

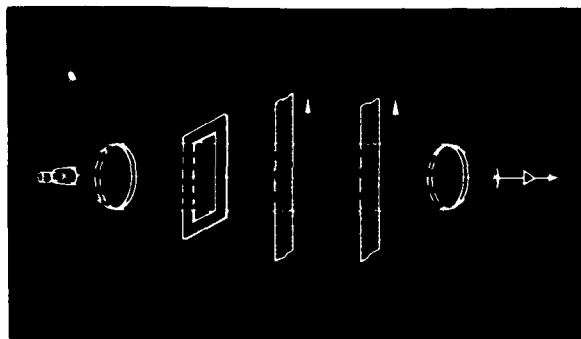
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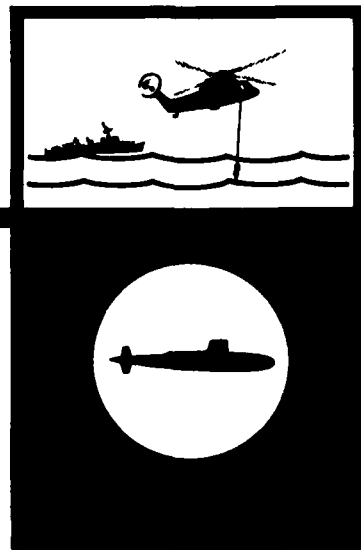
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1. THE IMPORTANCE OF OPTICAL SIGNAL PROCESSING

1.1 INTRODUCTION

Recent advances in sonar signal-processing techniques are expected to lead to significant improvements in the detection and classification capability of future ASW sonar equipment. Both passive and active sonars, and airborne (including sonobuoy) as well as shipboard systems, can be improved through better signal processing. In all cases, the improvements will result either from obtaining information at a higher rate on a given target or from making better use of the information that enters the sonar.

The importance of improved signal processing is especially apparent in the large, high-powered shipboard sonars such as the AN/SQS-26 and the AN/BQS-6. These sonars, working at peak power levels of 75 kilowatts, represent just about the practical limit to the "brute-force" approach of extending sonar ranges by increasing peak power levels. The signal-processing techniques to be discussed, however, do offer the possibility of increasing sonar range by substantially raising the average power level through use of the "pulse compression" techniques that are common in radar, and they can do so without sacrificing other desirable characteristics, such as good range resolution.

Because of the importance that advanced signal processing will assume, Lockheed Electronics Company has been engaged for over a year and a half in a Company-funded program to develop basic signal-processing techniques that are consistent with the Navy's needs and with the shipboard and airborne environments in which they must ultimately operate. This volume is a report on past results of this activity together with plans for the future.

In order that its signal-processing techniques may be of greatest use to the Navy, LEC has set several self-imposed restrictions on its approach:

1. All techniques must be consistent with operation and maintenance by Naval enlisted personnel aboard ships or aircraft without requiring extensive or unusual training.
2. Techniques must be consistent with temperature, humidity, shock, vibration, and space and weight limitations encountered in the Naval environment.
3. Techniques must be adaptable to a wide variety of ASW systems including both passive and active detection systems and classification systems.
4. Techniques must be avoided if they could lead to unusual logistic or supply problems (for example, techniques employing photographic film).

The first restraint is particularly important, since many of the more advanced signal-processing concepts currently being considered could lead to highly complex systems if implemented by standard electronic techniques. Most (if not all) of these concepts can, for example, be implemented through use of large general-purpose or special-purpose computers. General use of digital-computer equipment for signal processing could be expected to aggravate the Navy's existing problem of personnel training.

Having set the development objectives and the design restraints for ASW signal-processing techniques, LEC first considered a number of alternate approaches. After evaluating the advantages and disadvantages of each alternative, it selected the following basic approach:

1. Use of optical techniques. Optical techniques offer the most simple direct approach to performing the computations required for ASW signal-processing (as will be discussed more fully later).

2. Use of photochromic materials as the optical storage medium.

Photochromic materials, unlike photographic materials, are re-usable and they do not require development or other chemical processing. Unlike thermoplastic storage devices, they do not require handling in an evacuated electron device.

Most of the remainder of this volume is devoted to a discussion of the LEC program. Before beginning on that, however, it will prove helpful to consider the many system applications for which photochromic optical signal processing is considered suitable.

1.2 POTENTIAL SYSTEM APPLICATIONS

1.2.1 Passive Systems

Generally speaking, optical techniques can be used whenever (1) correlation-type signal processing, (2) spectrum analysis, or (3) beam-forming of array elements is required. Present systems whose concept lies within one or all of these general categories immediately suggest themselves. Two of the most obvious are the two-channel passive correlators and spectrum analyzers. Examples of the former are the AQA-3 and AQA-5 airborne systems. A further example, at least in basic concept, is the submarine-borne passive ranging system (PUFFS). In the category of spectrum analyzers, the airborne AQA-3 and AQA-5 and the submarine-installed BQQ-3 (LOFAR) systems are two well-known, operationally proven, system applications.

A third passive sonar system concept which is presently receiving considerable attention is the Multiple Array Correlator, examples of which are the DIMUS and PADLOC systems. Admittedly, optical approaches to advanced signal processing in this context become more difficult. However, the advantages of optical signal processing, even in these applications, can be demonstrated.

1.2.2 Active Systems

Communications. - The AN/BQA-2 (SESCO) underwater communication system is an example of the use of correlation techniques in active sonar systems. In this application, intelligence is transmitted in the form of coded pseudo-random noise bursts. At the receiver the burst is detected by correlation processing, which permits extraction of the encoded information. Long range would, hopefully, be achieved by trading off a low data rate against high signal processing gain at the receiver. SESCO is one of several means of implementing this basic communications philosophy, all of which require correlation-type processing. Hence, it is LEC's conclusion that they are adaptable to optical processing using photochromic signal storage.

Pulse-Compression Sonar. - LEC's experience in applying pulse-compression to radar and communications indicates that optics offer a means of applying the principles of pulse compression to underwater signals. These techniques allow sonars to transmit more average peak power into the water with longer pulses, without exceeding peak power or transducer cavitation limitations.

High-Data-Rate Sonar. - In an effort to provide data at a higher rate for shipboard fire control systems, LEC envisions a high-data-rate sonar, using pulse compression techniques, which would have several precoded transmissions in the water simultaneously. Return echos would be correlated against the stored optical replicas of the pulses such that each return echo could be identified with its corresponding transmitted pulse. Essentially, this technique is a potential alternative to frequency diversity, such as is used in the SQS-26 shipboard sonar equipment.

1.3 POTENTIAL ADVANTAGES OF OPTICAL PROCESSING

1. To the operator, the ultimate user who must live with, operate, and maintain his equipment to provide continued peak performance, optical signal processing offers several advantages:

- (a) Less complexity
- (b) Fewer training problems
- (c) Fewer components; hence, greater inherent reliability
- (d) Savings in weight and space; hence, increased "habitability"

2. To the Government, optics potentially provides equipment at less cost without sacrifice in capability.

3. To the engineer, optics provides a practical means for implementing extremely complex system concepts.

2. BASIC DISCUSSION OF OPTICAL PROCESSING

2.1 CHARACTERISTICS OF CORRELATION SYSTEMS

In any sonar signal processing sequence using correlation techniques, the problem can be expressed as one of evaluation of integrals of the form

$$O(t) = \int_{t-\Delta t}^t V_1(t) \cdot V_2(t) dt \quad (1)$$

where $V_1(t)$, $V_2(t)$ are time-varying signals (voltages, acoustic pressures, etc.)

t = time

Δt = integration period

and $O(t)$ = output signal from the processor

The requirement is for a repetitive solution in real time; in short, the integral must be evaluated over and over again.

In a correlation detector this integral takes the form of

$$O(t, \tau) = \int_{t-\Delta t}^t V(t) \cdot V(t - \tau) dt \quad (2)$$

where τ is the time delay. In correlation detectors, provision is usually made for sweeping through a range of values for time delay.

In a spectrum analyzer a like approach may be taken if the analysis is seen as the correlation of an unknown signal against a stored reference sinusoid that can be swept in frequency through the band of interest. Here, the integral being repetitively evaluated in real time takes the form:

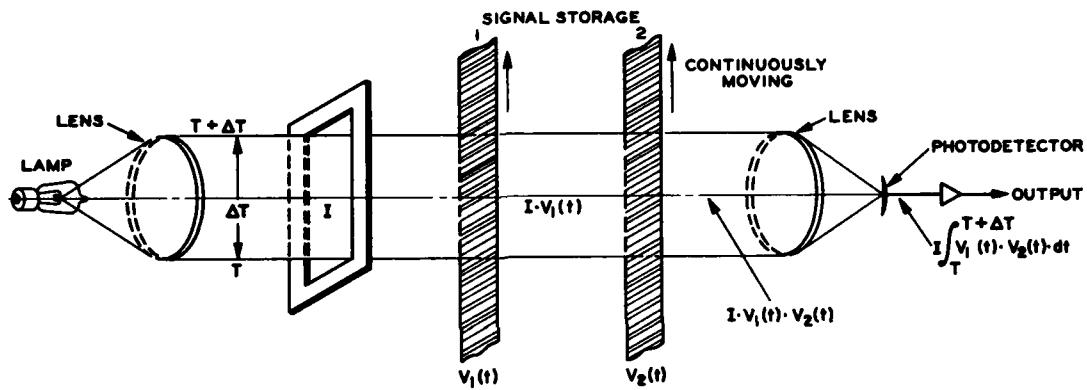


Figure 1. Elements of a Basic Optical Signal Processing System

$$O(t_1, \omega) = \int_{t-\Delta t}^t V(t) \cdot \cos \omega t \, dt \quad (3)$$

where $\omega = 2\pi \cdot$ frequency of the stored replica

In practice, a better "filter characteristic" can be obtained if a "weighting function" is applied to the signal. Hence, the above integrals may be modified to give:

$$O(t_1, \omega) = \int_{t-\Delta t}^t w(t) \cdot V(t) \cdot \cos \omega t \, dt$$

where $w(t)$ = weighting function. Typically, the form may be $w(t) = e^{-kt}$ (exponential weighting).

From the representative equations shown above, we may conclude that the basic problem in any approach to sonar signal processing is essentially one of designing a "computing" device to evaluate integrals of the form

$$\int_{t-\Delta t}^t V_1(t) \cdot V_2(t) \, dt \quad (4)$$

and to do so in real time.

2.2 BASIC OPTICAL PROCESSING SYSTEM

Figure 1 shows schematically the basic optical system which will demonstrate the principles underlying incoherent* optical processing. Its components are a light source, collimating lens, two moving film strips (optically transparent storage media), a condensing lens, and a photo-

* Incoherent, in this context, means that the process uses light intensity only, as distinguished from "coherent" processing where both light intensity and phase relationship must be used. Such a process would require a monochromatic light source.

detector tube that translates the output light intensity of the optical system into a usable electrical signal. Basically, this optical system is a computing device for evaluating integrals of the form given in equation (4).

Assuming that each film storage strip has uniform linear motion in the same direction and that the transmittance of any point on the strips is a controllable function of the incoming signal, the module's function as a "computer" is then as follows.

Light from the source is collimated by the first lens, passed through the two films in series, condensed by the second lens, and focused on the photodetector. The intensity of the light at any point on passage through the first film is $I_o \cdot T_1$, or the product of its original intensity (I_o) and the optical transmittance (T_1) of the film storage medium at the point under consideration. Similarly, the intensity of the light on passing through the second film is $I_o \cdot T_1 \cdot T_2$ (where T_2 = transmittance of a second strip) at any point. The system signal level output, then, may be considered to be the summation of the intensities of all the rays passing through the storage area or an integration across the entire optical plane. The equation representing this sequence is:

$$O = \int_0^x T_1 \cdot T_2 \cdot dx \quad (5)$$

If the transmittances are made linear functions of the input signals $V_1(t)$ and $V_2(t)$, then the optical system becomes a device for evaluating the basic signal-processing integral. That is, if

$$T_1 = a + b \cdot V_1(t) \quad (5a)$$

and
$$T_2 = a + b \cdot V_2(t) \quad (5b)$$

where a and b are constants, then (5) becomes (with a change of variable from x to t)

$$O(t) = \int_{t-\Delta t}^t [a + b \cdot V_1(t)] \cdot [a + b \cdot V_2(t)] dt \quad (5c)$$

which can be expanded to give

$$O(t) = a^2 \cdot \Delta t + ab \int_{t-\Delta t}^t [V_1(t) + V_2(t)] dt + b^2 \int_{t-\Delta t}^t V_1(t) \cdot V_2(t) dt \quad (5d)$$

Of the three terms on the right, the last is the one of interest. The first is a constant "bias" term, which represents a DC level in the output and, hence, is of no concern or importance. The second represents a "noise" in the output; provided the integration period is sufficiently large, it can be reduced to negligible levels as compared with the third term. Consequently, for all practical purposes, the output of the optical device is

$$O(t) = b^2 \int_{t-\Delta t}^t V_1(t) \cdot V_2(t) \cdot dt$$

Hence, an optical system that can store incoming signals in the proper form on optical transparencies can evaluate the necessary definite integrals with a modest amount of equipment. It is this simplification of equipment that makes optical systems attractive for sonar signal processing.

2.3 CONCEPTUAL SYSTEM APPLICATIONS

The systems applications discussed below are concepts for optically implementing the signal processing requirements of storage, multiplication, and integration based on the analogy derived above. The moving signal storage medium shown is photochromic material (described in detail in Section 3), whose characteristics and potential use in such applications are being explored in LEC research programs. Upon exposure to ultraviolet light, photochromic material undergoes an instantaneous change in color.

