteristics of the pumping beams used for tuning. The results of these calculations may be summarized as follows:

<u>Source of Broadening</u>	<u>$(\Delta\lambda/\lambda)$</u>
1. spatial divergence	$\sim \frac{1}{2} \sin^2 \alpha_1$
2. amplitude variation.....	$\sim 2\lambda_p/\lambda_{VAR}$
3. spatial incoherence.....	$\sim \sin^2 \alpha_2$
4. spectral width.....	$\sim \Delta\lambda_p/\lambda_p$

where

α_1 = angle of divergence

α_2 = angle of k-vector spread

λ_{VAR} = effective wavelength of amplitude variation

λ_p = pump laser wavelength

$\Delta\lambda_p$ = pump laser spectral width

and the symbol " $\sim$ " is to be interpreted as meaning "is on the order of."

3.6 Evaluation of the Nitrogen Laser as a Pumping Source for Interference Tuned Dye Laser Spectroscopy

In Section (1) it was decided that a desirable resolution for an ITDFB laser spectrometer is $10 \text{ m}\text{\AA}$, since this represents an improvement of at least an order of magnitude over the resolution of conventional spectrometers. At visible wavelengths, a $10 \text{ m}\text{\AA}$ resolution can be achieved if the fractional spectral spread $\Delta\lambda/\lambda$ of the ITDFB dye laser is less than 2×10^{-6} .

A transverse pumped superadiant nitrogen laser of the type commercially manufactured by Avco, Carver and other companies has the following characteristics:

$$\begin{aligned}\lambda_p &= 3371 \text{ \AA} \\ \Delta\lambda_p &\approx 0.6 \text{ \AA} \\ \alpha_1 &= 10^{-3} \text{ rad.} \\ \alpha_2 &= 10^{-3} \text{ rad.}\end{aligned}$$

As pointed out in Section 3.2, as long as the cross-sectional area of the nitrogen beam is larger than the dye cell by a considerable margin, or if a cylindrical lens is used to spread the beam in one direction so that the widened beam has an even intensity over the dye cell, the problem of amplitude variation is not significant. Typically, we may take

$$\lambda_{\text{VAR}} = 1 \text{ m.}$$

Thus, calculated values for the four broadening factors are,

1. spatial divergence	$\Delta\lambda/\lambda = 2 \times 10^{-6}$
2. amplitude variation	$\Delta\lambda/\lambda = 6 \times 10^{-7}$
3. spatial incoherence	$\Delta\lambda/\lambda = 1 \times 10^{-3}$
4. spectral width	$\Delta\lambda/\lambda = 7 \times 10^{-4}$

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Since we are seeking a fractional spread of less than 2×10^{-6} , even if the calculations of Section (3) are off by two orders of magnitude; it is clear that the superadiant nitrogen laser cannot be expected to yield a $10 \text{ m}\overset{\circ}{\text{A}}$ dye laser line in the ITDFB scheme because of its overwhelming failure to meet the 10^{-6} criterion on account of k-vector spread, the third broadening factor. It will be recalled from Section 3.3 that the dye laser linewidth is likely to be broadened out to roughly $18 \overset{\circ}{\text{A}}$ by the action of k-vector spread alone. The spectral width factor is also wide of the mark by several orders of magnitude. The first two factors, on the other hand, do not appear to pose any problem.

As a move toward improving the k-vector spread and spectral width of the nitrogen laser, a reasonable approach is to try to reduce the gain of the nitrogen laser and to put the laser tube into a resonant cavity. In this way, a transition from the superadiant regime to the more coherent cavity oscillator regime might be achieved.

(4) Experimental Nitrogen Laser Research

For purposes of investigating the possibility of using the nitrogen laser as a pump for an ITDFB dye laser, a transverse-field pulsed nitrogen laser was constructed.

1. Physical Description of Nitrogen Laser

A sketch of the nitrogen laser tube employed is shown in Fig. 14. The two electrodes, each 50 cm in length, were made from 1" O.D. polished brass tubing and were positioned within a 2½" I.D. acrylic plastic tube so that the separation between the electrodes was 1½". Each electrode was supported by three ¼" diameter brass studs which protruded through the acrylic tube and were cemented in place. Copper sheets were then used to connect the electrodes via the studs to the various discharge circuit components. The ends of the acrylic tube were fitted with 2" diameter circular windows cut from ¼" thick Corning No. 7740 pyrex glass. These windows were mounted perpendicularly to the axis of the laser tube. Two gas connections were made in the side of the tube near the ends to provide for a flow of nitrogen gas.

