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THE OHIO STATE UNIVERSITY, PH.D., 1979
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FOCUSING OF PHOTON ECHOES
IN SODIUM VAPOR

DISSERTATION

Presented in Partial Fulfillment of the Requirements for
the Degree Doctor of Philosophy in the Graduate
School of The Ohio State University

By

Neil Charles Griffen, B.S., M.S.

* * * * *

The Ohio State University
1979

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INTRODUCTION

Photon echoes are produced when an absorbing medium is subject to two collinear, non-synchronous pulses of monochromatic, coherent radiation of the proper intensity and duration. The emitted echo is coherent and collinear with the initial pulses. Figure 1 shows the sequence with two initial pulses and the resultant echo.

Spin echoes, the forerunner to the photon echo was first reported in nuclear hyperfine levels by Hahn in 1950. This phenomena was extended to the optical region by Abella, Kurnit, and Hartman in 1966 by observing photon echoes in a ruby crystal. Subsequently, photon echoes have been seen in many gaseous or vapor systems, including Cs, C\textsubscript{13}H\textsubscript{3}F, I\textsubscript{2}, SF\textsubscript{6}, and Na. The effects of a magnetic field and upper level lifetime measurements have also been studied.

In the experiments mentioned above, the initial pulses were plane waves as was the resultant echo. No attempt was made to vary the waveshape of the initial pulses and note the effect on the echoes. In 1973, Heer proposed that if the initial pulses had radii of curvature of $R_1$ and $R_2$, then the echo's radius of curvature could be calculated from $R_E^{-1} = 2R_2^{-1} - R_1^{-1}$. The analysis was extended to include Carr-Purcell Echoes in 1976. This experiment supports the theoretical work for the case of the photon echo.

Recently, there has been much interest in phase conjugation of
Figure 1. The Photon Echo Sequence.
coherent radiation by non-linear techniques.\textsuperscript{14} It has been observed in such media as CS\textsubscript{2},\textsuperscript{15,16,17} Na vapor,\textsuperscript{18} and Ge.\textsuperscript{19} As recently proposed\textsuperscript{20} and confirmed in this experiment, photon echoes can also be used to produce a phase conjugated beam.

The previous photon echo experiments conducted in this laboratory\textsuperscript{6,7,8} utilized a 3 meter sample cell which would be impractical for focusing experiments because the radii of curvature of the pulses would be tens of meters long in order for the source cell to appear "short." The high absorptivity of atomic vapors such as Na allow the cells to be much shorter and hence ideal for this type of experiment. Other reasons for using Na as the nonlinear media are the relatively low operating temperatures and the D lines are visible and at a wavelength compatible with Rhodamine 6G dye. This organic dye has high gain, is readily available, and is inexpensive.

Chapter I contains a general theoretical development for the production of focused photon echoes. From this basic development, other features such as phase conjugation, time of echo formation, and misalignment effects are discussed.

The experimental apparatus was designed and assembled for purposes of this experiment and is fully discussed in Chapters II and III. To produce the focused photon echoes, a monochromatic output from an optically pumped dye laser was amplified by a nitrogen pumped dye amplifier. These high intensity plane polarized pulses were then split
and recombined to form near collinear, nonsynchronous pulses. As these pulses passed through a sodium vapor cell, photon echoes were generated. These echoes were easily seen by the eye, but a high speed detector was needed to analyze the pulse shape and echo formation time. Focusing optics were introduced to allow the radii of curvature of the initial pulses to be independently varied. Circular polarization could also be produced by the use of a $\lambda/4$ plate.

In addition to the focusing data, an unexpected new phenomena occurred. When the beam attenuator which controlled the pulse intensity was removed, the resultant intense pulses created new pulses distinct from the echoes. These auxiliary pulses were coherent and had a well defined radii of curvature. The results of a simple experiment in which sum frequency generated pulses were formed is used to support an explanation for these auxiliary pulses.

Finally, the theory presented in Chapter I is extended to include damping effects by use of phenomenological equations. Using an IBM 360 Computer, the effects of off-resonant pulses and damping terms are calculated and compared with the results of Chapter I. A shift in the echo formation time which is a function of the radiative lifetimes is also predicted.
CHAPTER I

THEORY FOR THE FOCUSING OF PHOTON ECHOES

In this chapter, a general theoretical development from basic principles for the production of focused photon echoes is described. This development is similar to that used to explain the results of previous photon echo experiments in SF₆ and SiF₄ at this laboratory. For those experiments, the important parameters under investigation were the collision times and states of polarization of the plane wave pulses. In Na vapor, the collision times are too long to be significant, but the theory used to describe the polarization effects would be very similar to those already described and experimentally investigated. The development for these effects will not be repeated here but rather the effects of using spherical waves instead of plane waves will be presented.

A phase relationship between the echo and initial pulses is derived and used to predict the radii of curvature and direction of propagation of the echo as a function of these parameters in the initial pulses. In addition, the special case of phase conjugated echoes, the ideal electric field amplitudes of the initial pulses, the formation time of the echo, and the effects of non-collinearity of the initial pulses on the intensity of the echo are described.
General Considerations

The response of a 2-level degenerate atom or molecule to an intense pulse of monochromatic electromagnetic radiation can be described by the time dependent Schrödinger equation,

$$i\hbar \frac{\partial \psi}{\partial t} = [H_0 + V(t)] \psi.$$  \hspace{1cm} (1-1)

Using the dipole approximation, the interaction potential is

$$V(t) = -\hat{P} \cdot \hat{E}(\vec{r}_a, t)$$  \hspace{1cm} (1-2)

where $\hat{P}$ is the electric dipole operator, $\hat{E}(\vec{r}_a, t)$ is the electric field, and $\vec{r}_a$ locates the $a$th atom or molecule. The electric field can be expressed as

$$\hat{E}(\vec{r}_a, t) = E \hat{u} e^{i\phi(\vec{r}_a)} e^{-i\omega t} + \text{c.c.}$$  \hspace{1cm} (1-3)

where $E$ is the electric field amplitude, $\hat{u}$ is the polarization of the electric field at $\vec{r}_a$, and $\phi(\vec{r}_a)$ is the phase or spatial part of the eikonal. Using the rotating wave approximation and keeping only resonant terms with $|m_a\rangle$ representing the lower state and $|m_b\rangle$ the upper, $V(t)$ may be expressed as

$$V(t) = -\sum_{m_a, m_b} |m_a\rangle \langle m_b| \langle m_a| \hat{P} \cdot \hat{u} |m_b\rangle \hbar \hat{E} e^{i\phi(\vec{r}_a)} e^{-i\omega t} + \text{Hermitian Conjugate}. $$  \hspace{1cm} (1-4)
Similarly, $H_0$ may be expressed as

$$H_0 = \sum_{m_a} |m_a\rangle \langle m_a| + \sum_{m_b} |m_b\rangle \langle m_b|.$$  \hfill (1-5)

The eikonal can be removed from the expressions by using

$$\psi(\vec{r}, t) = A(t)\psi' (\vec{r}, t)$$  \hfill (1-6)

where

$$A(t) = \exp\left\{ \frac{1}{2} i \delta_0 [\phi(\vec{r}_0) - \omega t] \right\}$$  \hfill (1-7)

and

$$\delta_0 = \sum_{m_a} |m_a\rangle \langle m_a| - \sum_{m_b} |m_b\rangle \langle m_b|.$$  \hfill (1-8)

Substituting into Equation (1-1) gives

$$i\hbar \frac{\partial}{\partial t} \{A(t)\psi'\} = [H_0 + \mathcal{V}(t)]A(t)\psi'.$$

Multiplying on the left by $A^+(t)$ and rearranging yields

$$i\hbar \frac{\partial}{\partial t} \psi' = \left[ \frac{1}{2} \hbar \delta_0 \delta - \omega \right] + A^+(t)H_0 A(t) + A^+(t)\mathcal{V}(t)A(t)\psi'$$  \hfill (1-9)

$$= H_I \psi'.$$

Noting that $A(t)$ and $\delta_0$ commute with $H_0$, $H_I$ becomes

$$H_I = H_0 + \mathcal{V}_I + \frac{1}{2} \hbar \delta_0 \delta - \omega.$$  \hfill (1-10)
where
\[ V_I = A^+(t)V(t)A(t) \]
\[ = -\hbar E \sum_{m_a m_b} |m_a\rangle\langle m_b| (m_a |\vec{p} \cdot \vec{\mathbf{r}} |m_b) + \text{Hermitian Conjugate} \quad (1-11) \]

The time evolution operator, \( U' \), which describes the change in \( \psi' \) as time increases from \( t_n \) to \( t \) in the equation
\[ \psi'(t) = U'(t,t_n)\psi'(t_n) \quad (1-12) \]
is
\[ U'(t,t_n) = \exp\{-iH_I(t-t_n)/\hbar\} \quad (1-13) \]
and \( \phi \) is assumed constant. For \( U \) in the equation
\[ \psi(t) = U(t,t_n)\psi(t_n) \quad (1-14) \]
the time evolution operator is
\[ U(t,t_n) = A(t)\exp\{-iH_I(t-t_n)/\hbar\}A^+(t_n) \]
\[ = A(t) \exp\{-i\xi(t-t_n)\}A^+(t_n) \quad (1-15) \]
where
\[ \xi = (H_0 + V_I)/\hbar + \frac{1}{2} \delta_0(\phi - \omega) \quad (1-16) \]
and \( \xi \) can be diagonalized in some representation \( |\mu\rangle \).