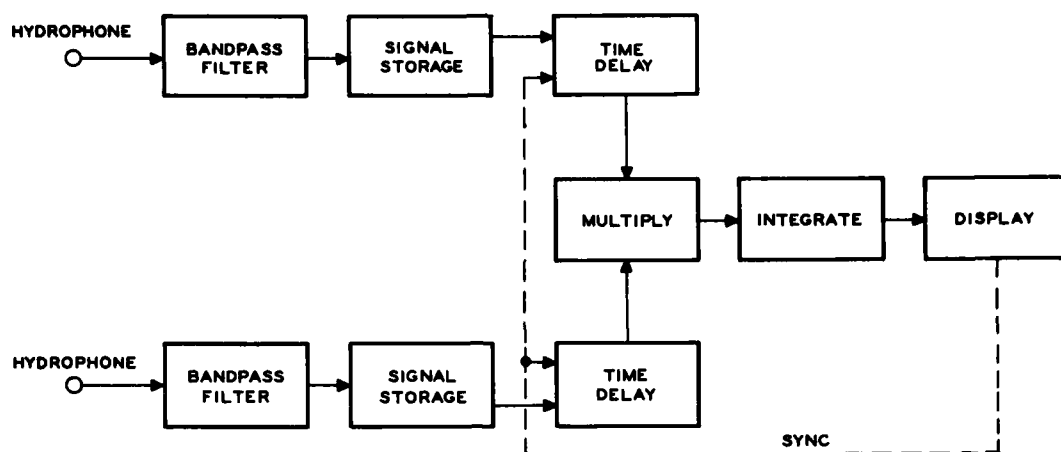


Figure 2. Two-Channel Correlator, Functional Block Diagram

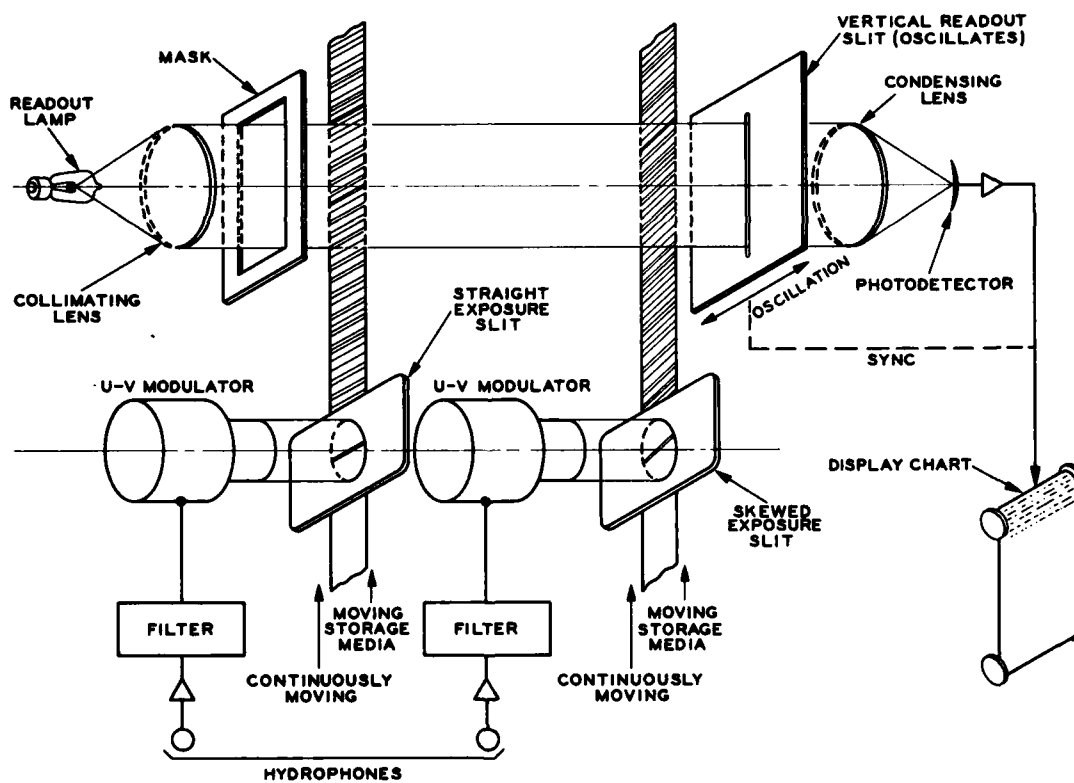


Figure 3. Two-Channel Correlator, Conceptual Schematic

Typically, the change is from colorless to a blue or purple. The optical transmittance in the colored state is approximately proportional to the ultraviolet exposure (time-intensity product) of the activating light source.

2.3.1 Passive System Concepts

Two-Channel Correlator. - With the basic principles of the computation sequence through optical means established, certain concepts for their application are evident. Figure 2, a block diagram of a two-channel correlator, shows the basic functional sequence of such a system. Signal filtering to retain only the band of interest and final display are common to electronic and optical processing alike. Hence, it is the functions of signal storage, multiplication, and integration that the optical system must be designed to handle. Figure 3 is a conceptual schematic for optically implementing this same correlation processing using the analogy described in paragraph 2.2.

Each incoming signal modulates a source of ultraviolet. Each of the film strips is exposed to one of the ultraviolet sources, the exposure taking place through a narrow slit in such fashion that the signal is stored as lines of varying transmittance. (Film strips are considered at this point for illustrative purposes only. They would not be used in a practical system, because of the problem of keeping them properly in register. Instead, rotating rigid disks or a single strip with signal recorded on each surface would be used, as will be discussed later.)

In order to develop time delay between the signals and to provide a means of sweeping through a range of time-delay values, the two recording slits are not aligned parallel to each other. The lines on one recording surface (representing stored signals) are not parallel to the lines on the other recording surface. On readout, the stored signal is scanned through a narrow slit that (1) is parallel to the direction of motion of the photochromic strip and (2) oscillates periodically in a direction at right angles to the motion of

the strip. Thus, at successive instants of time, the signal stored on one surface is correlated with varying values of time delay against the signal stored on the second surface. In order to present a display of output level versus time delay, the motion of the scanning slit is synchronized with whatever display device is used. Light passing through the two slits is focused on the photodetector, to obtain the integrating effect discussed previously.

Spectrum Analyzer, - The concept of a spectrum analyzer as a two-channel correlator is valid if the spectrum analysis is considered to be the correlation of an unknown incoming signal, recorded continuously on the first film, with a reference sinusoidal replica that can be swept in frequency through the band of interest.

Figures 4 and 5 show a functional block diagram and schematic representation of this process. The sinusoidal replica may be permanently stored on a glass plate. Readout, as above, is through an oscillating slit whose period of oscillation is synchronized with the time of signal storage and display.

Multi-Channel Correlators, - In order to achieve the combined advantages of the signal processing gain associated with the use of arrays and correlation techniques, LEC is conducting a preliminary investigation of concepts by which multi-channel correlation, such as is found in DIMUS and PADLOC systems, might be implemented. The functional sequence in such a correlator is shown in Figure 6. The problem, basically, is one of inserting the proper time delay into each of the signal channels so that a pair of preformed beams is formed and the signals received on each beam are in correlation. When this condition is satisfied, the combined effects of array gain and correlation-processing gain are realized.

The optical techniques that have been described offer a convenient means of developing the necessary time delay. As is true with magnetic recording drums, time delay can be developed between a pair of channels by varying

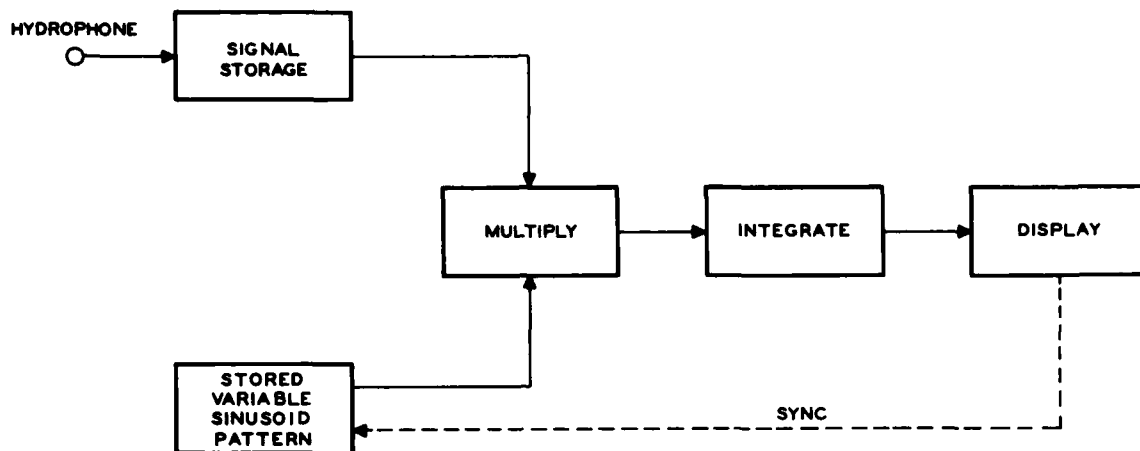


Figure 4. Spectrum Analyzer, Functional Block Diagram

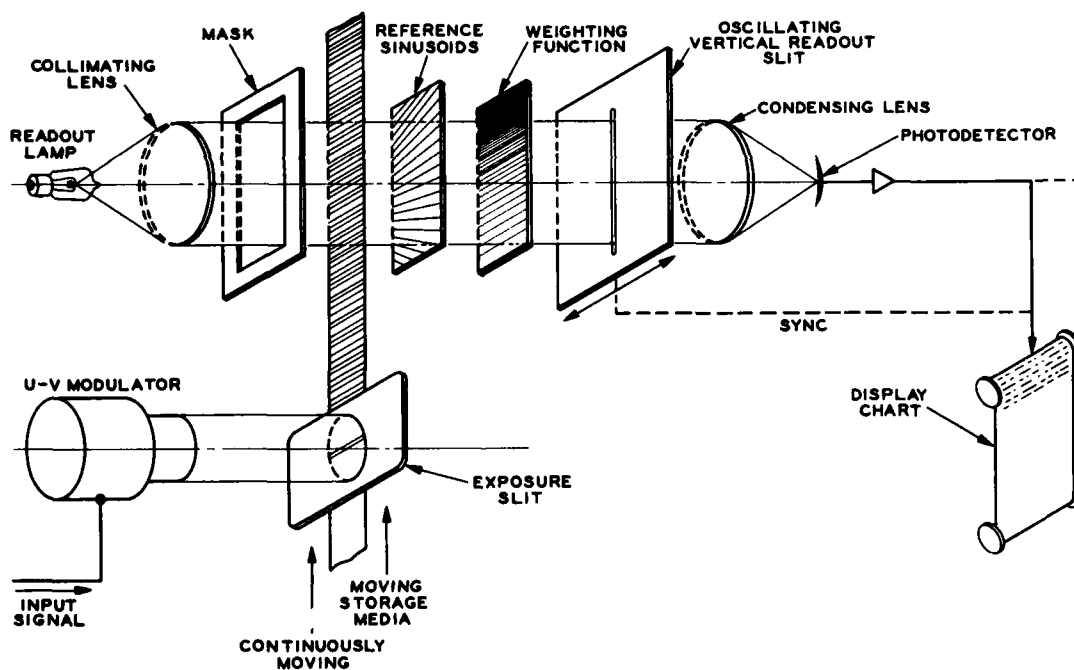


Figure 5. Spectrum Analyzer, Conceptual Schematic

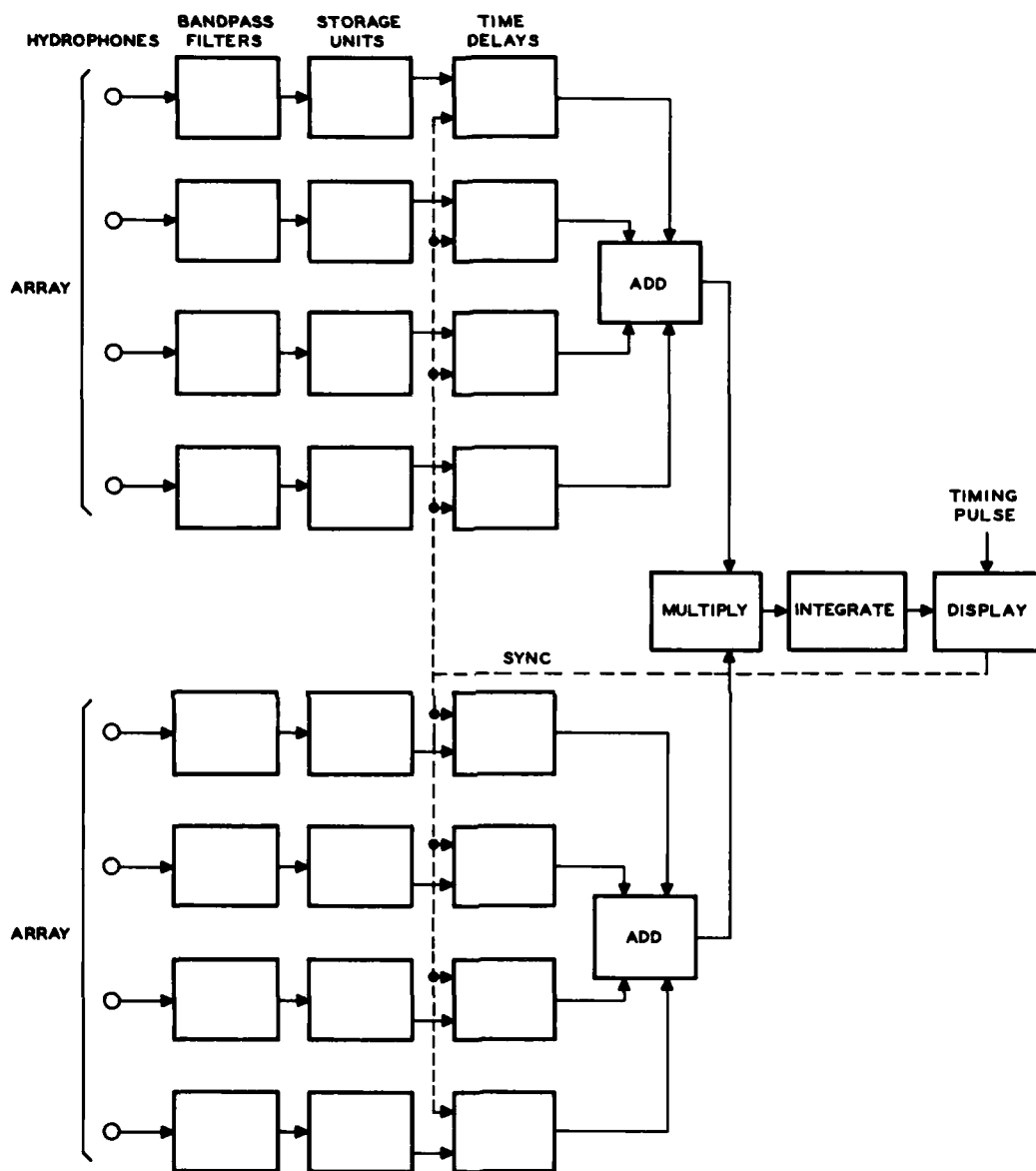


Figure 6. Multichannel Correlator, Functional Block Diagram

the position either at which the signal is stored or at which it is read out. For any technique, the time delay values must be correct to within a tolerance of about 3 percent of the period of the highest frequency in the system.

In any array correlator, information on the spatial distribution of array elements must be "stored" in the system in a form convenient for generation of the time delay. In a magnetic-drum system, this information is stored in the pattern of readout heads. In a DIMUS system, which uses shift registers for signal storage, the array geometry is "stored" in the pattern of collector buses that tap the signal from the shift registers. In the optical systems described here, the necessary information can be stored on photographic plates.

A preliminary study on the use of photographic plates for this purpose has been completed. A self-imposed requirement is that the system must be designed in such fashion that exposure and subsequent handling of the photographic plates can be carried out by U.S. Navy enlisted personnel aboard ship. Several alternative design approaches have been examined, each of which promises to be satisfactory. The choice of the best alternative depends, however, on the course of future developments, particularly on the development of ultraviolet light modulators. Consequently, it would be premature to discuss these design approaches at this time. It is anticipated that they will be covered in future reports.

2.3.2 Active System Concepts (Pulse Compression Sonar)

The term "pulse-compression" means the transmission of relatively long coded pulses by a sonar set, followed by "compression" of the echo returns into short pulses. The transmission of an "expanded" pulse results in a pulse of greater energy (average power) from a sonar set that is peak-power limited. Compression of the received echo results in good range resolution. The compression is accomplished by correlation processing, the return echo being correlated against a stored "replica" of the original pulse.

The ret effect is to put more total energy into the water and, hence, improve the system's inherent detection and target-resolution capabilities. Figures 7 and 8 are representative functional block diagrams of an active sonobuoy system and a scanning pulse-compression sonar system, respectively. Though the latter is inherently more complex in nature, each has essentially the same series of functional components.

The pulse compression feature in each is related to the "stored pulse pattern" which (1) identifies the transmitted pulse, and (2) is the reference against which the received echoes are correlated on return. In both applications optical implementation of the basic technique becomes a problem of developing and using this common optical replica.

Figure 9 illustrates one technique for optically implementing the pulse compression sonar concept. The transmitting sequence originates with the light source, shutter, and lens system shown at the top. Light energy passes through the moving shutter (focal plane shutter principle as used in photography) and then through the collimating lens and beam-splitter, respectively. That portion of the energy which passes through the beam-splitter is directed to a lens-slit-photodetector sequence; the output of which generates the range sweep on the range display. The energy that is deflected by the beam-splitter sweeps across the stored pulse pattern and passes through a condensing lens that focuses the energy on a photodetector. The output of the photodetector, in turn, keys the sonar transmitter sending out the "expanded" pulse.

The receiving sequence begins with the expanded return echo modulating a suitable ultraviolet source which records the signal on a moving photochromic film. The record processing area is illuminated with visible light of appropriate wavelength. When correlation of the return signal with the stored signal replica on the fixed plate is achieved, the peak of the "compressed" pulse appears at the photodetector output and on the range display.