The discharge circuit consisted of a 30KVDC power supply, two capacitor banks, and an EG&G GP-14 triggered spark gap. A schematic diagram of the discharge circuit is shown in Fig. 15. The inductances L shown in the schematic diagram represent the distributed inductance of the copper sheets which were used in place of wires to connect the discharge circuit elements. From transmission line calculations, the value of L was found to be about 30 nH. The inductance of the spark gap is 20 nH. The capacitor banks C_1 and C_2 were composed of Sprague 30 kV 2500 pF ceramic units. The inductance of these special purpose capacitors,

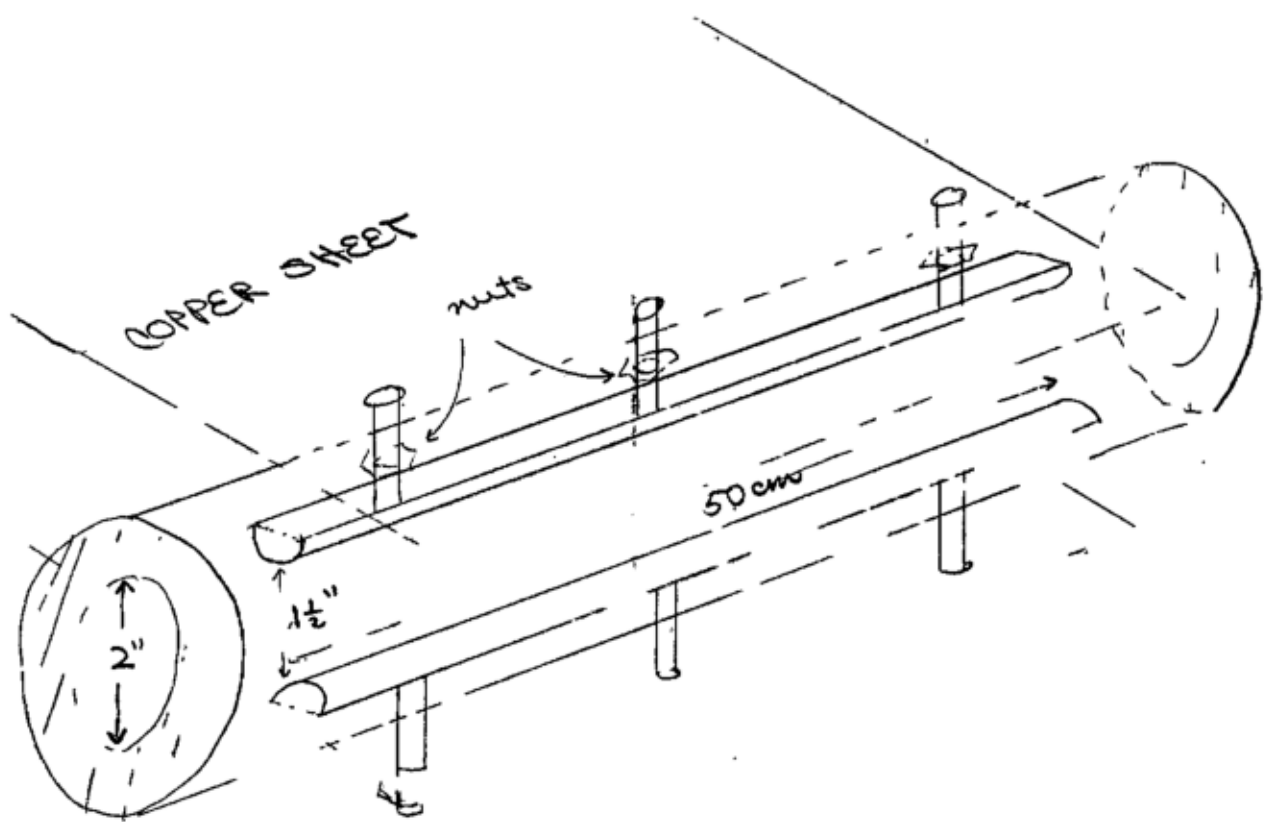
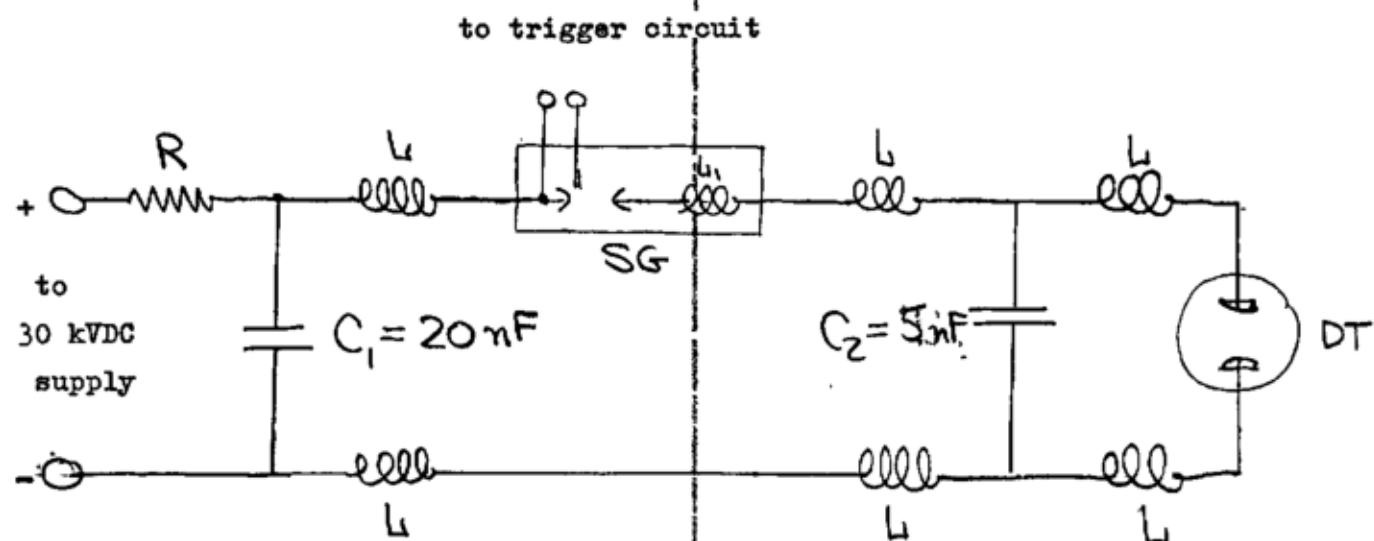