The time evolution of the density matrix for the \( \alpha \)th atom or molecule from time \( t_0 \) is
\[ \sigma_\alpha(t) = U_\alpha(t,t_0)\sigma_\alpha(t_0)U_\alpha^+(t,t_0) \quad . \] (1-17)

The stimulated dipole moment is

\[ \vec{d}_\alpha = \text{Tr}\hat{\rho}\sigma_\alpha(t) \quad (1-18) \]

\[ = \sum_{m_a,m_b} (m_b | \hat{\rho} | m_a)(m_a | \sigma_\alpha(t) | m_b) + \text{c.c.} \]

and the total polarization is

\[ \hat{\rho} = N\vec{d} \quad (1-19) \]

where \( N \) is the number \( / m^3 \).

The electric field generated by an oscillating dipole at a distance \( r' \) away is

\[ \hat{E}_\alpha(\vec{r},t) = -\pi(\hat{r} \times \frac{\hat{p}_\alpha}{\varepsilon_0\lambda^2 r'}) e^{-i\omega(t-r'/c)} + \text{c.c.} \] (1-20)

where \( \hat{p}_\alpha \) is the coefficient of \( e^{-i\omega t} \) when Equation (1-18) is written out in detail. \( \hat{r}' \) is related to \( \hat{r} \) by

\[ \hat{r}' = \hat{r} - \hat{r}_\alpha \quad (1-21) \]

Each atom or molecule will then generate a spherical wave which when summed over all the atoms create a new wavefront with phase \( \phi_\rho(\vec{r}) - \omega t \). The sum over the individual field amplitudes from each
source is proportional to

\[ F = \sum \sum \alpha \beta a_{a}b \exp \{-i[\phi_{E}(\vec{r}) - \omega t]\} \]  \hspace{1cm} (1-22)

Before the atoms are exposed to the radiation, the density matrix consists of equally probable lower states \( |m_b\rangle \),

\[ \sigma(t) = (2J_b + 1)^{-1} |m_b\rangle (m_b| . \]  \hspace{1cm} (1-23)

When the pulse is suddenly turned on at \( t_0 \), the density matrix takes the form

\[ \sigma_{ab}(t) = (m_a|U(t,t_0)\sigma(t_0)U_{1\dagger}(t,t_0)|m_b) \]  \hspace{1cm} (1-24)

\[ = (m_a|\exp[-i\xi(t-t_0)]|m_b)(m_b| \exp[i\xi(t-t_0)]|m_b) \times \exp[i\phi(\vec{r}_a)-\omega t] \]

\[ = -g_1 f_1 \exp[i\phi(\vec{r}_a)-\omega t] \]  \hspace{1cm} (1-25)

where \( f_1 \) and \( g_1 \) are the matrix elements of \( \exp[i\xi(t-t_0)] \). These elements are listed below, and detailed calculations can be found in Reference 21:

\[ f = \cos qt - i \frac{\Delta}{2q} \sin qt \]  \hspace{1cm} (1-26)

\[ g = \frac{i v}{q} \left(J_l m_b M|J m_a \right) \sin qt \]  \hspace{1cm} (1-27)
11

\[ 2q = [\Delta^2 + 4|v|^2 (J_b \cdot M_a - J_b \cdot m_a)^2]^{1/2} \]  

\[ v = \pi^{-1/2} (2J_a + 1)^{-1/2} E(J_a ||P|| J_b) . \]  

In the above equations, \((J_b \cdot M_a | J_b \cdot m_a)\) is the Clebsch-Gordan coefficient and \((J_a ||P|| J_b)\) is the reduced matrix element in the Wigner-Eckart theorem. Also, it was assumed that the perturbation connects only the state \(|m_a\rangle\) with \(|m_b\rangle\). Thus, for linear polarization \(m_a = m_b\) and for circular polarization \(m_a = m_b \pm 1\).

After the pulse has been turned off at \(t_1\), the system continues to evolve according to

\[ U_0(t, t_1) = e^{-iH_0(t-t_1)/\hbar} \]  

and

\[ \sigma_{ab}(t) = (-g_1 f_1) \exp\{i[\phi_1(\vec{r}_a) - \omega_1 t_1]\} \exp[-i\omega_{ab}(t-t_1)] \]  

\[ \text{(1-31)} \]

where \(\omega_1\) is the pulse frequency. Also by Equation (1-25)

\[ \sigma_{ba}(t) = \sigma_{ab}^*(t) \]  

\[ \text{(1-32)} \]

for \(t_1 \leq t \leq t_2\).

The density matrix following the second pulse which starts at \(t_2\) and ends at \(t_3\) is

\[ \sigma_{ab}(t) = (m_a \lvert U_0(t, t_3) U_2(t_3, t_2) \rvert m_b) (m_b \lvert \sigma(t_2) \rvert m_a) \]
\[ \times \langle m_a | U_2^+(t_3, t_2) U_0^+(t, t_3) | m_b \rangle \]
\[ = \sigma_{ba}(t_2) g_2^2 \exp\{i[\phi_2(t_3) + \phi_2(t_2)] - \omega(t_3 + t_2) - \omega_{ab}(t-t_3)\} \] + non-echo terms \hspace{1cm} (1-33)

In this system, the atoms or molecules are moving which requires that \( \phi \) be replaced with
\[ \phi[\vec{r}(t)] = \phi_0[\vec{r}(0)] + \dot{\phi} t . \] (1-34)

Substituting Equations (1-31) through (1-34) into Equation (1-22), yields
\[ F_\alpha = -g_1 \frac{g_2}{f_1} e^{i(\phi_E - 2\phi_2 + \phi_1)} e^{-i\Delta(t-t_3-t_2+t_1)} \] (1-35)

and the pulses are assumed to have the same frequency. Also, the assumption that \( \phi_1 = \phi_2 = \phi_E \) was made. When the sum over \( \alpha \) is taken, then two conditions must be met in order for the sum to be non-zero. The phase term \( \phi_E - 2\phi_2 + \phi_1 \) must not be allowed to vary which requires
\[ \phi_E - 2\phi_2 + \phi_1 = \text{constant} \] (1-36)

Also, when the sum over different molecules is taken, the \( \Delta \) term will vary according to the doppler shift of the particular molecule. Thus,
for the last exponential term in Equation (1-35), the time variable must be such that
\[ t = t_3 + t_2 - t_1. \]  (1-37)

Focusing of the Echo

When the gradient of Equation (1-36) is divided by k, the result is
\[ \hat{n}_E = 2\hat{n}_2 - \hat{n}_1 \]  (1-38)
where \( \hat{n} \) is the direction of the normal to the wavefront. If \( \phi = \hat{k} \cdot \hat{r} \), corresponding to a plane wave, then Equation (1-38) becomes
\[ \hat{k}_E = 2\hat{k}_2 - \hat{k}_1 \]  (1-39)

The phase of a spherical wave with its real or virtual source at \((x_0, y_0, z_0)\) and at large \( r \) can be expressed as
\[ \phi = kr \pm k(z-z_0) + \frac{k}{2(z-z_0)} \left\{ (x-x_0)^2 + (y-y_0)^2 \right\} \]  (1-40)
which is valid for the wave near the z axis. If the coordinate axis is chosen such that the source for the second pulse is on the z axis and the source for the first pulse is in the xz plane, then the phases at the position of the \( n \)th atom or molecule can be written as
\[ \phi_1(x,y,z) = k(z_\alpha - z_1) + \frac{k}{2(z_\alpha - z_1)} (x_\alpha - x_1)^2 + y_\alpha^2 \]  
(1-41)

\[ \phi_2(x,y,z) = k(z_\alpha - z_2) + \frac{k}{2(z_\alpha - z_2)} (x_\alpha - x_2)^2 + y_\alpha^2 \]  
(1-42)

\[ \phi_E(x,y,z) = k(z_\alpha - z_E) + \frac{k}{2(z_\alpha - z_E)} (x_\alpha - x_E)^2 + (y_\alpha - y_E)^2 \]  
(1-43)

Substituting these equations into \((\phi_E - 2\phi_2 + \phi_1) = \text{const}\), and using the condition that the sum must be independent of \(\alpha\), implies that the sum must be independent of \((x_\alpha)^2\) and \((y_\alpha)^2\). This is true if

\[ \frac{1}{z_\alpha - z_E} = \frac{2}{z_\alpha - z_2} - \frac{1}{z_\alpha - z_1} \]  
(1-44)

Normally, this condition is written as

\[ \frac{1}{R_E} = \frac{2}{R_2} - \frac{1}{R_1} \]  
(1-45)

where \(R = z_\alpha - z\) and is the general relationship between the radii of curvature.

When the gradient of Equations (1-41) to (1-43) is divided by \(k\) and substituted into \(\kappa_E = 2\kappa_2 - \kappa_1\), the result for the \(x\) component is

\[ \frac{x_\alpha - x_E}{R_E} = \frac{2x_\alpha}{R_2} - \frac{x_\alpha - x_1}{R_1} \]  
(1-46)

This expression is independent of \(x_\alpha\) and

\[ x_E = -\frac{R_E}{R_1} x_1 \]  
(1-47)
Repeating for the $y$ component yields a similar expression which is independent of $y_a$ and

$$y_E = 0 . \quad (1-48)$$

Thus, the echo will focus at

$$x_E = -\frac{R_E}{R_1} x_1$$
$$y_E = 0$$

and

$$z_E = z_0 - R_E \quad , \quad (1-49)$$

where $z_0$ is the position of the cell. The length of the cell introduces an uncertainty in the echo position which can be readily calculated by using Equation (1-45).

Also, the direction of propagation of the echo can be predicted from Equation (1-39). If the second pulse is at an angle of $\theta$ to the first, then the echo propagates along an angle of $2\theta$ to the first pulse.