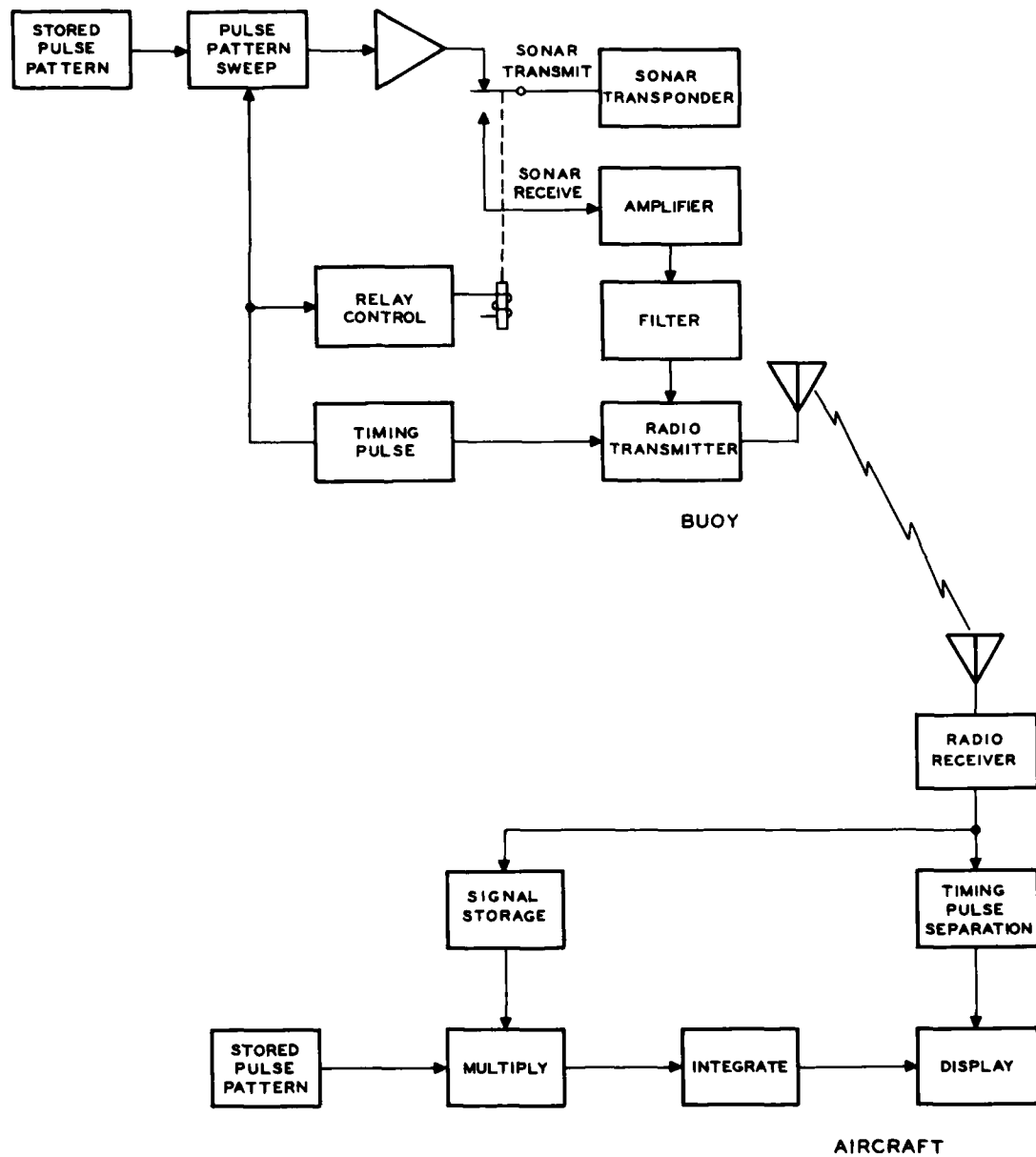


Figure 7. Pulse Compression Echo-Ranging Sonobuoy, Functional Block Diagram

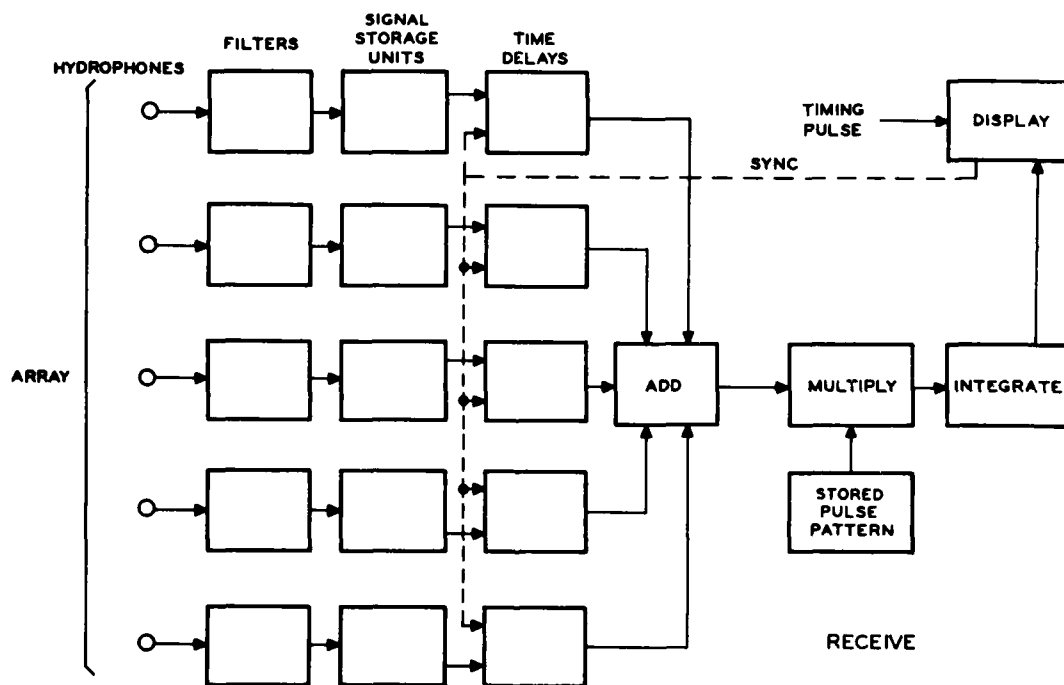
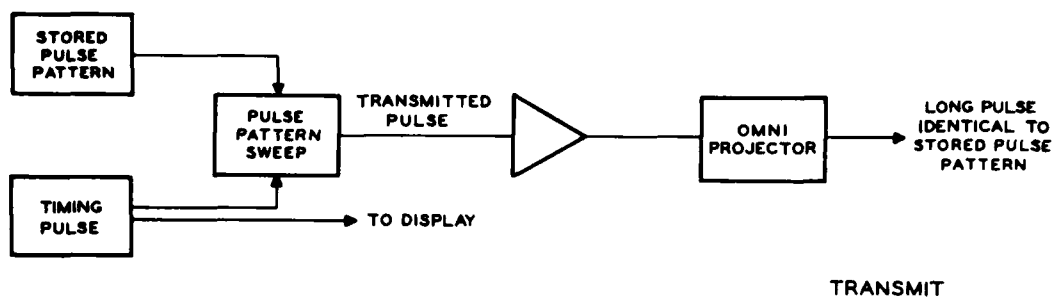


Figure 8. Pulse Compression Scanning Sonar, Functional Block Diagram

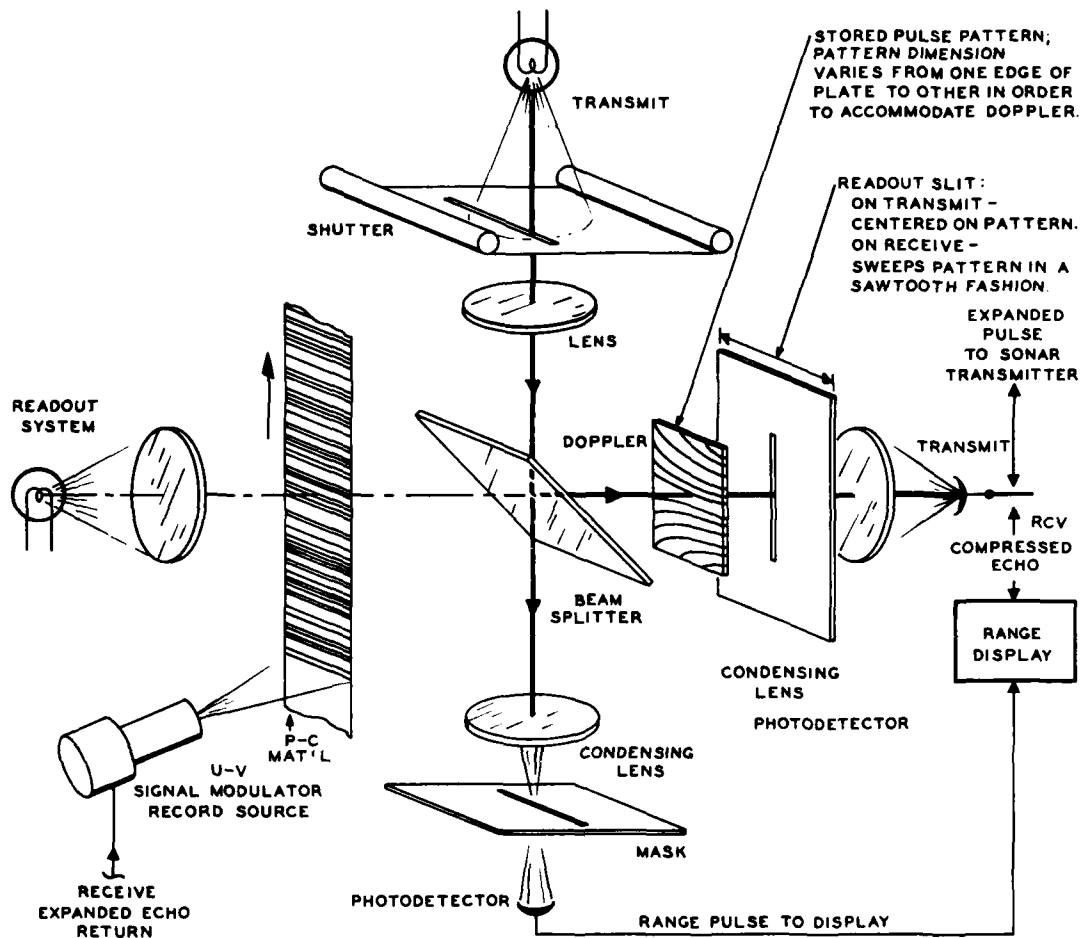


Figure 9. Pulse Compression Sonar, Conceptual Schematic

The effect, then, of this repeated correlation is one of "echo compression" or a continuous "piling up" of return echoes, correlated repetitively against the fixed pattern that established the character of the transmitted pulse.

Such a system must make provision for doppler. One approach to modifying the system to optically identify and determine the amount of doppler would be as follows:

- (1) Modify the stored replica of the pulse pattern so that the lines constructed upon it are increasingly divergent from their point of origin.
- (2) Install a readout mask with a vertical slip behind the replica, which remains centered during transmission.

In transmitting, then, the energy arriving at the condensing lens and photo-detector behind the mask would be limited to that portion which passed through the vertical slit on the mask. In the receiving mode, the return expanded echoes are again recorded on the moving photochromic film storage by the signal-modulated ultraviolet source. To effect correlation, however, the vertical slit is oscillated so that it sweeps in a sawtooth fashion across the replica. The presence and direction of doppler can then be displayed by indicating the relative position of this readout slit at the instant that correlation is achieved. For instance, if correlation is effected between the vertical axis and that side of the plate on which the lines are widely spaced, it is a "down doppler" situation. Correlation on the opposite side of the vertical axis would signify an "up doppler" situation. Correlation along the vertical axis would be interpreted as "no doppler." A further refinement of this concept would be the addition of a "range rate" display synchronized to the readout mask in the receiving sequence such that a direct readout of range rate (i. e. doppler) could be obtained.

2.4 CONCEPTUAL DIFFERENCES IN OPTICAL AND ELECTRONIC SIGNAL PROCESSING

From the foregoing discussion it can be concluded that certain basic differences exist in the philosophy of sonar signal processing using optical and electronic techniques. Electronic systems, by their nature, must read out (use) the stored signals in a sequential manner. In using these signals, time integration must be employed, the effect of which is to make "time compression" and "frequency multiplication" parameters of importance.

On the other hand, optical systems read out all stored information simultaneously; hence, they do not process the signal as a moving time series. Second, the integrator is an "area" related to time, not time as a moving series with which an integrator must be used. Finally, it is noted that the unique nature of optics is such that "time-compression" and "frequency multiplication" related to electronic storage techniques do not apply.

3. OPTICAL SIGNAL STORAGE

In order to process sonar signals optically in the manner previously described, a medium is required for signal storage. The most common medium being used is photographic film. A second medium is "thermoplastic" material. LEC, in its approach to the signal storage problem in optical processing, is using photochromic materials. A comparison of these three signal storage media is given below.

Photographic materials can and are being used in many signal processing applications. However, their use dictates certain definite requirements which in any photographic system cannot be circumvented:

- (a) They must be developed and fixed, which involves the use of wet chemicals.
- (b) They use expendable film which must be carried in quantity, creating a logistic problem.
- (c) In their storage and use, temperature controls may be required.
- (d) In any application the film must be handled in continuous strips with associated precise film transport requirements to minimize or eliminate associated problems of flutter, wow, etc.
- (e) The granular nature of photographic film may be a limiting factor in the degree of resolution that can be achieved.

Thermoplastic materials provide still another medium for optically re-recording signals. Their main disadvantage lies in the fact that imprinting on them by electronic beams must be done in a vacuum.

Photochromic materials are products of the analine dye industry. They can be applied in solution with transparent plastics as films on the surface of transparent materials. In their normal state they are colorless (transparent), in which state they are stable on exposure to most visible wavelengths of light. On exposure to ultraviolet light they switch from their colorless state to a blue-purple hue, in which state they are also stable and may be projected through with visible light. On exposure to infrared, heat, or visible light of the proper spectral wavelength, the material switches back to its original transparent state, and the cycle may be repeated.

Essentially, this interaction with light in the ultraviolet and infrared regions causes an alteration in the molecular structure of the photochromic substance such that it yields different absorption characteristics. (A theoretical discussion of photochromic phenomena is included in the Appendix.)

Photochromic materials avoid the difficulties associated with photographic and thermoplastic materials for the following reasons:

- (a) They are reusable.
- (b) They do not require chemical fixing.
- (c) They can be easily adapted to mechanical handling.
- (d) They require no unique environmental controls in storage, use, or processing; no vacuum devices are needed.
- (e) Their change in response to the application of energy (ultra-violet light) is molecular; hence, essentially infinite resolution is attainable.
- (f) The stored image appears instantly on exposure.

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From studies at LEC, it is clear that optical signal processing using photochromic materials as the signal storage media can be implemented with many advantages to the Navy. The following chapters discuss techniques of using these materials more fully.

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4. BASIC TECHNIQUES FOR USING PHOTOCHROMATIC MATERIALS IN OPTICAL PROCESSORS

LEC has experimented with two different techniques for use of photochromic materials in optical processors: (1) as coatings on (or in chemical combination with) transparent disks of suitable materials; or (2) as coatings on film strips.

4.1 DISKS

In the disk concept, the basic repetitive processing sequence of signal write-on, readout, and erase is accomplished near the circumference of the disk. Figure 10 is a schematic representation of the disk application. Signal write-on takes place by projecting the modulated ultraviolet light through a slit in a mask shown at the top of the disk. After passing through the signal processing area (represented here as a 90-degree sector of arc) the signal passes into the erase area where exposure to infrared (or heat) erases the signal and restores the disk to its original condition of opacity. It is then ready for reuse in the same sequence.