Figure 14 . The nitrogen laser tube.



L distributed inductance
 L_1 spark gap inductance
 R charging resistor
 SG spark gap
 DT discharge tube

Figure 15 . Discharge circuit schematic.

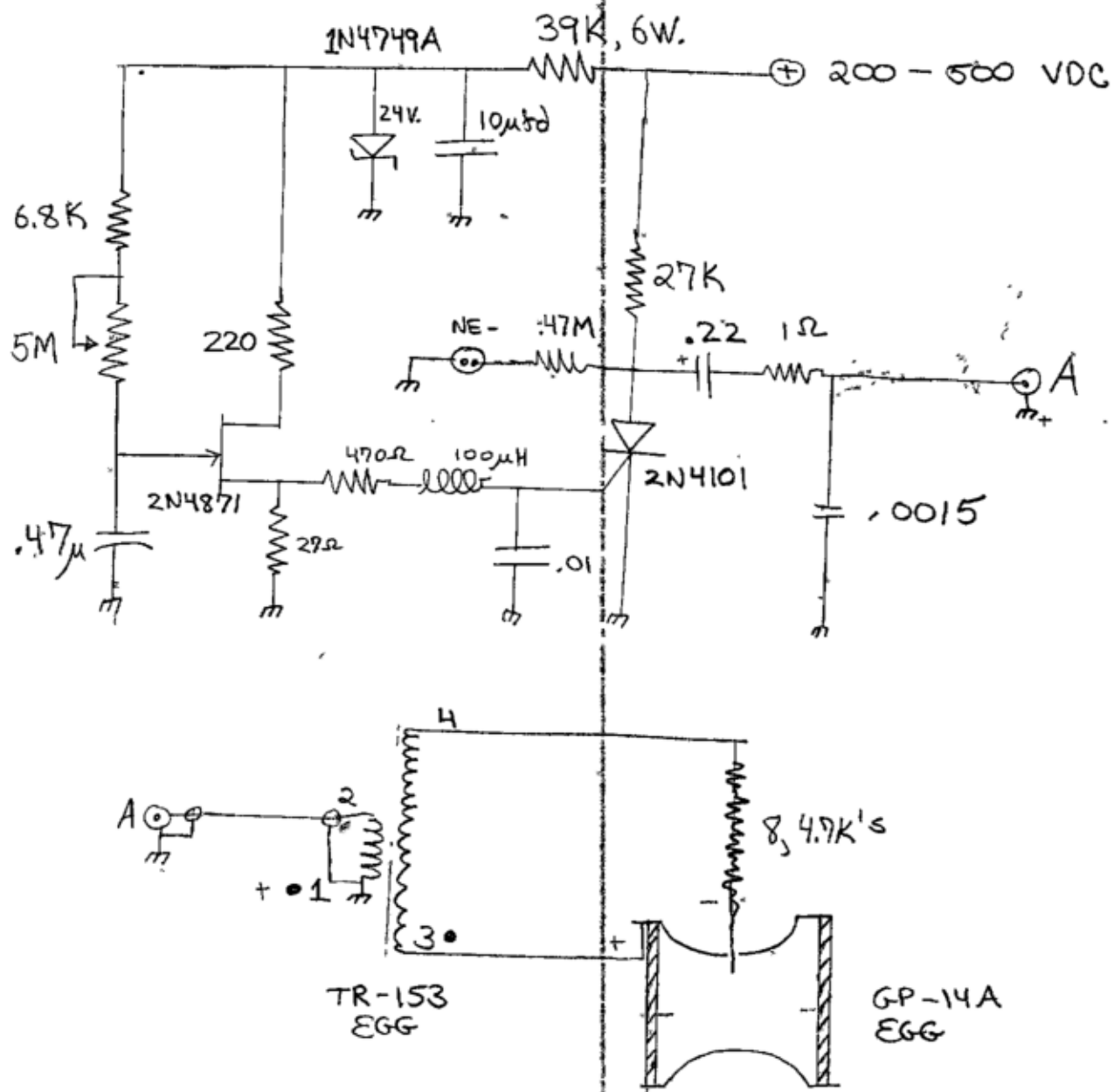


Figure 16 . Trigger circuit schematic.

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not shown in the schematic, is on the order of 2-4 nH.

The spark gap was triggered by an EG&G TR-153 trigger transformer driven by a variable frequency transistorized pulser. The complete trigger circuit is shown in Fig. 16. It was found that the polarity of the voltage pulse triggering the gap was very critical. If the leads from the trigger transformer