**Phase Conjugation**

The phases of the pulses can be rewritten as

$$\phi_0(x, y, z) = \gamma_0(x, y, z) + kz \quad (1-50)$$

where $\gamma_0$ represents the departure from plane wave behavior. If $\gamma_2 = 0$, ...
then the second pulse is a plane wave and $R_2 = \omega$. Equations (1-36) and (1-45) become

$$\gamma_E + \gamma_1 = \text{const} \quad (1-51)$$

and

$$R_E = -R_1 \quad (1-52)$$

The phase of the electric field vector becomes

$$\phi_E = -\gamma_1 + k_z \quad (1-53)$$

which is the phase conjugate of $\phi_1$. Figure 2 is a sketch of this when the first pulse is diverging and the second is plane. The initial pulse originates below the axis and the echo comes to focus below the axis. This is a property of phase conjugation which would not be seen if the Na vapor simply acted as a lens.

**Initial Pulse Intensities**

The conditions for echo formation $\phi_E - 2\phi_2 + \phi_1 = \text{constant} \quad \text{and} \quad t - t_3 = t_2 + t_1$ given in Equations (1-36) and (1-37), respectively, were based on maximizing the sum of the exponential terms in $F_\alpha$ and did not consider the coefficients $f_1^*$, $g_1^*$, and $g_2^2$. These terms are functions of $\Delta = \omega_{ab} - \omega + \delta$, $\nu$ the interaction strength, and $t$ the pulse length and provide a criteria for determining the initial pulse intensities.

For the first pulse and when $\Delta = 0$, the product $g_1^*, f_1^*$ from
Figure 2. Production of a Phase Conjugated Pulse. The First Pulse is Diverging, the Second Pulse is a Plane Wave, and the Echo Pulse is Convergent as Shown. The First Pulse Virtual Source and Focal Point of the Echo are Below the Principle Axis.
Equations (1-26) and (1-27) is largest when

\[ \sin (2q_1 \tau_1) = 1 \]

or

\[ 2q_1 \tau_1 = \pi/2 \quad \text{(1-54)} \]

This pulse is often called the \( \pi/2 \) pulse and \( f_1 = g_1 = 2^{-1/2} \). \( g_2^2 \) is largest when

\[ \sin (g_2 \tau_2) = 1 \]

or

\[ 2q_2 \tau_2 = \pi \quad \text{(1-55)} \]

The second pulse is often called the \( \pi \) pulse and \( g_2^2 = 1 \).

From the definition of \( q \) given in Equation (1-28), it follows that

\[ v_1 = \frac{\pi}{4\gamma \tau_1} \quad \text{(1-56)} \]

and

\[ v_2 = \frac{\pi}{2\gamma \tau_2} \quad \text{(1-57)} \]

where \( \gamma = (J_{b1} l_a M_b \, J_{a1} m_a) \). In the special case when \( \tau_1 = \tau_2 \), then \( v_2 = 2v_1 \).

The electric field of the \( \pi \) pulse from the definition of \( v \) in Equation (1-29) is

\[ E_\pi = \frac{\pi \hbar (2J_a +1)^{1/2}}{2\tau \pi \gamma (J_{b1} \alpha P || J_{a1})} \quad \text{(1-58)} \]
\((J_b | P | J_a)\) is the reduced matrix element and can be calculated from the relation

\[
A = \frac{16\pi^3}{3(2J_a + 1)\hbar^3 \epsilon_0} |(J_b | P | J_a)|^2
\]

(1-59)

found in Reference 22. For Na, the Einstein A coefficient is \(6.25 \times 10^7\) sec\(^{-1}\) which gives \((\frac{3}{2} | P | \frac{1}{2}) = 4.26 \times 10^{-29}\) coulomb meters.

The Clebsch Gordan coefficients are calculated for the special case of linear polarization with \(\hat{u} = \hat{z}\) where \(\hat{u}\) is the electric field polarization. In this case, the only value of \(M\) allowed is \(M = 0\). Thus

\[
(\frac{1}{2} \frac{1}{2} 0 | \frac{3}{2} \frac{1}{2}) = \sqrt{2/3}
\]

and

\[
(\frac{1}{2} - \frac{1}{2} 0 | \frac{3}{2} - \frac{1}{2}) = \sqrt{2/3} .
\]

(1-60)

Substituting these values into Equation (1-58) with \(\tau = 5 \times 10^{-9}\) seconds gives \(E = 1.91\) volts/meter and

\[
I = \frac{2E^2}{\mu_0 C} = 1.94 \times 10^4\) watts/m\(^2\).
\]

(1-61)

For the \(\frac{1}{2} - \frac{1}{2}\) transition, \(I = 3.88 \times 10^4\) watts/m\(^2\).

**Echo Formation Time**

When the conditions for echo formation were derived earlier, the
time of echo formation was based on a sum over $\Delta$ of the exponential term

$$e^{-i\Delta(t-t_3-t_2^+t_1)}$$

found in the expression of $F_\alpha$. See Equations (1-35) and (1-37). This sum did not include the terms $f_1^*$, $g_1^*$, $g_2^2$ which are also a function of $\Delta$. The effect of these terms is non-trivial and will be considered next.

From Equations (1-27) and (1-28)

$$g_1^*g_2^2 = \frac{-i\nu_1}{\frac{1}{2}(\Delta^2+4\nu_1^2\gamma^2)} \sin \left[\frac{1}{2}(\Delta^2+4\nu_1^2\gamma^2)t\right]$$

$$\times \frac{-\nu_2^2}{\frac{1}{4}(\Delta^2+4\nu_2^2\gamma^2)} \sin^2 \left[\frac{1}{2}(\Delta^2+4\nu_2^2\gamma^2)t\right]$$

where $\gamma = (J_b)_{b|a} (J_m)_{a|a}$. This product may be put into dimensionless form by using the substitution

$$x = \frac{\Delta}{4\nu_1\gamma} = \frac{\Delta\tau_1}{\frac{1}{\pi}} = \frac{\Delta\tau_1}{2\gamma\nu_2\tau_2} .$$

which yields

$$g_1^*g_2^2 = \frac{1}{\gamma^3} \frac{\sin \left(\sqrt{1+4x^2}\frac{\pi}{4}\right)}{\sqrt{1+4x^2}} \frac{\sin^2 \left(\sqrt{1+x^2}\frac{\pi}{2}\right)}{1+x^2}.$$
$g_1^*g_2^2$ remains large as $x$ increases from zero until $x$ becomes larger than 1. For $x > 1$, the behavior may be approximated by

$$\frac{i\pi^2 \sin^3 \left( \frac{x\pi}{2} \right)}{8y^3 \left( \frac{x\pi}{2} \right)^3}$$

which approaches zero rapidly and remains small as $x$ increases. Thus, the sum over $\Delta$ is important only when $x < 1$ or equivalently when $\Delta < \frac{\pi}{x}$. The effect of $f_1^*$ is now investigated. Using the same substitutions used to convert $g_1^*$ and $g_2^*$ into dimensionless form gives

$$f_1^* = \cos \left( \sqrt{1+4x^2} \frac{\pi}{4} \right) + \frac{2x}{\sqrt{1+4x^2}} \sin \left( \sqrt{1+4x^2} \frac{\pi}{4} \right).$$

This complex function can be put into exponential form by using the following relation

$$\cos \theta + F(\theta) \sin \theta = \{\cos^2 \theta + F^2(\theta) \sin^2 \theta\} \exp \{i \arctan [F(\theta) \tan \theta]\}$$

which yields

$$f_1^* = \{\cos^2 \left( \sqrt{1+4x^2} \frac{\pi}{4} \right) + \frac{4x^2}{1+4x^2} \sin^2 \left( \sqrt{1+4x^2} \frac{\pi}{4} \right)\} e^{i\Delta ts}$$

where

$$ts = \left( \frac{1}{\Delta} \right) \arctan \left( \frac{2x}{\sqrt{1+4x^2}} \tan \sqrt{1+4x^2} \frac{\pi}{4} \right)$$
The condition for echo formation from Equation (1-35) now becomes

\[ e^{+i\Delta t_s} e^{-i\Delta (t-t_3-t_2+t_1)} e^{-i\Delta (t-t_3-t_2+t_1-t_s)} = e \]  

(1-71)

where \( t_s \) is a function of \( \Delta \).

Recalling that the sum is only important for \( x < 1 \) or \( \Delta < \frac{\pi}{T_1} \), \( t_s \) may be calculated for \( x < 1 \). From the expression of \( t_s \) given in Equation (1-70) for small \( x \), \( \tan \left( \sqrt{1 + 4x^2} \pi/4 \right) \approx 1 \) and

\[ t_s \approx \frac{2x}{\Delta} = \frac{2\tau_1}{\pi} \]  

(1-72)

As \( x \) increases, \( t_s \) slowly decreases until at \( x = 1 \), \( t_s = 0.9 \left( \frac{2\tau_1}{\pi} \right) \) or a 10% decrease from when \( x = 0 \). \( t_s \) for \( x > 1 \) is unimportant because of the product \( g_1^s g_2^2 \) which is small for \( x > 1 \). Thus, \( t_s \) can be considered to be approximately constant and the sum over \( \Delta \) in Equation (1-71) taken. In order to ensure a nonzero result, it must be true that

\[ t = t_3 + t_2 - t_1 + t_s \]  

(1-73)

where

\[ t_s = \frac{2\tau_1}{\pi} \]  

(1-74)

Therefore, the peak of the echo is expected to occur at a time after the second pulse equal to the initial pulse separation plus \( (2/\pi) \)
times the pulse length of the first pulse.