4.1.1 Two-Channel Correlator Application

Figure 11 is a schematic representation of the disk principle in a two-channel correlator application. Each disk continuously records the filtered modulated ultraviolet signals from each single channel. The skewed slit on the second disk is associated with the time delay generation technique described in paragraph 4.1.4. Readout is accomplished by projecting through the processing area with visible light of appropriate wavelength, collimated at its source, and condensed (focused) on the photodetector after passage through the signal storage disks. Hence, the requirements for signal storage multiplication and integration as discussed in paragraph 2.2, have been accomplished.

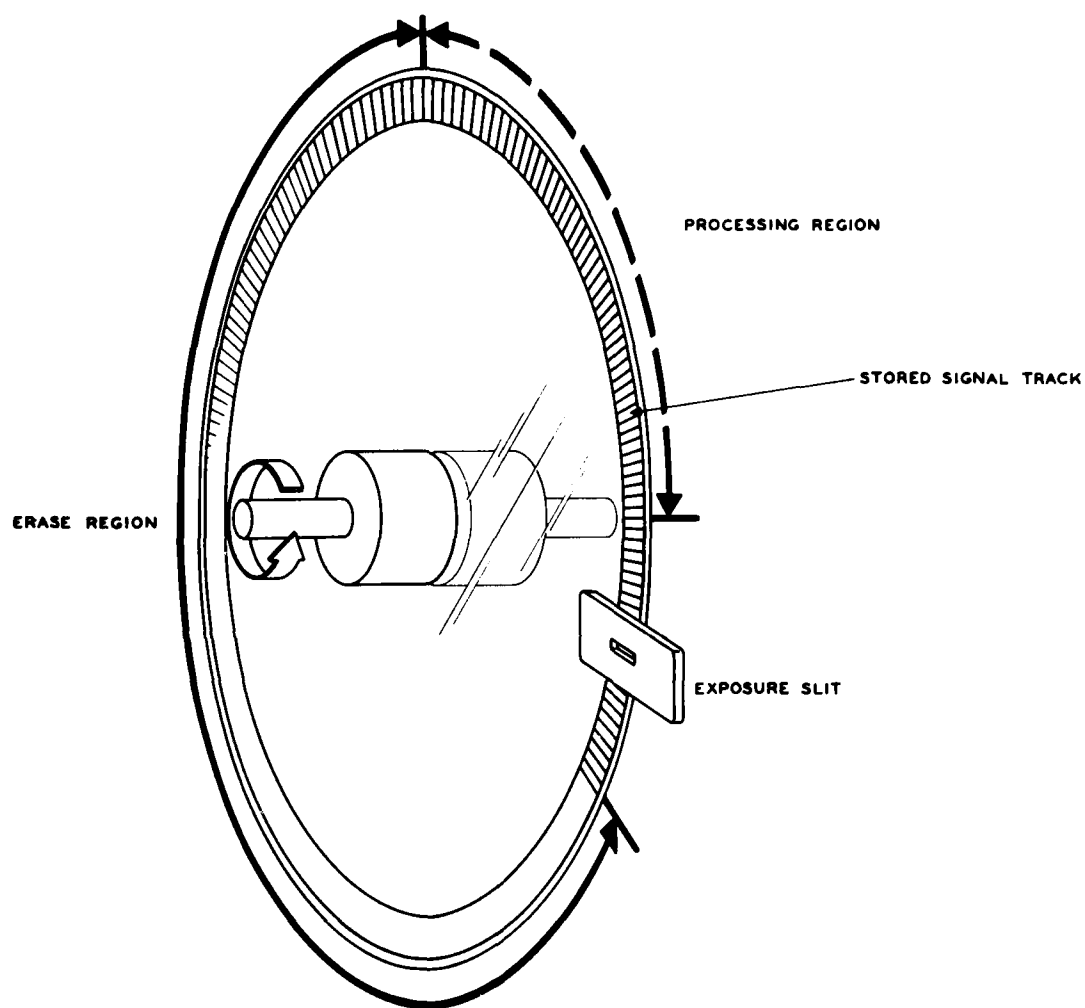


Figure 10. Photochromic Disk Processing Concept

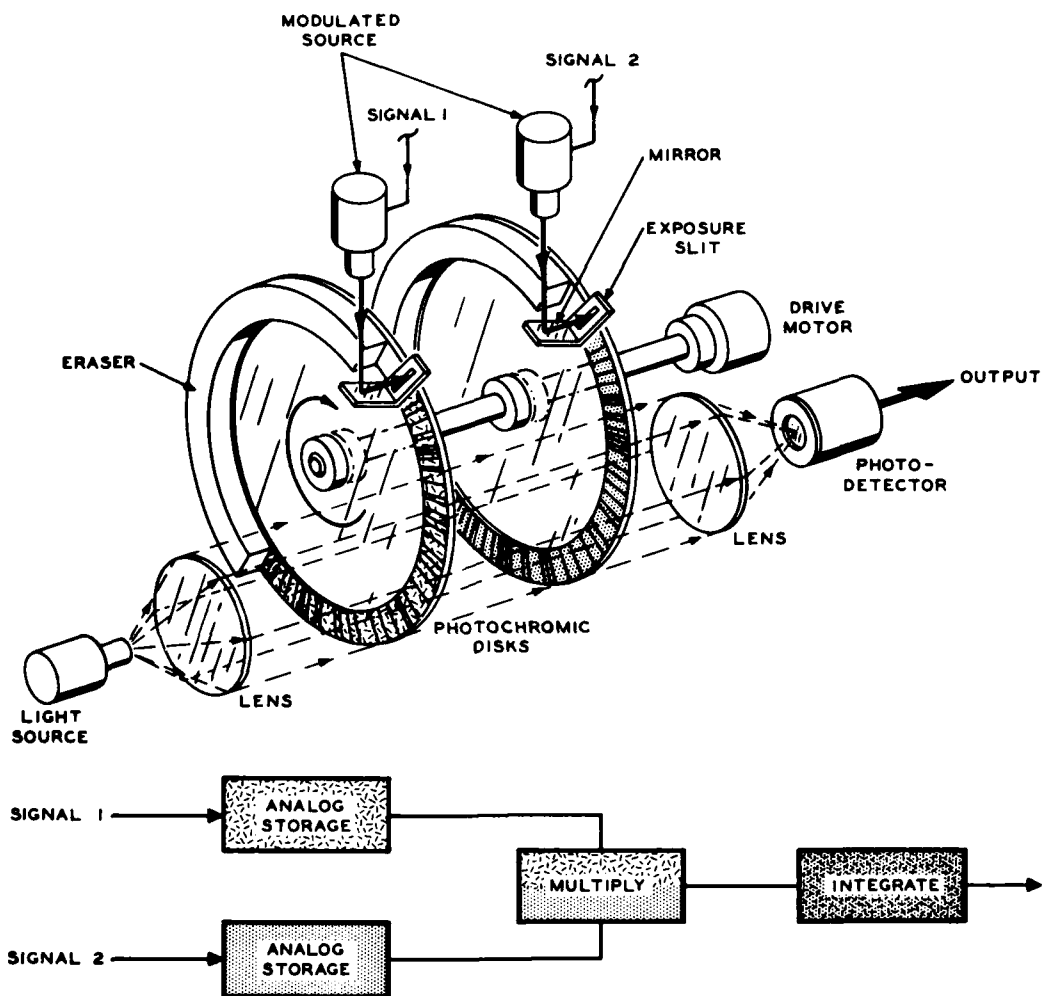


Figure 11. Two-Channel Correlator Using Photochromic Disks

4.1.2 Spectrum Analyzer Application

The two-disk principle may be applied to a spectrum analyzer if the spectrum analysis is envisioned as the correlation of an unknown signal against an appropriately "weighted" variable frequency sinusoidal reference. This sinusoid would take a permanent form built into the second disk. Weighting in this application might be optically implemented through appropriate shading (filtering) of the stored sinusoidal replica on disk no. 2.

4.1.3 Single Disk Concept

In either the two-channel correlator or spectrum analyzer application the alternative of using a single disk in lieu of two disks mounted on the same shaft is a possibility. Here both sides of the disk would be coated with the photochromic material, and the modulated signals would be recorded simultaneously on each side of the disk. If this approach proves feasible, greater equipment simplification will be realized.

4.1.4 Time Delay Techniques

Provision for the insertion of time delay is a requirement in any correlation processing, i. e., the system must provide for a varying time delay to compare (correlate) a signal with its stored replica, which arrived at some finite earlier time. One concept for implementing the effect of time delay in an optical system is shown in Figure 12. Here the stored signals on each disk take the same configuration as the slit in the mask through which they were passed in recording. The signals on the first disk are stored as segments of radii along the disk circumference. The signals on the second disk are stored as spiral segments. By generating an expanding arc of light that scans radially outward in a period at least equal to the total time of signal storage in the processing area, the effect of time delay has been created because:

- (a) As the arc passes the innermost end of the signals recorded on each disk, it is comparing information stored on disk no. 1 with information stored at some earlier time on disk no. 2. This might be thought of as a "negative" time delay situation.

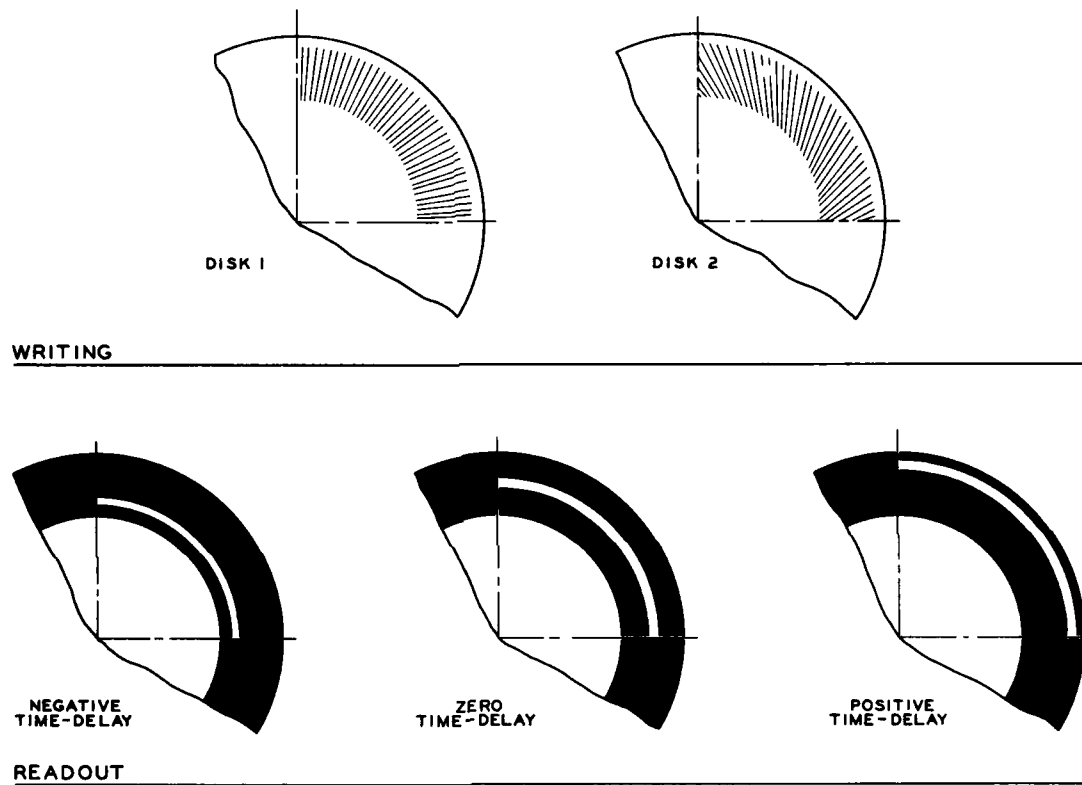


Figure 12. Time Delay Generation in Photochromic Disks

- (b) As the arc passes through the midpoint of its scan, it is comparing information recorded at the same time, hence, the same age. This could be considered a "zero" time-delay situation.
- (c) As the arc reaches its extreme outer position, it is scanning (comparing) information stored on disk no. 1 with information stored at some later time on disk no. 2. This then becomes a "positive" time-delay situation.

Hence, it may be concluded that the effect of variable time delay has been achieved within the optical system as long as the arc sweep period at least equals the time a signal is held in storage.

4.2 TAPE LOOPS

An alternative approach to using disks of photochromic material is that of using closed tape loops with photochromic materials coating the tape surface. Here, again, the processing sequence would be record, readout, erase, and reuse. For general correlation applications, either two tapes or a single tape with signal recording on both sides can be envisioned. The obvious drawback in the two-tape approach is the precise requirement for no relative movement between the two tape transports and the problems of flutter, wow, etc., which are inherent to some degree in even the most precise film transport equipments.

4.2.1 Two-Channel Correlators

In the application of closed tape loops to two-channel correlators, each signal-modulated ultraviolet source records on its tape through a slit (in a mask) whose axis is normal to the direction of motion of the film loop. Because the motion of each of the two films is linear (as opposed to rotational with disks) and is precisely controlled relative to the other, the requirement would be for vertical exposure slits only in each mask. Readout is accomplished with visible light of appropriate spectral characteristics through a

vertical slit in a mask whose period of oscillatory motion would be at least equal to the time of signal storage in the processing portion of the system. This mask movement is then the means by which variable time delay may be achieved in the system. The film then passes through the erase area where, by means of heat or infrared exposure, it returns to its transparent state and is readied for re-exposure.

4.2.2 Spectrum Analyzer

Again, in the concept of correlating an unknown signal against a built-in, stored reference, variable-frequency sinusoid (on the second tape), the spectrum analysis principle can be demonstrated using closed tape loops. The "weighting function" requirement for a filter to produce precise frequency response may be accomplished optically by design of a suitable graduated filter of varying degrees of shading.

4.2.3 Single-Tape Loop Concept

A photochromic film can be coated on each side of a tape that is transparent to visible light but opaque to ultraviolet. Consequently, the film transport problem of eliminating relative motion between two tapes is immediately removed, and the inherent problems of flutter, wow, etc., associated with film transports are at least halved, by the removal of one film transport.

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5. PROBLEM AREAS

There are several problems involved in implementing optical signal processing systems using photochromic materials. These can be grouped into two areas: problems associated with the materials themselves, and problems resulting from the use of these materials in signal processing systems. Each of these areas is discussed in the following paragraphs.

5.1 PHOTOCROMIC MATERIALS

Certain characteristics of photochromic materials relate to their use for signal storage in any optical system application:

1. Switching speed
2. Erasing speed
3. Fatigue
4. Optical noise
5. Resolution

5.1.1 Switching Speed

The rate at which the photochromic dye changes from the colorless to the excited colored state when activated by ultraviolet light, and the linearity of this change, will determine the frequency response of the recorder. It is important, therefore, that the relation between the switching speed and the ultraviolet exposure ($\text{watts/cm}^2/\text{sec}$) of various photochromic dyes of different optical path (dye concentration and path length) be investigated so that optimum systems can be resolved.

5.1.2 Erasing Speed

If the photochromic recorder is to provide a continuous and reusable media for recording and storing audio signals, it is equally important that the erasing effect be achieved in real time. The allowable erase time will vary according to the design of the recorder transport. Switching back to the transparent state can be accomplished by heat and/or illumination of the proper spectral distribution (500-600 mu). Thus, the rate at which this switching occurs for a particular dye as a function of the erase exposure (watts/cm²/sec) must be determined.