to the gap were connected incorrectly, the gap simply did not fire at all. The correct polarity is shown in Fig. 16.

2. Performance Characteristics

The 30 KVDC power supply employed was capable of delivering a maximum average power of 150 W. Since the energy required to charge capacitor bank C_1 for each pulse was

$$W_{ch} = CV^2 = (20 \times 10^{-9})(30 \times 10^3)^2 = 18 \text{ joule}$$

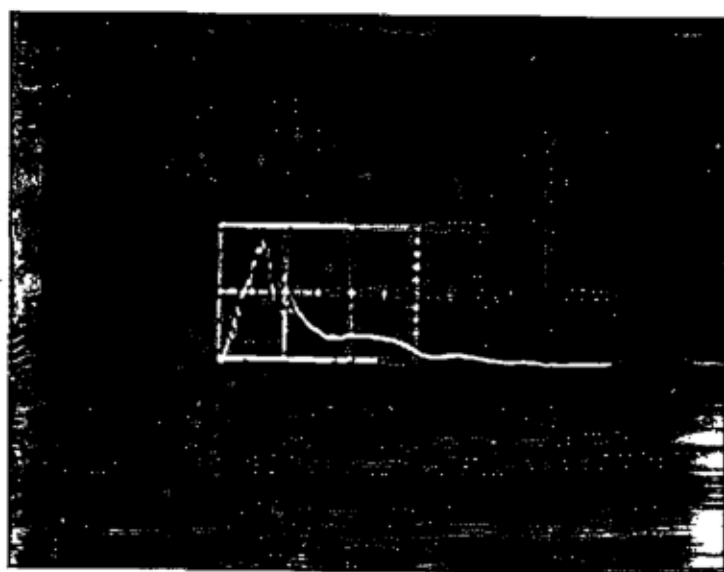
the maximum repetition rate was limited to about 8 pps.

As measured by a CDC TRG-100 calorimeter, the energy contained in each laser pulse was 0.50 $\pm$ 5 mJ. The efficiency of the laser, based on the 9 J stored in C_1 per pulse, was .005 %.