**Phase Matching**

When the initial pulses are not collinear, then the matching condition \( \hat{k}_E = 2\hat{k}_2 - \hat{k}_1 \) cannot be met and there is a resultant loss in echo intensity. This loss can be obtained in a straightforward manner by calculating \( \Delta \phi = \phi_E - 2\phi_2 + \phi_1 \). The general expression of the phase used is the same as before and is

\[
\phi_0 = k(z-z_0) + \frac{k}{2(z-z_0)} \left\{ (x-x_0)^2 + (y-y_0)^2 \right\}
\]

(1-75)

To simplify the analysis, let \( z_\alpha = z_c + \Delta z_\alpha \) where \( z_c \) locates the front of the cell. This substitution and the use of the binomial expansion allows the phase in the cell to be written in the form

\[
\phi_0 = \phi_0' + k_0 \Delta z_\alpha \left\{ 1 - \frac{(x_\alpha-x_0)^2+(y_\alpha-y_0)^2}{2(z_c-z_0)^2} + \ldots \right\}
\]

\[
= \phi_0' + k_0 \Delta z_\alpha .
\]

(1-76)

In the above \( \phi_0' \) is the phase at \( z_\alpha = z_c \) and \( k_0 \Delta z_\alpha \) represents the phase change as a function of \( z_\alpha \). The \( (x_\alpha)^2 \) terms in the expansion are now multiplied by \( \Delta z_\alpha \) and the expression is not independent of \( \alpha \). This last term is assumed small and ignored for \( (x_\alpha-x_0)^2 + (y_\alpha-y_0)^2 \ll (z_c-z_0)^2 \) which is equivalent to ignoring the effects of the radii of curvature. This condition can be written as \( d/2 \ll R \), where \( d \) is the
beam diameter. \( \Delta \phi \) can now be written as

\[
\Delta \phi = \phi' - 2\phi_2' + \phi_1' + (\Delta k)(\Delta z_a)
\]  

(1-77)

where \( \Delta k = k_E - 2k_2 + k_1 \). In this form, the phase term in \( F_\alpha \) becomes

\[
\exp[i(\phi' - 2\phi_2' + \phi_1')] = \exp[i(\phi' - 2\phi_2' + \phi_1')] \exp(i\Delta k\Delta z_a).
\]  

(1-78)

The exponential of \( \phi_E' - 2\phi_2' + \phi_1' \) is evaluated at \( z_a = z_c \). Thus, the sum over \( \alpha \) involves the terms \( x_\alpha ^2 \) and \( y_\alpha ^2 \) which is not multiplied by \( \Delta z_a \). As shown in the derivation of Equation (1-45), the exponential is independent of \( x_\alpha ^2 \) and \( y_\alpha ^2 \) leaving the sum over \( \exp(i\Delta k\Delta z_a) \) which must be evaluated. This can be accomplished by converting the sum to an integral over \( L \), the length of the cell. The result is proportional to

\[
\frac{e^{i\Delta kL - i\Delta kL}}{i\Delta kL}
\]  

(1-79)

which is also proportional to the electric field amplitude. The intensity of the echo is then proportional to

\[
\frac{\sin^2 (\Delta kL/2)}{\Delta kL/2}.
\]  

(1-80)

\( \Delta k \) may be expressed as a function of \( \theta \), the angle between \( \hat{k}_1 \) and \( \hat{k}_2 \) by using a vector sum as shown in Figure 3. Here \( \Delta k = k_E - 2k_2 + k_1 = 2(k_1 - k_2) = k\theta^2 \). Using this approximation \( \theta_{\text{max}} \), the angle at which
Figure 3. Vector Diagram for Determining Phase Mismatch, $\Delta k$.

$$\Delta k = k_E - 2k_2 - k_1 = k\theta^2$$
the intensity drops by a factor of $e^{-1}$, is

$$\theta_{\text{max}} \approx \sqrt{\frac{\lambda}{2L}}.$$  \hspace{1cm} (1-81)

This expression is good for plane waves and spherical waves where $d << 2R$. For typical values of $\lambda = 5.89 \times 10^{-7}$ meters and $L = 2.5 \times 10^{-2}$ meters, $\theta_{\text{max}}$ becomes 3.4 milliradians. When $L = 10^{-3}$ meters, $\theta_{\text{max}} = 17$ milliradians.

**Other Misalignment Effects**

In the derivation of $F_{\alpha}$ given in Equation (1-35), it was assumed that $\dot{\phi}_1 = \dot{\phi}_2 = \dot{\phi}_E$ which is valid if the initial pulses are collinear plane waves. When the pulses are misaligned or curved, then the derivation must be repeated. This yields

$$F'_{\alpha} = F_{\alpha} \exp\{i[(\dot{\phi}_2 - \dot{\phi}_E)_{\alpha}(t_3 + t_2) - (\dot{\phi}_1 - \dot{\phi}_E)_{\alpha}t_1]\}$$  \hspace{1cm} (1-82)

where $F_{\alpha}$ is the previous result. The expression can be simplified by using the time derivative of $\dot{\phi}_E = 2\dot{\phi}_2 - \dot{\phi}_1$ to replace $\dot{\phi}_2 - \dot{\phi}_E$ with $\dot{\phi}_1 - \dot{\phi}_2$. Also, $(\dot{\phi}_1 - \dot{\phi}_E)t_{\text{1}}$ is much smaller than $(\dot{\phi}_2 - \dot{\phi}_E)(t_3 + t_2)$ and will be ignored.

The difference term $\dot{\phi}_1 - \dot{\phi}_2$ is the doppler shift which may be rewritten as

$$(\dot{\phi}_1 - \dot{\phi}_2_{\alpha}) = \frac{\omega}{c} (\hat{k}_1 - \hat{k}_2) \cdot \vec{v}_{\alpha}.$$  \hspace{1cm} (1-83)
If $\theta$ is the angle between $\hat{k}_1$ and $\hat{k}_2$, then

$$\langle \hat{\phi}_1 \cdot \hat{\phi}_2 \rangle_\alpha = \frac{w}{c} \theta v_{ap} \quad (1-84)$$

where $\theta$ is small and $v_{ap}$ is the component of $\vec{v}_\alpha$ parallel to $(\hat{k}_1 - \hat{k}_2)$. Using this $F'_\alpha$, can now be written as

$$F'_\alpha = F_\alpha \exp \left\{ i \frac{2\pi}{\lambda} \theta v_{ap} (t_3 + t_2) \right\}. \quad (1-85)$$

The expression in the brackets can be made independent of $\alpha$ by replacing it with the thermal average over $v_{\alpha}$. Assuming a Maxwellian distribution, this leads to

$$F'_\alpha = F_\alpha e^{-b^2 \theta^2} \quad (1-86)$$

where $b^2 = \pi^3 \overline{v(t_3 + t_2)/2\lambda}^2$ and $\overline{v}$ is the average velocity.

For plane waves, $\theta$ is a constant for all points on the wavefront. Thus, $\theta_{max}$, the maximum allowed angle between $\hat{k}_1$ and $\hat{k}_2$, can be determined from $e^{-b^2 \theta_{max}^2} = e^{-1}$ or

$$\frac{1}{b} = \frac{2\lambda \pi^{-3/2}}{\overline{v(t_3 + t_2)}}. \quad (1-87)$$

When $\lambda = 5.89 \times 10^{-7}$ meters, $\overline{v} = 640$ m/s, and $(t_3 + t_2) = 20 \times 10^{-9}$ seconds, $\theta_{max} = 16.5$ milliradians.
CHAPTER II

THE EXPERIMENTAL APPARATUS

The experimental apparatus that was used to produce focused photon echoes in Na vapor is shown in Figure 4. Many of the pieces were commercially produced and will be discussed briefly in this Chapter. They include the argon-ion laser, dye laser, nitrogen laser, optics, and the oscilloscopes that were used in conjunction with the high speed detectors. The rest of the items were constructed in the laboratory and include the dye amplifier, Na oven, Na cells, two high speed detectors, optical tables, and holders. Construction details and general discussions of these items comprise the bulk of the Chapter.

The discussion begins with the argon-ion and dye lasers and follows the output beam through the apparatus until it reaches the high speed detector.

Argon-Ion and Dye Laser

A CR-6 Argon-Ion Laser was used to pump a 590 Dye Laser, both of which were purchased from Coherent Radiation, Palo Alto, California. The argon-ion laser was operated on all lines with the output adjusted to 4 watts. Electrical power was furnished by a 220 VAC line with the system using up to 70 amperes. Excess heat produce by the large current flow was removed by a 4 gallon per minute water flow
Figure 4. The Experimental Arrangement. B = Beamsplitter, G = Grating, L = Lens, T = Telescope, TC = Na Vapor Test Cell.
which passed through a 25 micron filter before entering the laser assembly.

The jet stream dye laser used a solution of Rhodamine 6G dye in an ethylene glycol to produce a continuous and tunable output from 580 to 630 nanometers. A birefringent filter was used to tune the vertically polarized output which had a 50 GHz linewidth. This linewidth was reduced to a single cavity mode by use of two etalons, which were 0.5 millimeter and 10 millimeters thick. The cavity mode spacing was 340 MHz and the output beam diameter was 0.5 millimeter. Further information on the argon-ion and dye lasers is contained in the operating manuals and will not be discussed here.

The lasers were mounted on a table that was separate from the rest of the apparatus. This table was constructed from six 4 foot x 8 foot x 1 inch particle boards purchased from a local distributor. The boards were glued together and covered with "NEVAMAR LAMINATE" purchased from the American Plastics Laminates, Inc., Columbus, Ohio.