5.1.3 Fatigue

Fatigue is a gradual loss of the inherent switching property of a photochromic dye when exposed repetitively to ultraviolet radiation. The degree of fatigue is a function of the number of switchings and the intensity of the ultraviolet exposures that initiated the switchings. Increasing either exposure time or exposure intensity will not compensate for this decay. Since the operational life of a photochromic recorder system is limited by this basic dye characteristic, the causes of this fatigue are being investigated and remedies are being developed. Modifications to the basic dye structure can be effected with additives, which substantially reduce the fatigue rate without distracting from its forward switching rate.

5.1.4 Optical Noise

Noise results from random variations in the transmittance over the surface of a photochromic film caused by variations in the optical path or inhomogeneities in its structure. If a thin dye coating is placed on either a glass or mylar base, non-uniformities in the film thickness will result in variable changes in the transmittance over the surface when activated with ultraviolet. The net effect is a noise-like fluctuation in the intensity of light transmitted through the system. Consequently, the effects of fabrication techniques on noise are being investigated.

5.1.5 Resolution

Since the photochromic films consist of a molecular dispersion of reversible light-sensitive dyes in a suitable coating material, they are grain-free and hence are inherently high-resolution films. They are, therefore, superior in resolution to photographic materials. High resolution can be used to reduce the size of a signal recording system by working with suitable optics.

5.2 SYSTEM APPLICATION PROBLEMS

5.2.1 Signal Writing

Upper Frequency Limit. - The frequency response of a record system is influenced by the frequency-cutoff characteristic of the exposure slit as well as the translation speed of the record medium. Based on a sinusoidal input, the 3-db frequency response is given by the following expression:

$$f = \frac{0.443v}{d}$$

where

d = slit width

v = translation speed of record medium

The upper frequency limit will depend on:

- (1) Switching speed of the photochromic samples.
- (2) Ultraviolet intensity available at the exposure source.
- (3) Efficiency of the optics in obtaining a large value of irradiance (watt/cm²) at the exposure slit.

Thus, activating light sources of the required spectral output (300-400 mμ) must be evaluated with available photochromic materials to develop and refine the techniques of recording signals at an audio rate.

Effect of DC Bias. - All of the important signals stored and processed are pure AC with no DC component. Hence, to store them in optical form, a

DC bias must be added, corresponding to a medium-gray tone on the optical replica. This addition is necessary so that the excursion of electrical signals from negative to positive values may correspond to optical variations of transparent to opaque or vice versa.

Linear Range. - This characteristic of a particular dye system is determined by the relationship between the transmittance and the ultraviolet exposure time when a photochromic dye is activated by a constant source intensity. Although this is an exponential relationship, a region does exist where the change in transmittance approximates a linear function of the exposure time. This region then defines the "linear range" of that dye system and optical configuration.

5.2.2 Signal Readout

Since photochromic dyes absorb selectively in the visible spectrum when in the colored form, it follows that maximum readout contrast will be obtained through the use of filters matched to the absorption of the dye. The erasing tendency of the readout process makes it desirable to minimize the exposure of the photochromic sample during readout. Therefore, the readout light wavelength to be employed must be based on the absorption characteristics of the dye and must make use of high sensitivity photodetectors. Hence, it may be concluded that the requirements for the readout optics will vary with the dye system being used.

5.2.3 Signal Erase

Signal erasure can be obtained by the application of heat or visible light. Photochromic coatings that are relatively insensitive to erasure by visible light are called heat erasable. Coatings erasable by visible radiation are called light erasable, although they are also erasable by heat. The basic requirement of any erase technique is that it be able to "wipe the recorder clean" before it is exposed once again at the write-on slit. The speed at

which this can be accomplished is a function of the erase exposure (watts/cm²/sec) and will be a determining factor in the size and mechanics of the record transport.

5.2.4 Signal Modulation

Any optical processing system requires a suitable means of modulating the write-on signal at an audio rate. Two modulation techniques have been used to date with the mercury ultraviolet source lamps: (1) current modulation and (2) light valve techniques.

Using current modulation, mercury lamps cannot be modulated directly at frequencies above 3 kilocycles per second because, at the conditions of extreme brilliance of which they are capable, their luminosity does not fall rapidly enough when the current decreases to provide satisfactory modulation. Current modulation is a "brute force" technique that is relatively inefficient in terms of its power requirements. However, it has proven a convenient technique for use in the laboratory.

In the case of the "light valve," a Western Electric unit whose string tension was intentionally lowered has been used. Hence, its response was flat up to about 600 cps only. Increasing the string tension could extend this frequency up to about 8 kc, but at some sacrifice in the driving sensitivity with a gain-bandwidth exchange.

A third modulation technique being investigated by LEC is that of phase interference. The principles of this concept have been established in the successful design, building, and testing of a liquid ultrasonic light modulator using the same basic principles.

A fourth technique under consideration is a light valve using a piezoelectric bimorph bender as the active element.

A more detailed discussion of these techniques is included in Section 6.

6. LEC PROGRAMS IN PHOTOCHROMISM TO DATE

6.1 PHOTOCHROMIC PROPERTIES INVESTIGATION

LEC is pursuing a program of further study of those characteristic properties of photochromic dye materials that bear on their use in optical signal processing. These properties are persistence, fatigue, reversibility, resolution, and switching rate from one colored state to another under varying degrees of suitable light stimulation. The photochromic materials, which are being supplied by the American Cyanamid Company, are being subjected to (1) spectrophotometric analysis of their visible and ultraviolet absorption characteristics, and (2) measurement of the time rate-of-change of optical transparency in a photochromic system and its correlation with ultraviolet levels required to initiate the change.

The spectrophotometric analysis provides information of the type shown in Figure 13. The ordinate of the curves is percent transmittance, and the abscissa is light wavelength measured in millimicrons for a particular dye system in its colored and uncolored states. Two types of information can be extracted from such an analysis: first, the measure of the contrast between the colored and uncolored states; and, second, the peak absorption characteristics of the dye system under analysis. From this information the optimum readout wavelength requirement can be determined.

The results of this program are being correlated with the parallel study being conducted by the American Cyanamid Company.

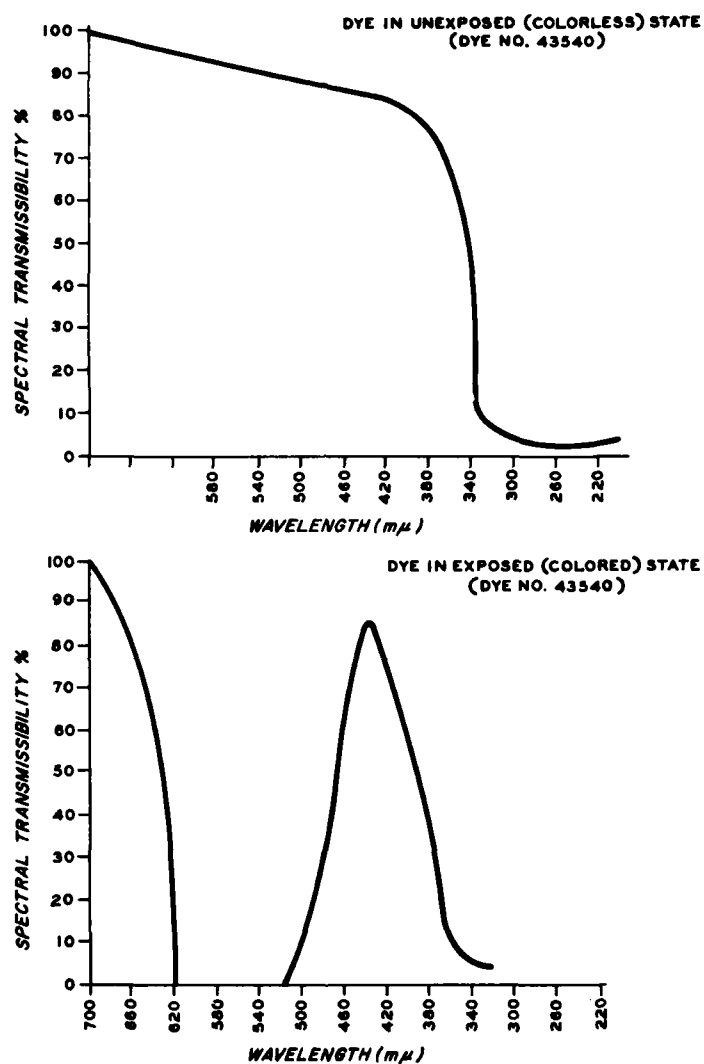


Figure 13. Spectrophotometric Analyses of Photochromic Dyes

6.2 SYSTEMS APPLICATION STUDIES

6.2.1 Signal Record and Readout

Various high-intensity ultraviolet light sources have been evaluated with fast-speed optical systems to determine a suitable activating source. This evaluation was accomplished by comparing the write-on speeds of several exposure sources with available photochromic materials. Short-arc mercury-vapor lamps have been found to be most efficient. Although it is most desirable to operate with low-power sources, switching speeds can be substantially improved by increasing the ultraviolet irradiance level (watts/cm²).

NOTE: An OSRAM type HB0200 W/2 lamp is currently being used as an ultraviolet light source. It has a typical operating rating of 53 volts, 3.6 amperes, and approximately 200 watts.

To measure the time rate-of-change of optical transparency in a particular photochromic dye system and correlate this with the ultraviolet level required to initiate the change, LEC designed and built an optical processor, shown in Figures 14 (schematic) and 15. With this processor, curves as shown in Figures 16 and 17 may be constructed wherein the effects of varying the optical path (dye concentration and/or path length) and ultraviolet irradiance (watts/cm²) can be observed.

Readout is obtained by passing a properly filtered light beam, which is matched to the maximum absorption characteristics of the dye, through the sample material onto a photomultiplier tube and observing the extent of the transmittance transformation on an oscilloscope.

6.2.2 Preliminary Signal Erase Investigations

Some success in erasing has been achieved by using projection lamps that have high spectral emittance in the visible and infrared regions. These

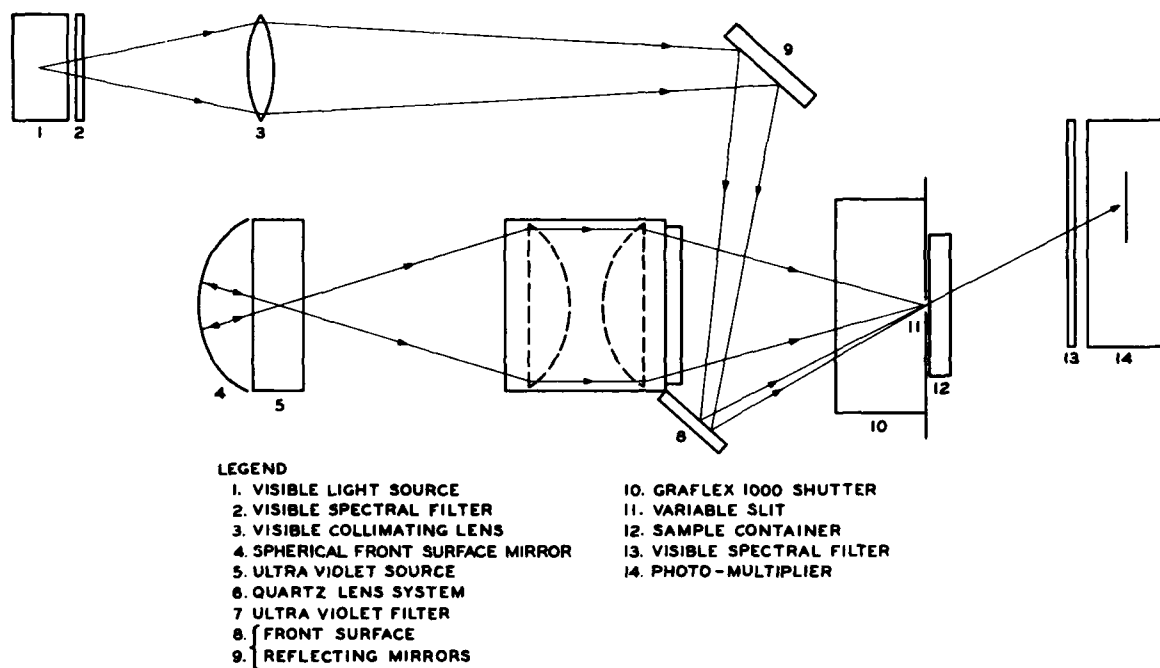


Figure 14. Optical Processor, Schematic



View
A



View
B

Figure 15. Optical Processor, Photograph

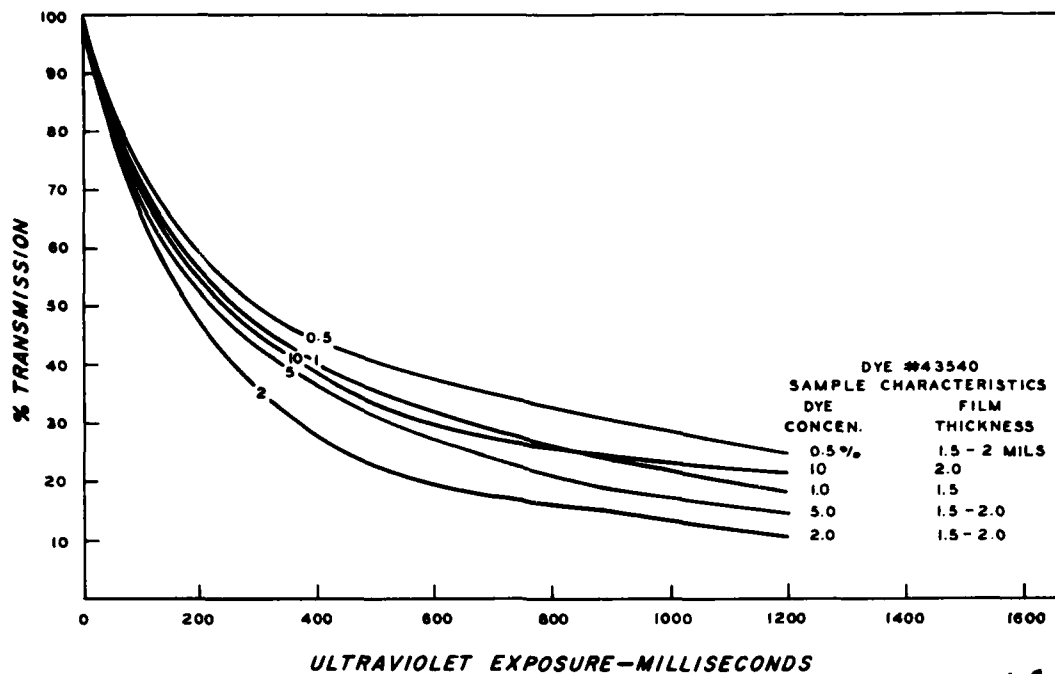


Figure 16. Photochromic Dye Concentrates vs. Percent Transmission

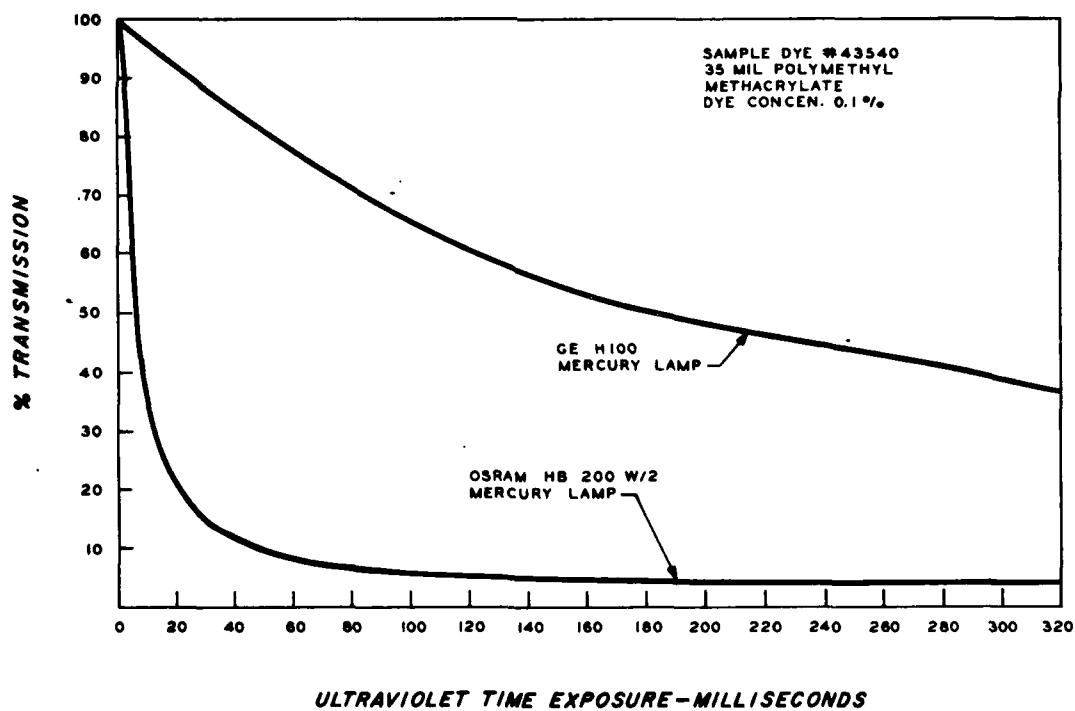


Figure 17. Percent Transmission vs. Ultraviolet Exposure Time

lamps have built-in reflectors within the glass envelope, which eliminate the requirement for external optics (lens and/or mirrors) to increase the lamp's inherent intensity. The techniques of erasing, as well as the exposure requirements of the erase source, are determined by the optical recorder design (disk or tape loop) and such considerations as the recorder speed (which, in turn, governs the amount of time available in the erase region) and the intensity of the erase source.