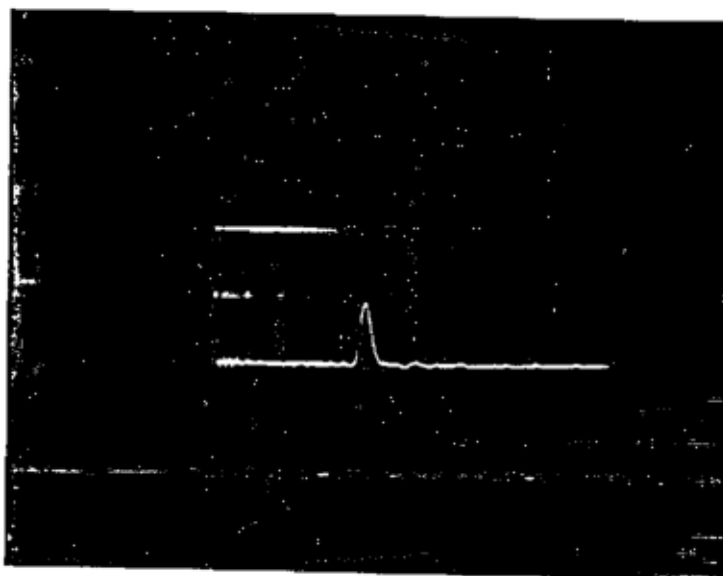
The wavelength of the nitrogen laser emission is known to be 3371 Å with a spectral width of about 0.6 Å. This width, as was noted previously, is due to the fact that the excited nitrogen gas lases on several rotational transitions at once. The duration of the laser pulse, as measured with an ITT FW-114A biplanar photodiode connected to a Techtronix Type 519 oscilloscope, was found to be 10 nsec. The pulse occurred some 80 nsec after the voltage across the nitrogen discharge began to break down. The discharge voltage and laser pulse oscillograms are shown in Fig 17a. Both traces have a time base of 50nsec/cm and were triggered by the voltage rise shown in (a). A triple trace of the laser pulse at 5 nsec/cm is shown in Fig 17c.

Since the laser pulse lasted 10 nsec and contained an energy of 0.50 mJ, the average power output of the laser during the pulse was

- (a) Voltage across discharge tube. Vert: 10 kV/cm,
Base: 50 nsec/cm.



- (b) Laser pulse.
Vert: uncalibrated,
Base: 50 nsec/cm



- (c) Three superposed laser pulses. Vert: uncal,
Base: 5 nsec/cm



Figure 17.

$$P_{av} = \frac{5 \times 10^{-4}}{10^{-8}} = 50 \text{ kW.}$$

Since the pulse was approximately sinusoidal in shape, the peak power achieved during the pulse was

$$P_{peak} = \frac{\pi}{2} P_{av} \cong 78 \text{ kW.}$$

The shape of the laser beam leaving the laser tube was rectangular, the dimensions being $1\frac{1}{4}" \times \frac{1}{4}"$. The divergence of the beam in the longer dimension was measured to be 15 mrad .

The optimum size of capacitor bank C_2 , which in our setup could ~~only~~ be changed in increments of 2.5 nF , was found to be 5 nF . The optimum nitrogen pressure was 20 torr .

A theoretical upper limit on the power available from the nitrogen laser at 20 torr can be calculated from the assumption of 100% inversion in the discharge channel,

$$P_{max} = \frac{(h\nu) n_0 V}{\tau}$$

where n_0 is the gas density, V the volume of the channel, and τ the pulse duration. For the laser described above we have $n_0 = 4 \times 10^{17} / \text{cm}^3$, $V = 75 \text{ cm}^3$, $h\nu = 7 \times 10^{-19} \text{ J}$, and $\tau = 10^{-8} \text{ sec}$. Thus, $P_{max} = 2.1 \times 10^9 \text{ W}$. Since the actual power of the pulse was 50 kW , the average percent inversion was $.025\%$.

3. The Cavity Experiment

In the conclusion of Section (3) it was pointed out that in order to obtain the desired 10 mÅ spectral width from a nitrogen pumped ITDFB dye laser, the characteristics of the nitrogen beam ~~had to be~~^{ac} improved. The purpose of the cavity experiment was to see if these characteristics could be improved by reducing the gain of the nitrogen laser and by putting it into a resonant cavity.

A choice of the type of resonant cavity to be used was governed by consideration of the volume of the optical mode that would fit in the cavity. To obtain a sufficient amount of power output for dye pumping, a large mode volume is needed. The diameter of the mode should be on the order of 1 cm, since this length is typical of the dimensions of the inverted region in the gas.

Our first inclination was to design a hemispherical cavity with a gaussian beam diameter of 1 cm. It immediately became apparent, however, that such a design was impractical because, at the ultraviolet wavelength of 3371 Å, the calculated radius of curvature for the curved mirror of the hemispherical cavity needed to obtain the 1 cm beam diameter was simply too large. This can be demonstrated as follows. The diameter of a beam formed in a hemispherical resonant cavity of length z and mirror curvature R is [8]

$$D = 2 \sqrt{\frac{\lambda}{\pi}} \left[z(R-z) \right]^{1/4}$$

The condition for cavity stability is simply that R be greater than z . For $z = 1$ m, the value of R needed to obtain $D = 1$ cm is,

$$R \approx \frac{\pi^2 D^4}{16 \lambda^2 z} \approx 6 \times 10^6 \text{ cm} = 60 \text{ km.}$$

If the wavelength were $10 \mu\text{m}$ instead of $3371 \text{ \AA}$, R would come out to around 100 m which is within the realm of practicality. Essentially, what the calculation ~~for R-tellurium~~ is that the only way to get a beam diameter of the order of 1 dm at UV wavelengths is to use a plane-parallel cavity. This is what we tried.