A four-legged steel support for the particle boards was constructed from four 30 inch long and 4 inch diameter steel pipes which were held in place by angle irons. Four of the angle irons were welded near the top of the legs and four more near the bottom forming a rectangular structure 60 inches long by 40 inches wide by 30 inches high. The particle boards were supported only by the tops of the four legs which caused the boards to flex in the middle and at the ends. Since the table was exposed to the air, humidity
changes caused the table to swell and contract over a period of time. This, along with the natural sagging of the particle boards, caused frequent realigning of the lasers. A 2 inch x 1/4 inch x 8 foot angle iron was placed lengthwise over two of the legs and beneath the particle board with the lasers situated on the table top directly above the angle iron. The stability of the lasers was greatly improved but still required some adjustment (see Experimental Procedures).

When the argon laser tube was new, an output of 4 watts as measured by a Coherent Radiation 210 Power Meter could be obtained at 22 amperes. After 400 hours running time, 26 amperes were necessary to produce the same output.

The argon laser was occasionally found to lase in the doughnut mode which lowered the dye laser output considerably. This could be easily detected by looking sideways at the beam. A dark line through the center indicated a lower intensity there which is not seen when the laser is operating in the TE_{M} mode. The doughnut mode was initially pointed out by a Coherent Radiation technician who corrected the problem by adjusting the aperture inside the laser.

The Brewster angle windows inside the argon laser were kept clean by using a flowing gas system. Small diameter teflon tubing and a needle valve was connected to a N_{2} regulator set for a pressure of a few pounds per square inch. The tubing formed a "Y" so both windows could be constantly exposed to clean N_{2} gas,
thereby creating a dust-free environment for the windows.

**Dye Amplifier**

The dye amplifier is composed of a beamsplitter, focusing lens, dye cell, telescope, diffraction grating and various mounts or holders. See Figures 4, 5, and 8. A single platform held the beamsplitter and 1 meter focal length focusing lens as shown in Plate I. This platform was mounted on an X-Y translator orientated so the platform could be moved transversely to the laser beam. The translator is similar in design to Model 405 Dual-Axis Translation Stage from Newport Research Corporation and was constructed by the Physics Shop. It is composed of two stages and a center support or guide. Each stage was held in place by two guide pins and translated by a micrometer head and opposing spring on the opposite side of the stage. The only difficulty encountered occurred when it was vertically mounted. The combined weight of the translator itself and of the platform exceeded the capacity of the spring to return the stage when the micrometer head was retracted. A stronger spring should alleviate this problem.

The dye cell was designed and constructed in our laboratory. See Figure 5. Stainless steel was used for the main body and brass for the window holders. These holders were designed so that the O-ring encircled the window and when tightened against the cell caused pressure on the window, sides and bottom of the holder, and cell body forming an effective seal. The quartz window, which was transparent to the U-V nitrogen laser output, rested against a ledge in the cell that
Plate I. The Beamsplitter, Focusing Lens, and Dispersing Prism for the Dye Amplifier.
A 34 millimeter from the bottom of the cell. A teflon O-ring was placed above this and forced into place by a thin-walled, "plunger type" clamp. This clamp was shaped like a "top hat" with a hollow end. The thin wall applied enough pressure to seal the window and was thin enough not to block the nitrogen laser beam. Teflon was used as it would flow under pressure and form a better seal between the quartz and the side of the cell. Before the clamp was tightened the window was checked to insure that it rested squarely against the ledge in the cell. If it did not it could easily chip or crack. The clamp was tightened and left overnight to allow the teflon to flow into place. The next morning it was tightened again. This procedure resulted in tight seals with little difficulties encountered.

The dye was introduced to the cell by 1/8 inch diameter thin wall stainless steel tubing soldered to the cell. The dye was introduced into the cell so that it flowed upwards as it passed between the quartz window and the cell body. A downward flow was not used as bubbles tended to form in the chamber which destroyed the optical quality of the cell. A 0.06 inch hole was placed on opposing sides of the cell to allow the dye laser beam through the cell. One of the holes was raised 0.05 inches so that the cell could be rotated 40 about a vertical axis perpendicular to the path of the dye laser beam. This caused reflections from the cell windows to be reflected out of the gain region.

The dye pump assembly was also constructed in the laboratory. See Figure 6. A magnetically coupled gear pump (Model 04-60-303),
Figure 5. The Dye Cell. a - Teflon O-Ring 20 Mills x 40 Mills x 1 Inch in Diameter, b - 1 Inch x 1/8 Inch Quartz Window, c - Thin Wall, about 10 Mills Thick, d - Ledge for Window b, e - Dye Feed Tubes, f - 1 Inch x 1/8 Inch Pyrex Window, g - Neoprene O-Ring, h - Window Holder, i - Entrance/Exit Hole for Dye Laser Beam.
Plate II. The Dye Cell for the Dye Amplifier.
Figure 6. The Dye Pump for the Dye Amplifier
purchased from the Micropump Corporation, Concord, California forced
the Rhodamine 6G dye in ethanol solution through a water cooled counter
flow heat exchanger, a 25 micron filter, a by-pass valve, through the
dye cell and finally back to the stainless steel reservoir where it
was recycled through the system. 1/4 inch polyethylene tubing with
Swagelock fittings were used to connect the pieces. The 25 micron
cartridge type filter was mounted in a standard filter holder. The
heat exchanger was constructed by slipping a 1/2 inch polyethylene
tubing over a 1 meter length of the smaller tubing which carried the
dye. T joints were fitted over the ends of the larger tubing so the
smaller tubing passed straight through the joint and the cooling water
could flow through the side port and past the inner tubing. When in
use this exchanger maintained the temperature of the dye to within
1°C of the water temperature. The same style of heat exchanger made
from copper could be made much smaller and still control the temperature
of the dye.

The pressure of the dye was set by the by-pass valve and could
be maximized at a pressure of 14 pounds per square inch corresponding
to a flow rate of 0.8 liters/minute. The major effect of different
flow rates was the distortion and "flickering" of the transmitted
laser beams with the minimum effect occurring at the maximum flow
rate.

A Molectron U-V 12 Nitrogen Laser was used as the pump laser for
the dye amplifier. This laser produced a pulsed output with a peak
power of 250 kilowatts in a 1 inch long and 1/8 inch wide beam. The
laser was operated at 20 pulses per second with a nitrogen flow rate of 12-15 cubic feet/hour. The laser was mounted on a 3 legged platform with adjustable legs.

Several difficulties were encountered with the operation of the laser. When initially used arcing occurred between the high voltage lines and the chassis causing the power supply to fail. A new power supply was sent by Molectron but arcing soon developed again. The laser was returned to the supplier where the power supply was replaced and new wiring installed. When returned to the laboratory a rectifier soon failed due to a lead reversal at the thyatron when repaired at the company. This was corrected, the rectifier replaced, and no further arcing problems developed.

Although it was first discovered on another nitrogen laser (NRG-0.35-8-170) it should be noted that a large vacuum line was necessary to allow the nitrogen to flow quickly through the laser tube to prevent degradation of the power output. When first set up, the output of the NRG laser was low for no apparent reason. When the 1/2 inch polyethylene tubing which connected the nitrogen exhaust port on the laser to the vacuum line was replaced by 1 inch diameter copper tubing, the output rose to expected levels. The UV-12's vacuum exhaust line was constructed from 1 inch diameter hose leading to 2 inch copper vacuum lines and has always operated according to specifications. The 2 inch vacuum lines are about 3 meters long and join 4 inch copper tubing which lead to the vacuum pump in the basement.
At 20 pulses per second the recommended flow rate is 10 cu. ft./hr. To achieve this the nitrogen supply regulator was set at 20 pounds per square inch, the vacuum adjust fully opened, and the pressure adjustor set to 15 torr. The vacuum adjust was then closed until the pressure inside the laser tube read 50 torr.

The nitrogen supply was a 160 liter liquid nitrogen dewar ordered through Howard Baker, a staff technician. This method of nitrogen supply was superior to bottles of compressed nitrogen because 1) the supply lasted about a month, 2) the system was self purifying, 3) a supply of liquid nitrogen was available, and 4) the cost was about one third of that available from the university supply of compressed gas. Also these cylinders had a maximum capacity of 225 cubic feet which would require replacements at least twice a week when the lasers were operating. Only one major difficulty was encountered with the dewars. On a number of occasions the dewars had a small leak or were poorly insulated so the pressure inside the dewar would rise until nitrogen was lost through the safety "blowout" valve which opened when the pressure reached 235 pounds per square inch.

The output of the UV-12 passed through a 15 centimeter focal length cylindrical quartz lens located 4.6 centimeters from the gain region in the dye cell and was focused down to form a line 25 millimeters long and 0.5 millimeter wide.

To allow for adjustment of the lens a lens mount was inserted into the end of a 9.7 centimeter long brass tube which was 3.81
centimeters (1 1/2 inches) in diameter. An aluminum block 3 inches x 4 1/2 inches x 1 inch was used to hold the tube in place. The block was fastened to the dye cell holder by two screws fastened in a vertical line near the bottom of the block. The height of the block was adjusted by loosening the screws, sliding the block in the slit, and retightening the screws. See Figure 7. A hole was cut into the face of the block to allow the brass tube to slip easily through. A slit was cut from the top of the block and a tightening screw placed in the side to allow the hole to be tightened when final adjustments had been made. This construction allowed the lens to be rotated and translated vertically and horizontally.

Figure 8 shows the arrangement of the dye cell, telescope, grating and various holders used to mount the apparatus on the invar rod purchased from Carpenter Steel Inc., Cincinnati, Ohio. Invar was chosen because of its low thermal expansion which minimized the realignment necessary due to changes in room temperatures. The 1 5/8 inch diameter of the 26 inch long rod was reduced to 1.600 inches in diameter and polished to remove surface scratches.