Obviously, light erasing is preferable to heat erasing, since residual heat in the photochromic material will affect its capability to store signals in such a repetitive process. Using 6-inch diameter disks, for instance, where 270 degrees of the outer circumference is devoted to the erase sequence, at least 2 seconds is required to completely erase signals recorded with the best dye system presently available using the standard projection lamp previously described.

6.2.3 Modulation Techniques

A description of the current and light valve modulation techniques which LEC has been using in its programs to date is given below.

Light Modulation by Current Variation. - In this method the mercury lamp used as an ultraviolet source is modulated directly at the source. The light intensity is varied by superimposition of audio-frequency currents on the DC quiescent current through the lamp. The power modulation that results is essentially proportional to the change in light intensity, as indicated by

$$\text{Percent of Modulation} = \frac{E_{\text{max}} I_{\text{max}} - E_{\text{min}} I_{\text{min}}}{E_o I_o} \times 100$$

Maximum and minimum values correspond to the extreme peak excursions, and $E_o I_o$ to the DC quiescent point. Since the mercury lamp is essentially a constant-voltage device, the light intensity is, to a first approximation, chiefly a function of the corresponding change of current through the arc.

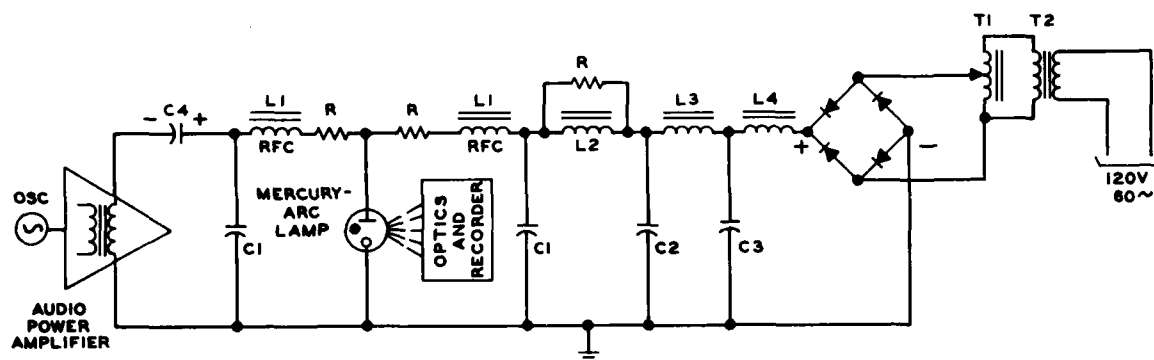


Figure 18. Current Modulation, Schematic

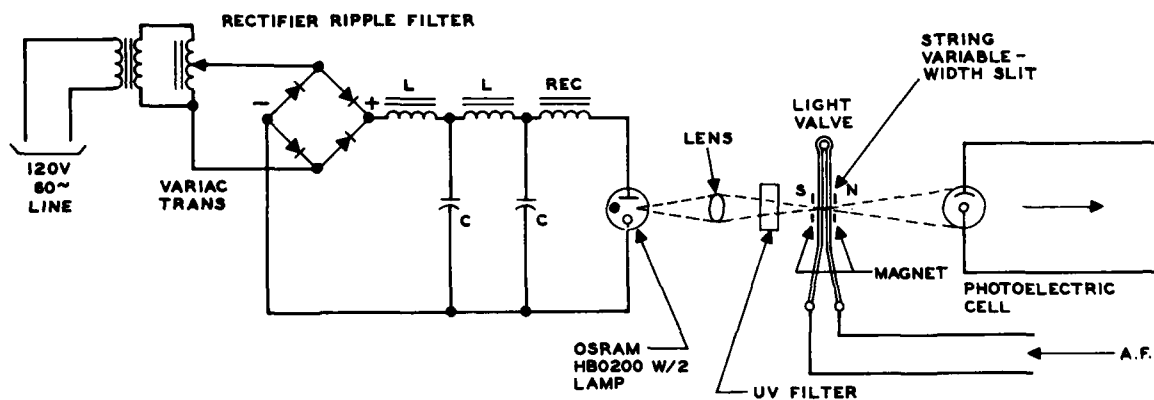


Figure 19. Light Valve Modulation, Schematic

The current modulation equals

$$\frac{I_{\max} - I_{\min}}{I_o} \times 100 \text{ percent}$$

Figure 18 shows a schematic for current-modulation of the mercury lamp. Direct current is supplied to the lamp from a rectifier well-filtered by L3, L4, and C2, C3. Audio-frequency signals are superimposed on the lamp from an oscillator and power amplifier combination. A large electrolytic capacitor, C4, isolates the DC from the amplifier, while L2 keeps the audio signal out of the power supply. L1, C1 prevent any stray RF, generated by possible negative resistance of the arc, from flowing in either the audio or power supply circuits.

Light Modulation by a Mechanical Gate (Light Valve). - A second technique of modulating the light intensity uses a light valve mechanically to gate the ultraviolet beam of light such as is used in recording sound tracks on motion picture film. The sequence is schematically shown in Figure 19. This technique employs a thin, flat duraluminum hairpin-ribbon stretched in the air gap of a strong magnetic field, the two parallel edges of which nearly touch. In the quiescent state, the ultraviolet light focused thereon is blocked off. When audio current is circulated in the string, motor action results in the opening and closing of a slit bounded by inner edges of the string. The frequency characteristics of the light valve are shown in Figure 20. Maximum modulating frequency with a modern light valve is about 8 kc. Approximately 60 percent modulation has been obtained in laboratory tests with this technique.

A second type of light valve, based on the use of piezoelectric materials, is under study. The use of high-compliance bimorph-benders shows promise of leading to an inexpensive, rugged light valve suitable for Naval shipboard use.

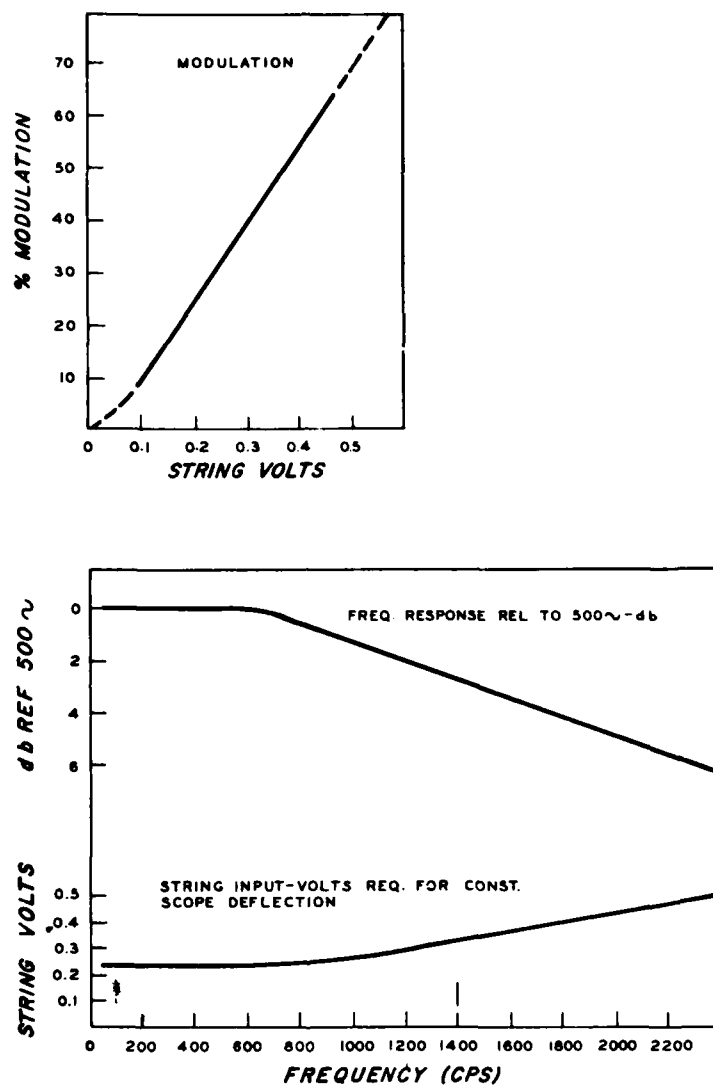


Figure 20. Frequency Characteristics of a Light Valve

Coherent Modulation Using Phase Interference. - Another possible technique of modulating the ultraviolet source is a mechanical-optical approach using the principle of phase interference. This principle can be implemented by inserting a quartz plate covered by a film of variable wavelength thickness in the ultraviolet beam of light. If the plate is mounted on a crystal transducer, vibrational motion of the transducer produced by the audio signal would, in turn, impart a like motion to the quartz plate and set up a phase interference of the light beam. Consequently, a modulated intensity would result at the exposure slit. LEC is further investigating this technique now.

6.2.4 System Noise and Frequency Response

A special experimental photochromic recorder has been designed and built to study system noise and frequency-response capabilities (see Figures 21 and 22). System noise is important because it determines the peak signal response of a recorder and, therefore, can limit its dynamic range. Frequency response depends on the characteristics of the material and on the ultraviolet exposure system. The faster the switching response of the system and the more intense the ultraviolet source, the higher the frequency response that can be attained.

Frequency response is measured by modulating the ultraviolet and imaging it through a slit onto the sample under study. The signal is then reproduced by illuminating through a second slit at some appropriate wavelength and detecting the signal on a phototube. The results are then correlated with the physical parameters of the sample under study. Present signal record and readout capabilities of this experimental recorder provide good system response up to 600 cps and peak response up to 1500 cps. The primary objective of tests now being conducted is to refine the details of recording until an audio signal can be printed on the photochromic disk, recovered at good signal-to-noise ratio, and then erased so that the sample is transparent once again by the time it arrives back at the record slit.

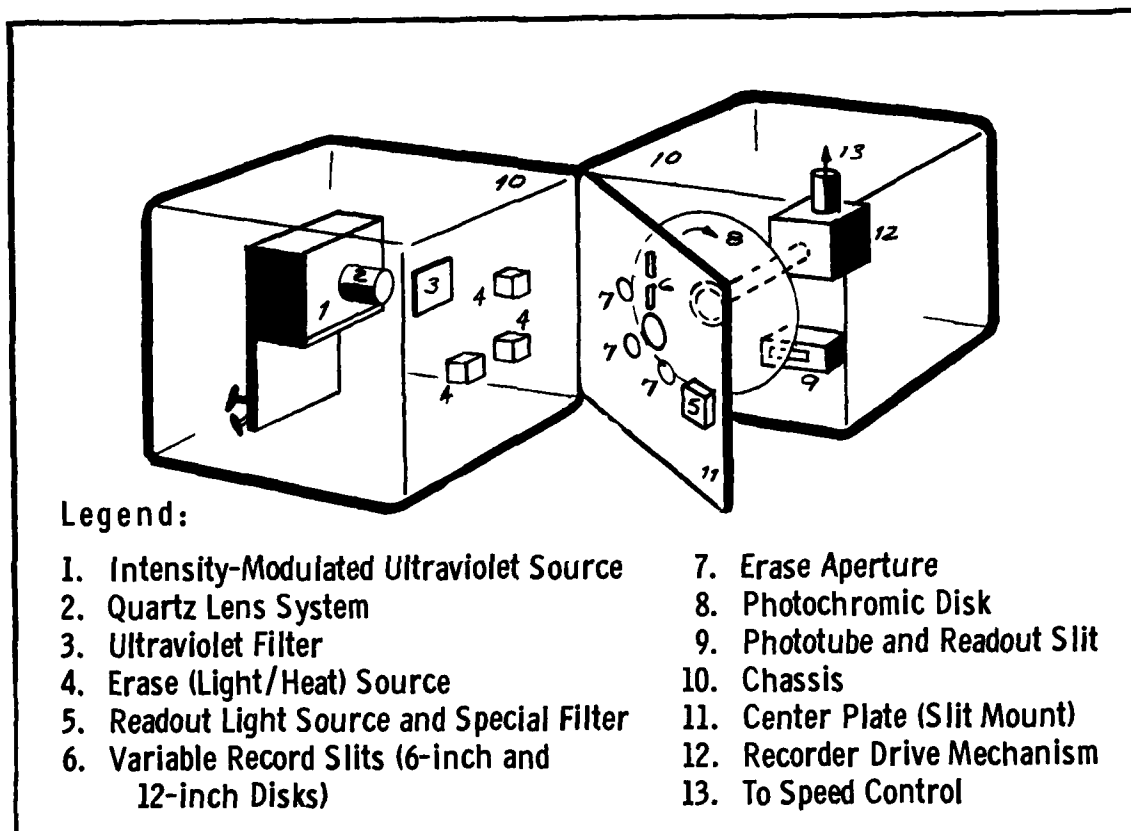


Figure 21. Photochromic Recorder, Schematic

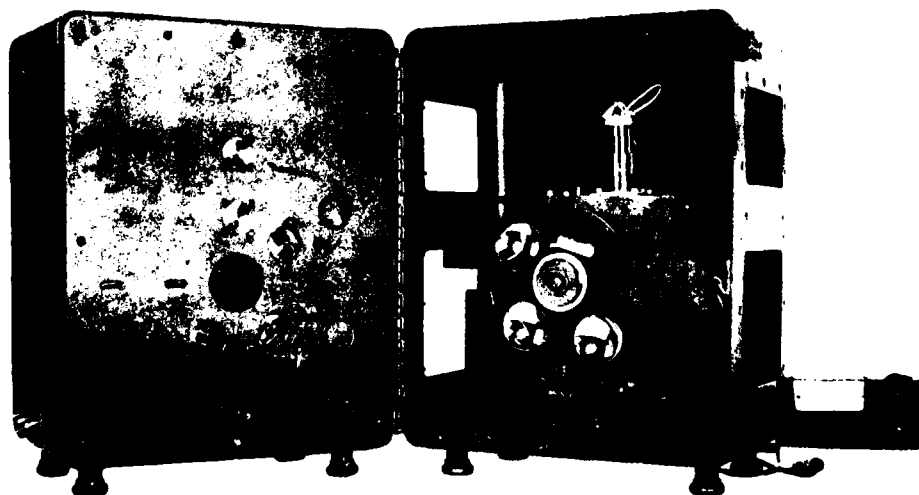
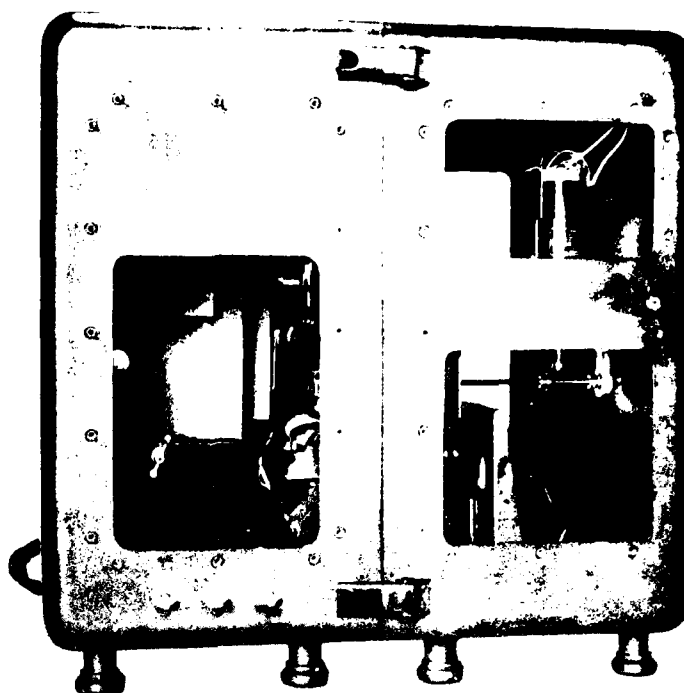


Figure 22. Photochromic Recorder, Photograph

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7. FUTURE LEC PROGRAMS

LEC programs in photochromic materials and their use in system applications fall into two general categories: (1) basic studies of the material properties, and (2) their use as signal storage media in specific system applications.