A plane-parallel cavity was formed by a 100% reflector and one of three available quartz beam splitters. These beam splitters, with low-loss evaporated aluminum surfaces, had reflectivities at $3371 \text{ \AA}$ of 30%, 60%, and 95%. The gain of the nitrogen tube was reduced so that the output beam, as observed by fluorescence from a moveable screen, only appeared when the second mirror of the plane-parallel cavity, the beam splitter, was in place and was perfectly aligned. Several different methods of reducing the nitrogen gain were tried: reduction of the power supply voltage; adjustment of the nitrogen pressure away from its optimum value; the addition of helium to the discharge tube. With all of these methods, and trying all three beam splitters, the smallest beam divergence that could be obtained was 5 mrad . If a coherent mode were forming in the cavity, we would expect its divergence to be on the order of

$$\Delta\theta \approx \frac{\lambda}{D} = \frac{3.4 \times 10^{-5}}{1} \approx .03 \text{ mrad.}$$

The failure of the cavity experiment to achieve a low divergence nitrogen beam with its low concomitant k-vector spread casts doubt upon the

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feasibility of using a single-tube nitrogen laser as a pump for a high resolution ITDFB dye-laser spectrometer. A possible explanation of the large divergence obtained is presented in the next section along with a suggestion for improvement.

(5) Conclusion

1. Interpretation of experimental results.

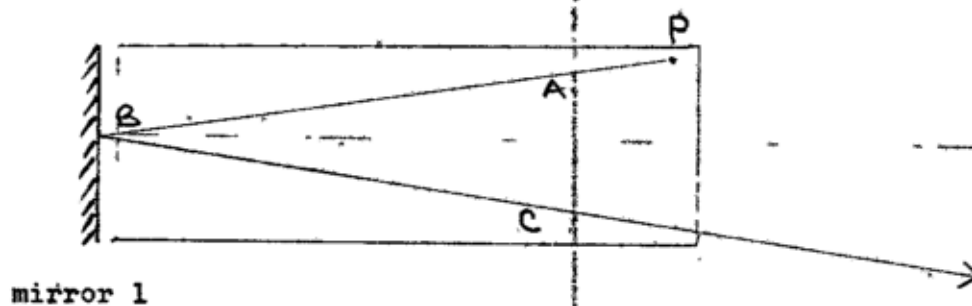
In Section 4.3 we found that the divergence of the output beam of a 50 cm long nitrogen cavity laser was many times larger than what would be expected from the diffraction of a coherent mode. It appears that the laser is operating as a multipass, spatially incoherent source rather than as a coherent source. That is, by adding a second plane mirror and by reducing the gain, the laser still behaves in a superadiant manner but with a longer effective length and corresponding decreased divergence.

The situation is illustrated in Fig. 18. When the gain of the nitrogen medium is reduced below a certain value, the amplification of the noise generated at point P along the path A-B-C in Fig. 18a, where only one mirror is used, is insufficient to produce a significant output power. However, when mirror 2 is added as shown in Fig. 18b, a longer amplification path appears along A'-B'-C'-D' and the overall gain along this path is sufficient to produce an output of several kilowatts from the spontaneous noise generated at point P'.

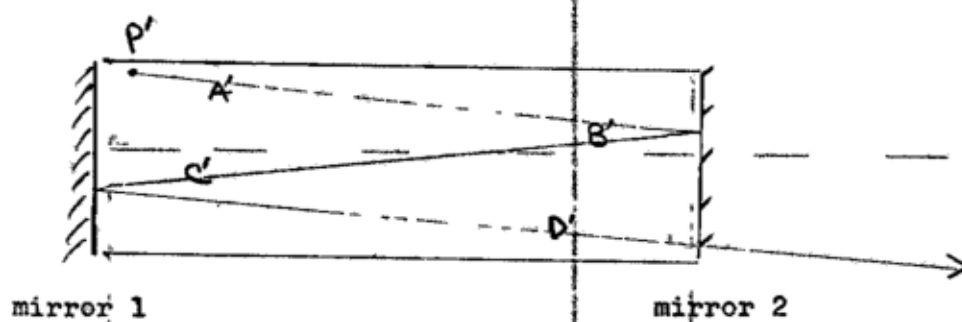
To compare the intensities of the two beams depicted in Fig. 18, we may write,

$$\left(\frac{I_b}{I_a}\right) = \frac{e^{\alpha(3L)}}{e^{\alpha(L)}} = e^{\alpha(2L)}$$

where L is the length of the cavity, α is the exponential gain in intensity, and we have assumed that the medium is unsaturated. A rough estimate for the value of α can be made from an experimental result of D.A. Leonard.^[9] He found that the small-signal (unsaturated) gain of his transverse-field nitrogen laser, which was comparable to ours, was 75 db/m. That is,



(a) A two-pass path in a one-mirror superadiant laser.