The aluminum holders were fastened to the invar rod by a flexible clearance hole and tightened with a screw similar to the method used to hold the brass tube for the nitrogen laser beam focusing optics. See Figure 7. A 0.005 inch clearance was allowed between the invar rod and the clearance hole to allow it to move easily but to hold securely when the screw was tightened.
Figure 7. Lens Holder and Mount for Focusing of Nitrogen Beam. Lens is Adjusted by Rotating or Sliding Brass Tube and Loosening the Two Screws that Hold the Mount to the Dye Cell Holder.
The dye cell holder was milled at a $4^\circ$ angle to the horizontal to allow the cell to tip as mentioned in the dye cell discussion. The cell was held in place by 2 set screws and a support which held the cell by the sides. Minor adjustments were accomplished by use of thin shim stock under or on the sides of the cell.

The 13X telescope was purchased from Metrologic Instruments, Inc., Bellmawr, New Jersey (60-270 13X Collimator). To mount the telescope a hole 0.005 inch larger than the telescope was drilled into the top of a piece of aluminum 2 inches x 2 1/2 inches x 4 inches at the proper position so that the laser beam passing through the dye cell parallel to the invar rod would pass through the center of the telescope. Two set screws at the top of the holder and two at the side held the telescope firmly in place.

The focus of the telescope could be adjusted by rotating the threaded lens holders with a special "key" supplied by the manufacturer. The adhesive used to hold the lens in the lens holder was very soluble in acetone which should be noted when cleaning the optics.

The one inch square 2380 line/mm diffraction grating blazed at 500 nanometers was purchased from Phase-R Company, New Durham, New Hampshire (#BGR23805). The grating was mounted in a Precision Mirror Mount, Model B-44-60 from Oriel Corporation, Stamford, Connecticut. This mount was attached to an aluminum wedge as shown in Figure 8.
Figure 8. The Dye Amplifier
Plate III. The Dye Amplifier and Nitrogen Laser.
The invar rod and attached components was mounted on an aluminum block 8 inches x 32 inches x 3/4 inches and the block could be adjusted by the three adjustable screws which served as legs. The amplifier was secured to the platform by two 3/4 inch x 3/4 inch x 3 1/2 inch aluminum blocks screwed to the platform on either side of the dye cell holder and support mount. Two screws were placed in one of the blocks which when tightened forced the amplifier into the other and held it securely.

Mirror and Beamsplitter Mounts and Lens Holders

The optics in the beam delay line with the exception of the two mirrors used to redirect the second pulse were mounted on an aluminum block measuring 3 feet x 2 feet x 1 3/4 inches. The mirrors and beamsplitters were 1 inch in diameter and mounted in precision mounts which are shown in Figure 9. Before these mounts were constructed numerous other mounts were tried but the orientation of the holders was difficult to adjust to better than $10^{-2}$ radians. These mounts were used to hold the lenses, neutral density filters, and the crossed polaroid beam attenuator which did not require precise orientation changes.

The precision mounts were constructed in the Physics Shop from aluminum stock. As shown in the diagram the front plates pivoted on a 3/8 inch diameter steel ball which separated the plates by 1/8 inch. Two extension springs held the front plate while adjustments were accomplished by turning the adjustment screws. The 10-32 screws
Figure 9. Precision Mirror Mount. a - 10-32 Tap Hole; b - 1/4 inch Hole with 4-40 Screw to Hold Small Spring; e, c - Small Taped Hole to Hold Pivot Ball; f, c - 0.196 inch Hole with 1/8 inch Steel Ball Mounted in Bottom. See Insert, e - Small Extension Springs; f - 3/8 inch Steel Ball; g - 10-32 Stainless Steel Screws with Smooth Ends.
Plate IV. A Precision Mount with Mirror and Mirror Holder.
were flattened on the end and allowed to push against the two 1/8 inch steel balls mounted on the front plate. The balls were in the bottom of 0.196 inch clearance holes that fitted the screws and prevented the plates from shifting. The holes in the center of the plates were 2.005 inches in diameter. This mount was very precise and able to be adjusted routinely to $10^{-4}$ radians.

The optics were held in 2 inch diameter lens holders designed to hold the 1 inch diameter optics in place by a screw-on cap. The holder was attached to the precision mount by glue or by setscrews inserted into the sides of the front plate.

The lenses, neutral density filters, and polaroids were 49 millimeter camera optics purchased from a local camera store. They provided inexpensive and readily available optics that was of comparable quality to the other optics used in the system. These other optics include the dye cell windows, Na oven and Na cell windows which were flat to 3-10 waves across the surface.

The precision mounts were screwed to 3 inch x 3 inch x 1/2 inch aluminum blocks which put the center of the optics 2 1/4 inches above the table. Once into position the precision mounts were fastened to the table by double sided "Tiger" tape from the Stewart Company. This tape grips firmly to metallic surfaces but dissolves readily in acetone for removal. The lens holders $L_1$, $L_2$, and $L_3$ were mounted on a base plate with three height screws adjustment which also served as legs. Translation was accomplished by sliding the mounts across the table.
The beam attenuator was composed of 2 polaroids with the first adjusted vertically and the second able to rotate freely. This provided an easy method of beam attenuation.

**Sodium Oven**

Before the sodium cell oven used in this experiment was constructed, several prototypes were built and tested. The basic design was to use a long heat conducting pipe with open ends. The cell was mounted in the middle of the pipe, baffles inserted to restrict the convection currents and holes drilled in the center of the baffles to allow the laser beams through the oven. Heater tape was then wrapped around the oven to provide an even heat distribution. This method failed miserably. The temperature distribution across the cell was very uneven usually resulting in the sodium collecting on the cell windows rendering them opaque.

An evacuated oven (see Figure 10 and 11) was constructed in which radiative rather than conductive or convective heating was used. The outside shell was constructed from a copper T joint for 4 inch tubing. 6 inch flanges were soldered to the ends and coverplates bolted on. One inch pyrex windows were placed in opposite ends and sealed with black wax. A vacuum port was soldered in one side and a viewing port in the other.

The inner cell was a 6 inch long section of a 2 inch diameter copper tubing. Copper discs were placed in the ends and fastened by 2-56 Stainless Steel screws. A 1 centimeter aperture was drilled in each the centers of the discs. One of the discs was attached to
Figure 10. The Outer Shell of the Sodium Oven.
Plate V. The Outer Shell of the Na Cell Oven.
Figure 11. The Inner Shell of the Sodium Oven.
Plate VI. The Inner Shell of the Na Cell Oven.
an outer shell flange by three stainless steel screws which maintained a 3 centimeter separation between the inner and outer shell. Stainless steel was used because of its low thermal conductivity.

An electrical vacuum feed-through was inserted under a window in the outer shell allowing access to the heating coil and thermocouple. The heating coil was Tophet A wire inside a small diameter pyrex tubing which prevented the heating coil from shorting out. A rheostat was used to control the temperature and a thermocouple tip was placed near the cell but not touching the cell or the sides of the oven.

The sodium cell was held in place by 2 sets of 3 equally spaced screws located on the inner shell. These screws held the front and back of the cell and did not obstruct the windows. The tip of the cell was allowed to protrude through the wall of the inner shell resulting in a lower tip temperature. This caused the sodium to collect there and not on the windows. The viewing port consisting of a 1 inch diameter window on the side of the outer shell and a slit in the inner shell allowed direct viewing of the cell during the experiment.

The operating temperature of the oven was 170°C. To maintain this steady temperature at the cell, 4 amps at 30 volts or 120 watts was needed to compensate for the heat loss due to blackbody radiation. To insure only radiative heating, the oven was pumped to less than 1 torr pressure. A leak rate of 1-3 torr per hour forced daily evacuations, but over the course of an eight hour period had no
observed effect on the cell operating parameters.

**Sodium Cell**

The sodium cell itself was constructed from a 1 inch long section of 1 inch diameter pyrex tubing to which 1/8 inch thick pyrex windows were fused. A 5 millimeter diameter stem was attached to the side to allow introduction of sodium into the cell. See Figure 12. Several other types of glass, including quartz and uranium glass were also tried but pyrex had the best resistance to the sodium vapor.

The cells were constructed by the glass blowing shop on campus with the principle drawback being the deformation suffered by the windows during the fusing process. Uneven heating caused the windows to "bow" in, causing a non-uniform lens effect. A second type of distortion in the finished cell was caused by a brown coating on the windows. This caused a random scattering of the beam that interfered with many of the measurements. Cell quality was checked by shining an expanded He-Ne laser beam 1 centimeter in diameter through the cell and onto a screen located 5-6 meters away. The distortion could be noted by observing the changing shape of the pattern as the cell was moved. A second method was to use a long focal length lens to focus the pattern into a point. Two apparent foci could be seen, one for the undistorted beam and one for the distorted beam.

One of the cells was constructed in a slightly different manner and seemed to have less distortion. See Figure 12. A 5 millimeter diameter stem was attached to a length of 28 millimeter diameter
Figure 12. The Two Types of All Pyrex Sodium Cells.
Plate VII. The Na Cell.
pyrex tubing. The tubing then was cut to a length of 1 inch by a diamond saw located in the Technical Operations Shop. Using a lathe and small grind stone with flat edges, a counterbore was made into the glass until a 1 inch diameter window would fit in leaving about 2 millimeter of the tubing protruding above the window. Next, one window was placed in the cell and held so the window was horizontal. A small, hot flame from a oxy-methane torch was used to heat the cell until the window was lightly attached to the tubing. The cell was inverted and heat applied to the side of the glass until the thin wall had fused to the window. When inverted the ridge was down and away from the heat so it would not flow. The flame was kept small to prevent overheating and had to be moved frequently to prevent one side of the glass from cooling while working on the other. When fusion was complete, the cell was returned to the original position and heated to reduce the ridge left until the edge was smooth. It is important to always heat from the side and not the top. The heat will rise quickly when applied to the side keeping the window relatively cool. This was repeated on the other end allowing a minimum of 15 minutes to flame cool the cell when complete. The cell was immediately placed in an annealing oven, heated to 580°C for at least 1 hour and allowed to slowly cool to room temperature.