7.1 PHOTOCROMIC MATERIAL STUDIES

This program is a continuing one that essentially parallels the system application programs. The properties of new dye samples of varying chemical composition are being continually evaluated to ascertain their characteristics related to persistence, reversibility, resolution, switching time, and fatigue factor. Each of these obviously bears on the material's system use.

Fatigue of the material is associated with the number of "switchings" to which it is subjected. American Cyanamid Company advises that through the use of proper additives ("doping agents"), material life (i. e., resistance to fatigue) can be considerably increased once the best choice of dye sample for a particular system application is made. LEC believes the important immediate goal at this stage in the program is that of proving the feasibility of the photochromic technique for signal storage. Once this is established, a comprehensive program aimed at solving the material fatigue problem will be initiated.

7.2 SYSTEM APPLICATIONS STUDIES

The first step in this program was to design and build an optical bench photochromic recorder, essentially a breadboard model, so that system

noise and frequency-response capabilities could be studied. It is now being replaced by a much improved, newer version with which we will continue these same investigations. Concurrent programs aimed at developing the best time-delay generation and signal erase techniques are also in progress.

Once techniques for incorporating these requirements into a system are firmly established, LEC will design, build, and evaluate a two-channel optical correlator. When this correlator's capabilities have been satisfactorily demonstrated under laboratory conditions using artificial signal sources, LEC would hope to make similar tests under realistic "at sea" conditions. Hence, the optical correlator's performance could be compared with its electronic counterpart using the same input signal source.

Beyond this, LEC anticipates application of the same principles to the design and building of a spectrum analyzer, with laboratory and "at sea" testing to follow in that order, if possible. Once, this application is proven, the LEC program will investigate and develop active sonar system applications for this unique process.

From its considerable experience in pulse compression techniques, particularly in radars such as the AN/SPS-40 and AN/TPS-36 and a current ASW radar for NADC, LEC is convinced that these same principles can be applied to acoustic signals so that new active system concepts, such as high-data-rate sonars for fire control purposes, can be feasibly implemented.

8. CONCLUSION

From its programs and knowledge in the areas of photochromic materials, signal processing, system's application investigations, and pulse compression techniques, LEC has concluded that

- (1) optical techniques will play an important role in signal processing for certain present and many future ASW systems;
- (2) photochromic materials appear to offer a unique, yet simple, approach to optical processing with broad and practicable system implementation.

LEC will continue its work within the scope of these programs to demonstrate conclusively the feasibility of using photochromic materials in optical signal processing applications.

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APPENDIX A
TYPICAL PHOTOCHROMIC MATERIALS INVESTIGATION PROGRAM

The following section covering photochromic materials has been proposed by American Cyanamid Company.

Statement of the Problem

The incorporation of photochromic devices into the optical information processing systems of Lockheed Electronics Company will offer the advantages of great compactness of the system, great versatility in scanning rate and virtually unlimited resolution. We are convinced that the greatest promise for the development of suitable photochromic devices lies in the synthesis of novel photochromic compounds designed specifically for a specific end use. To pursue this approach on a rational basis certain basic information is required. This information includes a knowledge of the types of chemical structures which could exhibit the desired photochromic behavior, the influence of chemical structure on the kinetic processes involved, and the influence of environmental factors on these processes. It is also of importance to establish the kinetic feasibility requirements for a suitably operating device in terms of measurable fundamental parameters, e. g., the rate constants for the coloration and decoloration processes.

General Approach

On a previous contract with the U. S. Quartermaster Corps¹ a theoretical analysis was undertaken to explore the feasibility of using reversibly photo-bleaching dyes in combat clothing to protect the wearer from the effects of nuclear flash radiation. In that analysis, rate equations were derived to describe the flash response behavior of a model system and its behavior under normal sunlight exposure conditions. Those equations, with some modifications, are applicable to the problem considered here. In the initial

1. Weissbein, L., and O'Brien, S. J., "Attenuation of Thermal Radiation with Phototropic Colorants," Contract DA-129-QM-1438(01-9170), Project No. PO-8(7-12-01-002), Final Report, August 4, 1960.

phase of this project, we will use these modified equations to establish the kinetic feasibility requirements of a suitable photochromic device in terms of the measurable fundamental parameters.

The second phase, which will constitute a large part of this project, will be devoted to a kinetic study wherein flash spectroscopic techniques will be used to measure the coloration and decoloration rates, under controlled exposure conditions, of the most promising photochromic materials available. With these techniques the effect of environmental factors, e. g., temperature, polarity and viscosity of the medium, etc., on the rates of the coloration processes will be explored. The data obtained will be correlated with structural parameters to establish the direction in which any future synthetic program should proceed. The data will also be useful in establishing the mechanism of the particular photochromic phenomenon involved and in optimizing performance. In addition, the data will provide an opportunity for experimental verification of the theoretical rate equations.

Contingent upon the outcome of the previous studies, and provided that time permits, an effort to explore certain aspects of photo-decomposition is contemplated.

Theoretical Principles

The well accepted principles that are pertinent to any photochemical study have already been reviewed by Dryer.¹ Therefore, we shall restrict this discussion to principles which pertain specifically to photochromic phenomena.

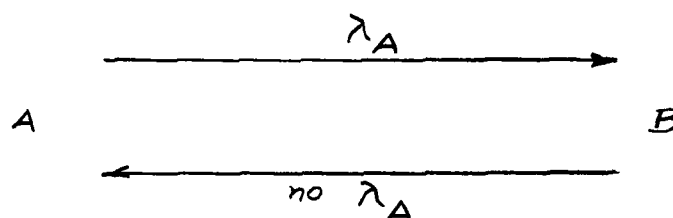
Since no universal definition of photochromism exists, let us formulate a provincial one that is satisfactory for our needs. Photochromism is herein taken to mean an appreciable change in the absorption spectrum of a system during the time it is subjected to "activating" electromagnetic radiation,

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1. Dryer, J. F., "Feasibility Study and Design of a Self-Attenuating Light Valve," WADC Technical Report 59-81, October, 1959.

with the absorption spectrum reverting to the original condition when the activating radiation is removed. Thus, this change is reversible.

The problem at hand calls for a photochromic system consisting of a clear plastic sheet with a photochromic material dispersed therein. It is imperative that this system be reasonably transparent in some region of the spectrum, but under illumination it must develop absorption in this region. It is also imperative the the resolution of the system be as high as possible.

Energy levels with separations in the visible and near ultraviolet regions of the spectrum usually are associated with "local systems," that is, units of molecular size and their immediate environment. Therefore, systems operating in these regions would be desirable from the standpoint of ultimate high resolution. Suppose we designate a "local system" in its normal colorless state by A and in its colored state by B. Then a functional definition of the photochromic phenomenon sought would be given by



The number of local systems which must be in state B to alter the absorption spectrum appreciably will depend in part on the extinction coefficients associated with B and hence is a specific property of each system. The number of local systems which can be placed in condition B by a given intensity of λ_A will depend on the number in condition A, the extinction coefficient of the local system in condition A for λ_A , and the lifetime of the system in condition B, the latter two factors again being specific properties. In order to proceed we must distinguish among various kinds of "local systems" and consider their possible utility. This involves consideration of the photochromic mechanisms.

a) Photochromic Mechanisms. - A major distinction between mechanisms for photochromism is whether or not "appreciable" atomic displacements are involved. (The term "appreciable" applies to displacements much larger than those associated with several quanta of vibrational energy.) In the latter category (designated i) color development relies primarily on electronic displacements, i. e., so-called "triplet-triplet" transitions in organic systems. In the former category (designated ii) there are more possibilities for color development, but perhaps at the expense of rate.

i. No Appreciable Atomic Displacement. - Under normal conditions of illumination in the visible or ultraviolet regions of the spectrum it is impossible to build up a sufficient concentration of local systems in the first excited singlet state which might then lead to an appreciable change in the absorption spectrum. However, under suitable flash conditions certain internal conversions may occur to such an appreciable extent that the system excited by λ_A is converted to another excited state with a longer lifetime. The states with longer lifetimes (metastable states) usually have this characteristic because they are the lowest energy states of a given multiplicity, and hence optical transitions to the ground state are forbidden. For stable organic compounds the ground state usually is a singlet and the metastable state is a triplet. Absorptions due to transitions from the lowest triplet state to higher triplet states (triplet-triplet transitions) are responsible for the color development.

The metastable states responsible for coloration are sometimes subject to other internal conversions which result in degrading of the energy to the ground state more rapidly than direct optical transitions. This type of radiationless decay is strongly influenced by environment. Normal ambient temperatures usually favor the rapid decay of the metastable states. Furthermore, systems in these metastable states are subject to chemical reactions with the environment.

ii. Appreciable Atomic Displacements. - We may envisage several distinct mechanisms within this category. In these cases the colored state B represents atoms in energy minima for which thermal energy (ambient

temperature) is necessary for reversion to state A. It is conceivable that in practice combinations of the various mechanisms listed below may occur.

Geometrical Rearrangement. - This is a situation in which the local system in state A undergoes a geometrical arrangement upon illumination, so that the system in state B is a stereoisomer with no change in sequence of atoms joined by chemical bonds. An example could be cis-trans isomerization, or it could be an excited electronic state with an appreciably different geometrical configuration from the ground state, and an environment which caused temporary trapping of the distorted configuration.

Bond Scission. - This is a case in which state B is obtained via an actual dissociation of one or more bonds of the system in state A. Chemically, this can be a homolytic or a heterolytic scission of a bond, with the fragments representing the system in state B. Certainly, the environment will exercise considerable influence on the lifetime and reversibility of state B. For example, if the bond broken was in a ring structure, the chances for recombination will be greatly increased.

Color formation through photoinduced salt isomerism, as with the leuco form of certain triphenylmethanes, is an example of the combination of geometric rearrangement and heterolytic bond scission. In this case λ_A activates the formation of a planar triphenylmethyl carbonium ion and the gegenion from the tetrahedral triphenylmethyl leuco compound. Here the polarity of the environment should influence both the charge separation and the life time of the triphenylmethyl carbonium ion.

Bond Association. - In this case λ_A promotes association of two parts of the local system which were not bonded in state A. This is somewhat the reverse of the previous case. In a sense it represents a trapping of an excited state by the environment, since an atomic displacement is involved as well as the formation of a new absorbing species.

b) Kinetic Analysis. - The feasibility of employing photochromic phenomena in optical data processing systems will depend largely on whether or not photochromic materials exist or can be synthesized with suitable switching times (coloration rates). In this connection, it is essential to define

"switching time" in terms of certain parameters, so that feasibility requirements can be expressed by related fundamental parameters. In the following discussion rate equations are derived with an aim toward defining switching time in terms of the rate constants associated with the $A \rightarrow B$ and $B \rightarrow A$ conversion processes.

In deriving these equations we shall consider a model system operating according to an idealized scheme which generally represents the mechanisms included in category ii. In this idealized scheme the thermal or photodegradation of the local systems is negligible. The $A \rightarrow B$ conversion process is activated with light contained in a specific band of radiation in the visible or near ultraviolet region. The $B \rightarrow A$ reversion process is thermally induced.

The energy diagram shown in Figure A-1 is a highly simplified picture of the photochromic mechanisms represented by our idealized scheme. In the figure, A is the colorless ground state of the local system with energy e_A . B is the colored, trapped state with energy $e_B > e_A$. B can be a trapped higher ground state or a trapped excited state. The quantity e_{AB}^* represents the energy B must attain in the process of reversion to the A state, i. e., the energy barrier. e_X is the excitation energy acquired by a local system when it is raised to the initial excited state by absorbing a quantum of activating radiation of wavelength λ_A . The initial excited state may be the first excited singlet state if no bond scission occurs or an unbonded state if bond scission is involved.

Photoexcitation of a single local system to the first excited singlet state is the primary step in the $A \rightarrow B$ conversion process and it occurs instantaneously. This process is immediately followed by collisional deactivation to the original A state or collisional deactivation to the B state, accompanied by atomic displacement and trapping. In the solid state at normal or elevated temperatures the probability is high that the deactivation collisions would occur within one or two vibrational periods of about 10^{-13} seconds after photoexcitation. If appreciable atomic displacement is involved, conversion to the B state could take considerably longer than this time. Even so, we

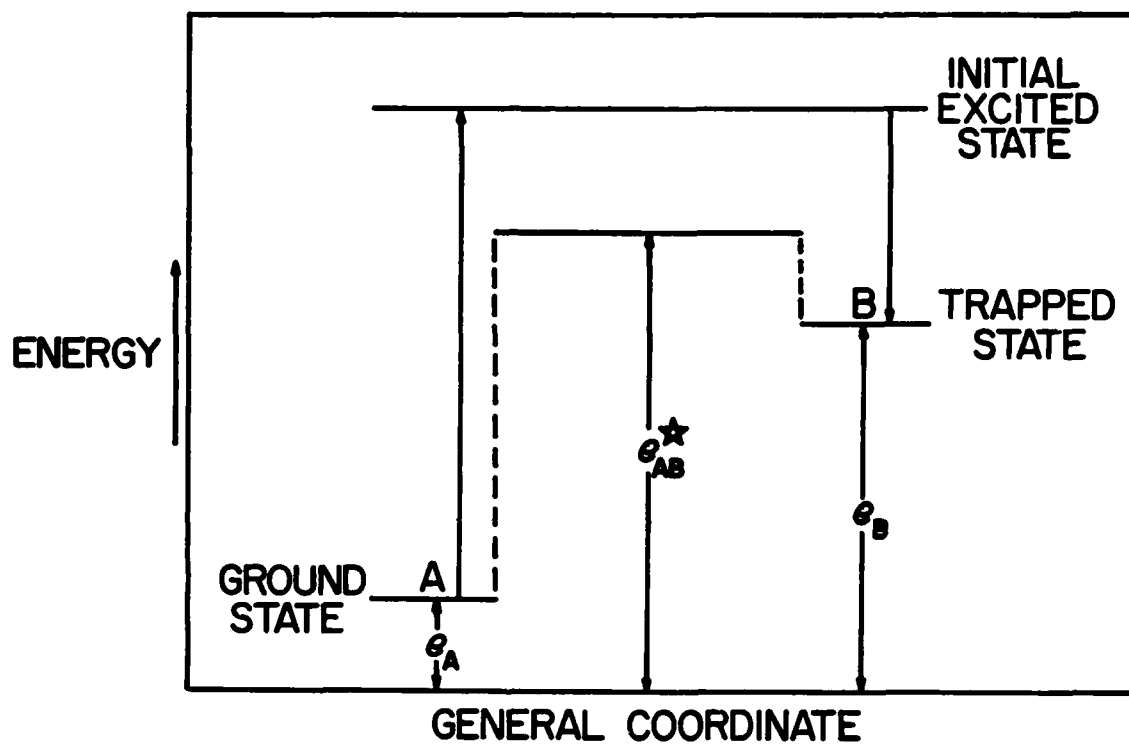


Fig. A-1 Energy Diagram of Phototropic Mechanisms Included in Category ii

can assume that the $A \rightarrow B$ conversion of a single local system will be virtually instantaneous relative to the time required to excite the assembly of systems. It follows then that the rate of $A \rightarrow B$ conversion process, taken by itself, is determined by the frequency of photoexcitations. The frequency of photoexcitations is a function of I_{λ_A} , the intensity of activating radiation of wavelength λ_A , and $[A]$, the concentration in moles per cm^2 of medium of thickness l of local systems in the A state at any given time of exposure.