(b) A three-pass path in a two-mirror cavity. (Mirror 2 is partially transmitting.)

Figure 18 .

$$10 \log \frac{I(1m)}{I_0} = 4.3 \alpha = 75$$

Hence,

$$\alpha \cong 17$$

and

$$\frac{I_b}{I_a} = e^{17 \times \frac{1}{2}} \approx 5000.$$

The extra pass makes a very significant difference.

The obvious suggestion that comes to mind is to reduce the gain of the nitrogen medium still further so that only paths which have, say, 50 or more passes will have sufficient gain to produce kilowatts of output. Then the divergence of the output beam would be improved over that shown in Fig. 19a by a factor of 25. Since our experiment failed to yield such a large divergence reduction, we conclude that the population inversion of the nitrogen discharge did not last long enough to allow for ~~that~~ long multipass amplification paths.

The best divergence obtained was 5 mrad. For a cavity of length 50 cm and width $1\frac{1}{4}" \cong 3$ cm, the number of passes implied by the divergence figure is

$$n \cong \frac{W}{L \Delta \theta} = \frac{3}{50(5 \times 10^{-3})} \cong 12$$

The time required for light to make this number of passes through the cavity is

$$\tau = \frac{nl}{c} = \frac{12 \cdot \frac{1}{2}}{3 \cdot 10^8} = 20 \text{ nsec}$$

The conclusion, then, is that the population inversion in the laser tube only lasts for roughly 20 nsec. This figure is reasonable on two accounts. First, 20 nsec is the time scale for the voltage breakdown and current pulse as evidenced by the oscillograms of Fig. 17. Secondly, the lifetime of the upper state of the 3371 Å laser transition is 40 nsec. Once a nitrogen molecule is pumped to the upper state, it must be "used" before its lifetime expires.

2. Improving Spatial Coherence

Apparently, according to our conception of amplification in terms of rays and passes, the divergence of the nitrogen laser beam cannot be improved by decreasing the length of the cavity. Although a shorter cavity allows for more passes, the geometric factor W/L is increased by the same amount and the same overall divergence results. That is,

$$\Delta\theta = \frac{W}{nl}$$

but

$$nl = \frac{c}{2f}$$

so

$$\Delta\theta = \frac{Wf}{c}$$

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Another, perhaps more realistic, approach to the question of nitrogen laser beam divergence is to ask: what is the divergence of the dominant steady state mode of the cavity, and how long does it take for that mode to build up. The divergence of a steady state field configuration in a plane-parallel cavity is limited only by diffraction and was shown in Section 4.3 to be on the order of .03 mrad. The question of the time it takes for a mode to grow and saturate the medium was investigated by Fox and Li.¹⁰ One gets the impression from reading their paper that it takes roughly 200 single passes for a mode to build up in a well-aligned plane-parallel cavity of reasonable dimensions, as long as the Fresnell number is greater than 5. Thus, it appears that contrary to the conclusion of the oversimplified ray analysis, there is an advantage to be gained by a reduction in cavity length. If it is true that the inversion in the nitrogen laser lasts at least 20 nsec, and if 200 passes is a sufficient time for the modal buildup, a diffraction limited beam should be obtainable from a cavity of length

$$L = \frac{c\tau}{n} = \frac{(3 \cdot 10^{10}) (2 \cdot 10^{-8})}{200} = 3 \text{ cm.}$$

However, the power output of such a short nitrogen laser would be much too low to pump any dyes.*

*At this point the reader may be wondering why the ruby pumping laser used by Kogelnik and Shank (see Section 2.3) had such a high spatial coherence to be able to meet the criteria presented in Section 3. One reason is that high power ruby lasers can be fitted into very short cavities. Far more important, however, is the fact that the lifetime of the upper state of the lasing transition in ruby is several milliseconds. Once a chromium ion is excited, it is willing to wait millions of nanoseconds before it adds its quantum of energy to the cavity mode. Secondly, ruby lasers are pumped by flashlamps. The duration of a flashlamp pulse is several thousand nanoseconds, much longer than the 20 nsec pumping period in the nitrogen discharge. The modes in a ruby cavity laser, then, have plenty of time to build up.