Several methods for transferring sodium into the cells were tried with varying degrees of success. After a number of attempts, it was
discovered that the operating temperature of the cells and the cell lifetimes were strongly dependent on the initial purity of the sodium. This was tested by immersing the cells in an oil bath and noting the lowest temperature at which spontaneous emission created by the dye laser beam was observed. For the early cells this was over 200°C, but with the transfer method discussed in the next section, this was reduced to 130°C.

The main failure mechanism for the early cells was a dark brown coating that formed on the inside of the cell. At temperatures above 200°C this coating formed rapidly, but at 150-170°C the lifetimes were extended to many hours. The first "good" cell had a lifetime of over 100 hours at 170°C. Although the dark coating was evident, the cell failed because of a lack of Na vapor. When first filled, there was an excess of sodium which formed in the stem as small condensed droplets. After the cell had been in use for well over 100 hours, it was noticed that over the space of a few hours the cell temperature had to be continually increased in order to maintain the proper cell absorptivity at the Na D lines. When the cell was examined at room temperature, no condensed Na was visible within the cell. The sodium had apparently reacted slowly with the pyrex and/or contaminates in the cell until all the free sodium was gone.
Sodium Transfer

The basic configuration for the Na transfer station is shown in Figure 13. The system was constructed from pyrex tubing by standard glassblowing techniques. It was assembled in sections with each section being cleaned by immersion in a glass cleaning solution and then were annealed in an annealing oven. The cells were also boiled in a NaOH solution to further remove impurities. Unfortunately, when the sections were fused together, they were recontaminated to a certain extent. Chamber 1 was initially left open so the sodium could be introduced. Stopcock 1 was closed and the cold trap baked out for 1/2 hour under a vacuum. The sodium, which was normally stored in kerosene, was dried off, and the outer surface removed with a sharp knife. The remaining sodium was silver in color, and was placed in Chamber 1. When about 1-2 grams of sodium was in the chamber, it was closed off by fusing the end. The system was pumped down by the cold trap and diffusion pump, and the entire glass system except Chamber 1 was baked out until the glass was clean and bright.

Chamber 1 was at a 30° angle to the horizontal to allow the melted sodium to flow into Chamber 2. The neck between Chamber 1 to 2 was made from capillary tubing. When the sodium was heated with a "cool" flame and flowed through the tubing, many of the impurities were left behind as residues on the glass surface. Chamber 1 was then removed and Chamber 2 heated slowly. At this point some con-
Figure 13. The Sodium Transfer Station.
taminates were visible on the surface of the liquid sodium.

When first heated, the sodium bubbled as gaseous contaminates were released. The sodium was heated for well over an hour until the sodium had repeatedly vaporized, condensed on the side, ran back to the bottom and the bubbling had ceased. Chamber 2 was now heated in a manner to maximize the amount of sodium passing into Chamber 3. Small diameter thick wall tubing removed from the ends of stopcocks were used to join these and the rest of the chambers. The openings were larger than for capillary which allowed faster sodium transfer and pumping speed.

When most of the sodium was gone from Chamber 2, the chamber was removed. The connection between Chamber 3 and 4 was heated carefully until Chamber 3 could be tipped about 30° down from its original position. This was to prevent a "slug" of sodium from forming and running into the next chamber. The tubing here must be thick-walled to prevent collapsing.

Chamber 3 was heated until the sodium had been transferred by vaporization into Chamber 4. Chamber 3 was then removed. Helium was allowed to pass through the nitrogen trap and into Chamber 4 until 1/4 to 1/2 atmosphere of pressure was reached. The helium was then pumped out "dragging" along any impure gases still around. This procedure was performed several times as the sodium was vaporized into the cells.
When the cells were opaque with the condensed sodium, they were "pulled off" by collapsing the stems. The stem was carefully annealed to prevent the development of pinholes. The body of the cell was gently heated until the sodium was condensed in the stem which made the first time use in the sodium oven much easier.

The discarded chambers often contained large amounts of sodium and had to be treated carefully. Normally, the chambers were wrapped in paper towels and broken into smaller pieces by a hammer. Small pieces of the broken glass and attached sodium were put a piece at a time in the sink and allowed to react with the water. Safety glasses were worn in case the escaping hydrogen ignited and exploded. The glass pieces were then discarded in the regular manner.

**High Speed Detector**

In order to analyze the pulse shape of the amplified signal and echo, a high speed detector with a response time of 2 nanoseconds or less was needed. To achieve this an impedance matched photodiode detector similar to that described by McCall was constructed. A Hewlett-Packard 5082-4220 photodiode was mounted on the center terminal of a Tektronix 017-0033-00 connector which provided a short, low inductance lead to the diode. Coax cable was then used to connect the detector to the 7904 Tektronix oscilloscope with 7A19 amplifier and 7B92 dual time base.
The photodiode was imbedded in the center of a brass disc which served as the positive plate of a capacitor. The area and thickness of the plate separation was designed to yield a capacitance of 1500 picofarads which then matched the detector to the 50\(\Omega\) cable and 50\(\Omega\) internal impedance of the oscilloscope.

The capacitor was constructed from 2 round brass plates 3 inches in diameter and with thicknesses of 1/2 and 1/4 inch. To insure a constant plate separation, the interfaces were polished smooth and a mylar sheet was used as the dielectric material.

After the connector was mounted on the positive plate, the negative plate was positioned and held in place by 4 nylon screws. A small tapered hole was placed in the center so the case of the photodiode could be forced against the plate to insure a good electrical contact. Care had to be exercised here as too much pressure would crack the window on the photodiode. Proper care in initially soldering the photodiode minimized this problem.

This assembly was mounted on the support as shown in Figure 14. The batteries were connected in series, with the negative terminal connected to the thick plate via one of the mounting screws. To minimize the inductance, a small wire was run through a hole in the aluminum support at the base of the capacitor and connected the positive terminal to the front plate at the lowest nylon screw. A 1 megohm resistor was also placed in this line. To minimize the
Figure 14. The High Speed Detector
Plate VIII. The High Speed Detector.
effect of radio frequency noise, the batteries were completely enclosed by 1/32 inch aluminum sheets which were formed into a rectangularly shaped box and screwed to the base.

A plastic light guard was constructed from the top of a round plastic box. The inside was painted black and covered with black paper. Felt was placed along the inside edges so when it fitted over the detector the felt blocked out light coming around the edges and held the cover in place.

The detector was mounted on a base with adjustable legs to allow height variations. This assembly was then mounted on an X-Y translator to allow horizontal adjustments.

The fastest risetime signal for measuring the response of the detector was the nitrogen laser which exhibited a 2 nanosecond risetime as expected. The sensitivity of 1 milliamp/watt/cm² allowed measurements of signals as low as 0.2 watt/cm² or 0.4 milliwatts striking the sensitive area of the detector which had an area of 2 x 10⁻³ cm². This sensitivity is based on the specification sheet for the diode and has not been independently calibrated. The sensitivity of the photodiode was wavelength sensitive and not calibrated for the U-V. When the output of the nitrogen laser was allowed to strike the photodiode, the expected pulse length was much longer than expected. This effect was caused by the fluorescence of the window and corrected by illuminating a dye such as Rhodamine 6G with the
nitrogen laser output and measuring the spontaneous emission of the dye. The dye lifetime is on the order of 1 nanosecond and the resultant pulse shape gave a good representation of the expected output shape.
CHAPTER III
EXPERIMENTAL PROCEDURE

This chapter contains a detailed explanation of alignment procedures and operational information. As in the last chapter the general operating procedures for the commercial items are contained in operating manuals and will not be discussed at length here. Finer points of operation which are not contained in the manuals are discussed especially for the case of the dye laser alignment. The theory and alignment procedures for the dye amplifier, which was constructed in the laboratory, are also contained herein.

The alignment of the beam delay line was straightforward and two methods of checking the beam alignment are discussed. On a number of occasions the equipment seemed to be working properly but no echoes were formed. Based on these experiences a trouble shooting guide was prepared and is contained in this chapter. Finally the methods of measuring the position and size of the echo are described.

Argon-Ion and Dye Lasers

Discussion of the experimental procedures for the argon-ion and dye lasers is presented in two parts. The first is a description of frequently used procedures encountered in daily operations. When these steps failed to produce a satisfactory dye laser output a
general system realignment was necessary and is discussed in the second part.

The instructions for turning the lasers on are in the manuals and were followed exactly. It was found that a warmup time of at least 1 hour at operating levels was necessary to stabilize the output of the argon-ion laser. After this time the output was set to 4 watts at the minimum current flow. These adjustments were usually small and the dye laser often produced a good signal with no adjustments. Adjustments made prior to the warmup period were of little value as the settings would change as the argon tube heated up and then would have to be readjusted to near the initial settings.