The $B \rightarrow A$ reversion process, which is thermally induced, should follow first order kinetics if the rearrangement of a single species is involved, or higher order kinetics if more than one species is generated by the $A \rightarrow B$ process. For the sake of simplicity we will assume the $B \rightarrow A$ process to follow first order kinetics.

In this discussion we will consider a thin plastic sheet containing a total concentration of $[A]_0$ moles of local systems per cm^2 of the sheet having the thickness l . It is convenient to consider the integrated optical density developed (at the readout wavelength λ_R) by the $A \rightarrow B$ conversion of local systems through the entire thickness of the sheet, rather than that developed at each point through the thickness. If the local systems in the B state do not absorb at wavelength λ_A we could express the net rate of $A \rightarrow B$ conversion upon exposure to activating radiation simply by

$$d[B]/dt = k_1 I_{\lambda_A} [A] - k_2 [B] \quad (1)$$

where $[A]$ = concentration of local systems in the A state at any given exposure time in moles per cm^2 of exposed sheet of thickness l .

$[B]$ = concentration of local systems in the B state at any given exposure time in moles per cm^2 of exposed sheet of thickness l .

I_{λ_A} = intensity of incident activating radiation in einsteins per cm^2 per sec. This value could be a time dependent function.

t = exposure time in sec.

k_1 = rate constant of $A \rightarrow B$ process with dimensions cm^2 per einstein.

k_2 = rate constant of $B \rightarrow A$ process with dimensions sec^{-1} . This constant is temperature dependent.

The relation between $[A]$ and $[B]$ at any given time can be expressed by

$$[A]_0 = [A] + [B] \quad (2)$$

By combining equations (1) and (2) we obtain a rate expression in terms of the variable $[B]$:

$$d[B]/dt = k_1 I_{\lambda_A} ([A]_0 - [B]) - k_2 [B] \quad (3)$$

If the local systems in the B state absorb at λ_A , I_{λ_A} would be attenuated to some extent. Assuming that the light path is short, so that I_{λ_A} is only partially attenuated as it passes through the sheet, we could, as a first approximation, consider an effective radiation intensity I'_{λ_A} as being operative through the thickness of the sheet such that

$$I'_{\lambda_A} = I_{\lambda_A} \left[(\epsilon_{\lambda_A, A} [A]) / (\epsilon_{\lambda_A, A} [A] + \epsilon_{\lambda_A, B} [B]) \right] \quad (4)$$

where $\epsilon_{\lambda_A, A}$ = molar extinction coefficient of A at wavelength λ_A .

$\epsilon_{\lambda_A, B}$ = molar extinction coefficient of B at wavelength λ_A .

In that case, equation (3) becomes

$$\frac{d[B]}{dt} = \frac{k_1 I \epsilon_{\lambda_A, A} ([A]_0 - [B])^2}{\epsilon_{\lambda_A, A} ([A]_0 - [B]) + \epsilon_{\lambda_A, B} [B]} - k_2 [B] \quad (5)$$

The increase in concentration of B with time of exposure to activating radiation can be directly related to the increase in optical density of the sheet at the readout wavelength λ_R by the Beer-Lambert relationship given in the form

$$d[B]/dt = KdD_{\lambda_R}/dt \quad (6)$$

where D_{λ_R} = optical density (inverse of log % transmission) at wavelength λ_R .

$K = 1/\epsilon_{\lambda_R, B}$ in which $\epsilon_{\lambda_R, B}$ is the molar extinction coefficient of B at wavelength λ_R .

It is reasonable to assume that the normal ambient temperature will be maintained during irradiation with monochromatic activating or monochromatic readout radiation, so the k_2 can be taken as a constant. In that case, we can define switching time in terms of the rate constants k_1 and k_2 with the aid of a hypothetical experiment. Let us imagine that the plastic sheet is maintained in the dark until [B] and, therefore, D_{λ_R} are zero. At this time, which we will call t_0 , the sheet is subjected to activating radiation of constant intensity $I_{\lambda_A, 0}$ as shown in Figure A-2.

The response of the photochromic system contained by the sheet can be obtained by replacing I_{λ_A} with $I_{\lambda_A, 0}$ in equation (3) or (5) and solving the equation by integration between the limits $[B] = 0$ when $t = t_0$ and $[B] = [B]_t$ when $t = t_{>0}$. The solution in either case will be exponential in nature and have the general form shown in Figure A-2. The values of the extinction coefficients $\epsilon_{\lambda_A, A}$, $\epsilon_{\lambda_A, B}$, $\epsilon_{\lambda_R, B}$ for any given system can be measured spectroscopically in separate experiments.

The switching time can be thought of quantitatively as the time required for the development (with a given length if $I_{\lambda_A, 0}$) of a given value of D_{λ_R} in the hypothetical experiment described above. This requirement will no doubt depend upon the contemplated application. With this knowledge and a knowledge of $[A]_0$, which is limited by the solubility of the photochromic material

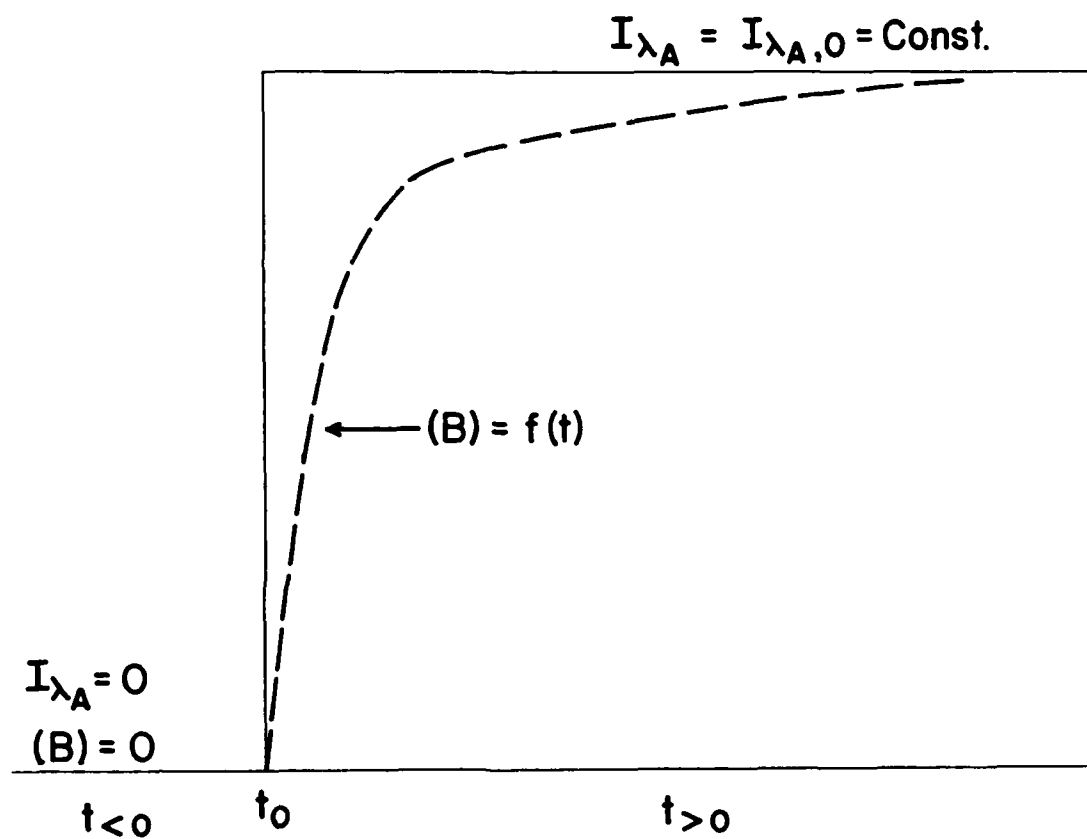


Fig. A-2 Schematic Representation of Switching Time Function

in the plastics medium, the aforementioned considerations would enable us to define the switching time requirement by certain required combinations of k_1 and k_2 .

The difficulties anticipated with the analytical approach outlined above are those that would be expected with any such approaches. These difficulties include the possibility of oversimplification in formulating the model system on which the basic equations are derived, making simplifying assumptions in manipulating or applying the equations which are not justified, etc. Attempts will be made, however, to verify experimentally the results of the analysis wherever possible and, perhaps, to modify the equations where such modification is indicated.

Flash Spectroscopy

The experimental approach will be based largely on the technique of flash excitation and absorption spectroscopy. This technique was first developed for the study of transient reaction intermediates and fast reactions by Norrish and Porter at the University of Cambridge in England. A schematic representation of the apparatus involved is shown in Figure A-3.

With this technique the photochromic system to be studied is activated by a short, intense burst of radiation. This burst of radiation is delivered by lamps composed of heavy metal electrodes set within a fused quartz jacket filled with an inert gas. The electrodes are energized by a bank of capacitors that delivers an electrical pulse at 5000 to 25,000 volts. Discharging in a few thousandths of a second, they can deliver power as high as a billion watts.

Synchronized with the large lamp, and set to fire after a short, predetermined interval, is a much weaker auxiliary flash lamp, usually designated as the spectro-flash lamp, which passes a light beam through the activated system. The beam then enters a spectrograph that records the absorption spectrum of the activated system on a photographic plate.

A series of such photographs not only identifies the absorption peaks of the colored species developed, but also indicates the rate at which they appear

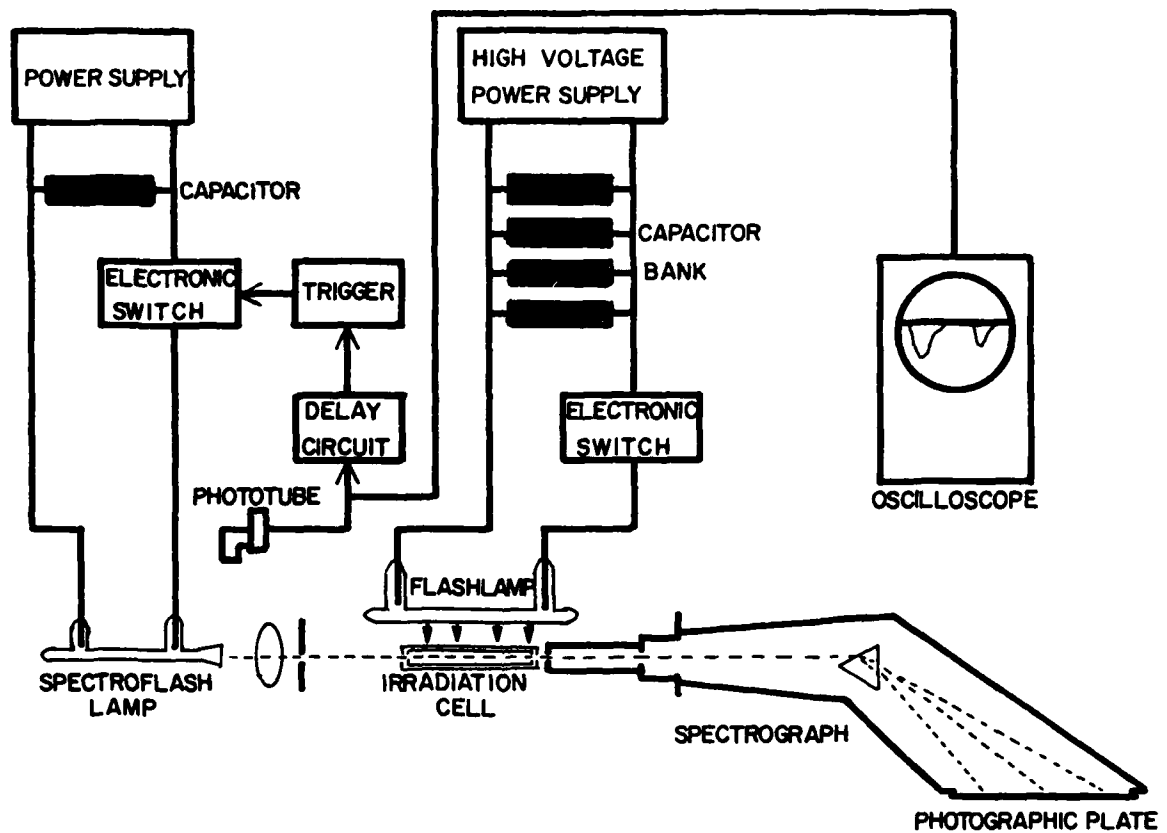


Fig. A-3. Schematic Representation of Flash Excitation and Absorption Spectroscopy Apparatus

and disappear. Once such an absorption peak has been located, its transient behavior can be tracked more accurately by replacing the auxiliary flash lamp with a small continuous lamp and the photographic plate with a photomultiplier tube set at the maximum wavelength of the absorption peak. The output of the photomultiplier tube is then fed to an oscilloscope, where it produces a graph showing the growth and disappearance of the absorption peak. In this way, the transient color produced by the flash is monitored continuously. The technique is quite sensitive. It can readily detect changes in light transmission as small as one part in 10,000. Technical details concerning the design and application of the flash spectroscopy apparatus appear in the literature.^{1, 2, 3}

With the flash spectroscopy equipment assembled at the Bound Brook Laboratories, absorption spectra and kinetic absorption measurements can be obtained in microseconds after the peak intensity of the initial flash is reached.

If the coloration process takes appreciably longer than a few microseconds, the coloration and decoloration cycle can easily be followed. If the color development occurs in less than a few microseconds, but the decoloration process is appreciably slower, then only the color decay rate would be measurable. In that event, or in the event that the color develops and is present only at the high light intensities generated in the initial flash, it would become necessary to modify the technique so that we could follow the color development during the flash. The problem here is to prevent the photomultiplier detector from becoming saturated with stray light from the flash. With suitable modification of the apparatus, this problem can be overcome.

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1. Grossweiner, L. I., and Matheson, M. S., J. Phys. Chem 61 1089 (1957).
 2. Grossweiner, L. I., Scientific American, p 135-145, May, 1960.
 3. Porter, G., Proc. Roy. Soc., A 200 284 (1950).

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Many of the presently known photochromic materials undergo some permanent change upon repeated exposure to ultraviolet radiation. In fact, it is axiomatic that any organic compound will experience some degree of permanent change or photodecomposition when exposed to ultraviolet radiation. However, certain dyes and ultraviolet absorbing compounds are known to exhibit remarkable stability, even upon prolonged exposure to ultraviolet light. On this basis, and on the basis of experimental findings thus far, it is expected that photochromic materials with sufficient stability can be developed.

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