5.3 Proposal for an Oscillator-Amplifier Nitrogen Pump

We found that, with a 3 cm cavity, a nitrogen laser should be capable of producing a spatially coherent beam. In order to obtain both the power and the spatial coherence needed for ITDFB dye laser pumping, a short 3 cm nitrogen laser could be used as an oscillator to drive another nitrogen tube employed as an amplifier. Such a configuration is shown in Fig. 19. As long as the amplifier is short enough so that it does not superradiate on a single pass, the scheme ought to work with both discharge tubes triggered at the same time. If more power is needed, one could use a longer amplifier tube if trigger delaying were used. By delaying the firing of the amplifier discharge, the amplifying population inversion is not turned on until the coherent oscillator pulse has had time to build up and is ready to drive the amplifier.

Unfortunately, we had time neither to experimentally investigate the divergence characteristics of short discharge lasers, nor to implement a dual tube apparatus.

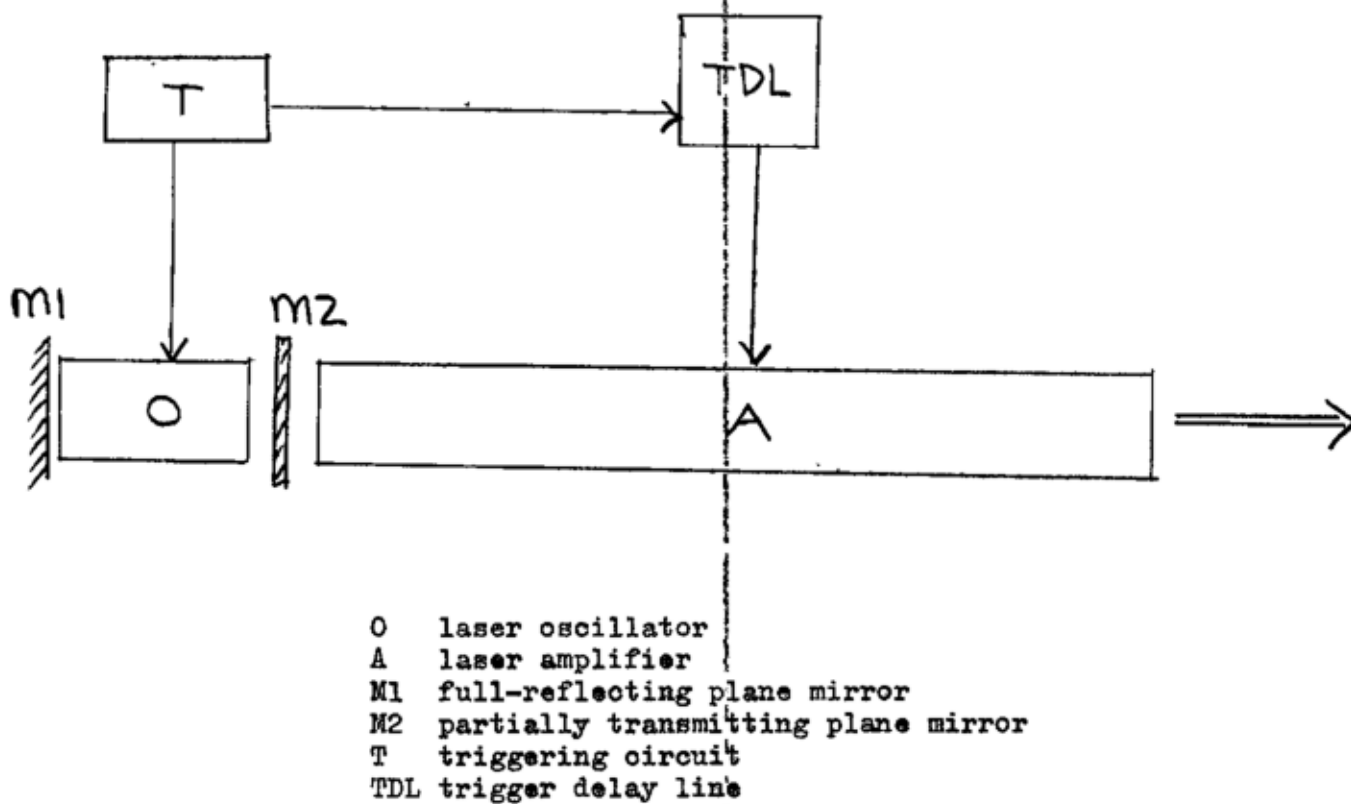


Figure 9. The Oscillator-Amplifier Configuration.

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