If the output of the dye laser was low, adjustments were made by physically realigning the body of the dye laser. There are three external adjustable legs for height changes and horizontal changes were accomplished by sliding the dye laser on the table top. Internal adjustments were not made because the internal mechanism was stable and the misalignment was primarily due to the positioning of the argon beam. This procedure is described in the operating manual and will not be further discussed here. When the dye laser output was in excess of 70 milliwatts with both etalons or in excess of 200 milliwatts with one etalon, the etalon(s) were removed and the birefringent filter tuned to the proper sodium line. To accomplish this, the output was reflected off the first beam splitter and through a sodium cell as shown in Figure 4. The cell was then heated with a methane/air torch and the dye laser tuned. Maximum intensity
from the spontaneous emission was used to indicate proper tuning. This procedure was repeated for the etalons. The thin etalon was tuned first, but if the laser would not lase when the thick etalon was inserted, the thin etalon was removed. Adjustment of the thick etalon was then accomplished by using the small screw adjustment on the etalon mount while the etalon carriage which held both of the etalons was not moved. When the thin etalon was replaced, it was already aligned. Final tuning was then accomplished by rotating the carriage via a screw worm-gear combination.

A second method of tuning with the etalon(s) in place was to heat the cell until near total absorption occurred. The relative shortness of the resultant absorption "tail" indicated line center. This method was the most useful for tuning the thick etalon.

The rest of the system could now be aligned but the dye laser output usually drifted up to a few doppler widths. The frequency had to be checked every 15-30 minutes for the first 2-3 hours. After that the drift was very small and corrections became infrequent. These frequency shifts were small enough that it did not affect the alignment of the rest of the apparatus. After system alignment, the dye laser could be tuned by observing the intensity of the beam that passed through the sodium cell that was used to produce the echoes. This method for final tuning was superior to those mentioned above.

If this simple tuning did not produce good results, the etalons were removed and the birefringent filter retuned to line center and the etalons adjusted as before. It was not uncommon to retune the
dye laser in this manner in the first 2-3 hours.

When the above techniques were unable to produce sufficient output power, the dye laser was realigned internally. This procedure is described in the operating manual and only variations to that procedure are described here. It is assumed that the reader will have access to the manual in order to completely understand the following procedures which deal only with the internal adjustment of the laser and are too long for inclusion here.

The internal arrangement of the dye laser is shown in Figure 15. The removal of the optics, initial alignment of the laser body and remounting of the mirrors was accomplished according to the manual instructions. At this point instead of trying to find the focal distances produced by the mirrors before the output mirror was mounted, the output was reflected off a mirror and onto a wall 3.6 meters from the dye laser. The cavity mirrors were adjusted until the spots on the wall were each about 3 centimeters in diameter and superimposed. Both beams were checked to insure they came to focus between the laser and the wall similar to the procedure described in the manual. The output mirror was mounted, aligned for maximum intensity, and the beam centered in the cavity mirrors.

Eyeglasses designed to filter out the sodium line, such as those used in glass blowing, are useful here. To inspect the focal point of the argon laser in the jet stream underexposed film negatives were found to be useful in attenuating the output to tolerable levels.
Figure 15. The Jet Stream Dye Laser
At this time there were usually 1 or 2 spots on the ceiling that resulted from reflections off the jet stream. The positions of these spots were adjusted by moving the jet stream until there was only one spot which was in line with the laser axis at 26° from the vertical in the direction of the laser output. This was a difficult task as the jet was secured by a "friction" mount with the adjustments hard to control. A better holder is now available from the manufacturer.

If the output was not at least 700-800 milliwatts, the jet focus controls and mirror adjustments were varied slightly. Power outputs greater than 800 milliwatts were not normally obtained. Inserting the birefringent filter normally reduced the output to less than 500 milliwatts. With both etalons in an output of 150-200 milliwatts was considered good.

The Rhodamine 6G dye was normally replaced every six months although no evidence of degradation was observed.

The modes of the dye laser were analyzed by an L-20 Spectrum Analyzer in a Model 545A Tektronix Oscilloscope. With both etalons in place the output was usually single mode as long as the thick etalon was not perpendicular to the laser axis which caused a multi-mode output. Because the alignment was easier, the laser was often operated with the thick etalon removed. This resulted in a 2-3 mode output as determined by the spectrum analyzer, but did not seem to affect the production of the photon echoes.
Dye Amplifier Theory

The dye amplifier used in this experiment is similar in design to that described in Reference 23. A brief theory of operation is presented next followed by the alignment procedures used to produce maximum gain and reasonable beam quality.

A typical dye molecule energy level diagram is shown in Figure 16. The structure is composed of electronic states which have been broadened by the rotational and collisional levels. These levels lie very close together causing each electronic state to become a band of states. The relaxation time of the rotational and collisional levels is at least 3 orders of magnitude smaller than the typical electronic lifetimes of $10^{-9}$ seconds. Thus the molecule is normally in the bottom of a band of states. The triplet state is normally filled by the $S_1 - T_1$ transition which has a lifetime of an order of magnitude larger than the $S_1 - S_0$ transition. Thus for short pulses, like those from a nitrogen laser, the triplet states do not play an important role. The model is further simplified by ignoring the $S_2 - S_1$ transitions which are few in number.

To a good approximation the gain can be described by assuming a two level dye molecule with upper state $a$ and lower state $b$. The rate equations for transitions between these states are:

\[
\begin{align*}
\dot{n}_a &= - \dot{n}_b = n_b \sigma_p F_p - n_a A - n_a \tilde{\sigma}_k F_k \\
\frac{dF_k}{dz} &= n_a \sigma_k F_k \\
n_a + n_b &= n_0
\end{align*}
\]
Figure 16. Energy Level Diagram for a Typical Dye Molecule.
where \( n_a \) = density of the upper state
\( n_b \) = density of the lower state
\( n_0 \) = total dye density
\( \sigma_p \) = absorptive cross-section of the dye at 337 nanometers
\( \sigma_k \) = absorptive cross-section of the dye at 589 nanometers
\( k \) = identifies the \( k \)th mode
\( F_p \) = the number of photons per m\(^2\) per sec. of flux of the pump beam
\( F_k \) = flux of the \( k \)th mode in the cavity
\( A \) = Einstein coefficient.

The reaction time of the dye to changes in the incoming beam and pump beam is near \( 10^{-12} \) seconds which allows the above equations to be solved using steady state conditions. From Equations (3-1) with \( n_a = 0 \):

\[
n_a = \frac{\sigma_p F_p n_0}{A + \sigma_p F_p + \sum_k \sigma_k F_k}
\]  

(3-4)

In the linear gain region the number of photons in the cavity are much smaller than numbers of absorbed pump photons which means

\[
\sum_k \sigma_k F_k \ll \sigma_p F_p.
\]  

(3-5)

Substituting for \( n_a \) in Eq. (3-2) with the above yields

\[
\frac{dF_k}{dz} = \frac{\sigma_p F_p n_0}{A + \sigma_p F_p} \frac{\sigma_k F_k}{\sum_k \sigma_k F_k}
\]  

\[
= \frac{n_0 \sigma_k F_k}{1 + A/\sigma_p F_p}
\]  

(3-6)

Integrating this equation over \( z \) leads to:
\[ F_k(z) = F_k(0) \exp \left( \frac{n_0 \sigma_k z}{1 + A/\sigma_p F_p} \right) = F_k(0)e^{\alpha z} \]  

(3-7)

where \( \alpha \)

\[ \alpha = \frac{n_0 \sigma_k}{1 + A/\sigma_p F_p} \]  

(3-8)

Defining the Pump Power, \( P_p \), as:

\[ P_p = F_p \left( \frac{\hbar c}{\lambda_p} \right) L d \]  

(3-9)

where

\[ \lambda_p = N_2 \text{ pump wavelength} \]

\[ L = \text{length of gain region} \]

\[ d = \text{width of gain region} \]

and \( n_0 = c_d A_0 \) where \( c_d \) is the dye concentration and \( A_0 \) is Avogadro's number allows the gain term \( \alpha L \) to be expressed as:

\[ \alpha L = \frac{n_0 \sigma_p F_p \sigma_k L}{A + \sigma_p F_p} = \frac{c_d P_p}{d} \left( \frac{A_0 \sigma_p \lambda \sigma_k}{\hbar c} \right) \left( \frac{1}{\sigma_p \lambda} \right) \]  

(3-10)

From Reference 25, \( \sigma_p = 1.17 \times 10^{-21} \text{ meter}^2 \) at 337.1 nanometers and a typical value of \( \sigma_k \) is \( 10^{-20} \text{ meters}^2 \). This along with \( L = 2.5 \) centimeters and \( d = 0.5 \text{ millimeter} \) shows the expression \( \sigma_p \lambda / \hbar c L d \) is much smaller than \( A \) and can be ignored. Thus \( \alpha L \) can be simplified as

\[ \alpha L = (1.2 \times 10^{-8}) \frac{c_d P_p}{d} \]  

(3-11)

Changing units allows the equation to be written as
\[ \alpha L = 1.2 \frac{C_d P}{d} \]  

(3-12)

where \( C_d \) is in units of \( 10^{-3} \) kg-mole/mm\(^3\), \( P \) is in \( 10^2 \) kilowatts and \( d \) is in millimeters.

**Pulse Length of the Amplified Signal**

The pulse length of the amplified signal is an important parameter because of the short radiative lifetimes of the Na D states. According to Equation 1-73 the formation time of the echo is a function of the initial pulse lengths and the time separation of the initial pulses that enter the sample cell. Thus with the apparatus used in this experiment the minimum echo formation time is nearly equal to or greater than the 16 nanosecond lifetimes of the Na D states. Shortening the pulse length of the dye amplifier output will then decrease the time needed for echo formation and correspondingly increase the echo intensity.

The intensity of the amplified pulse as a function of time can be calculated from the gain equation

\[ G(t) = e^{\alpha L t} \]  

(3-13)

where \( \alpha L = 1.2 \frac{C_d P}{d} \) as given in Equation (3-11). Since \( P_P(t) \) appears in the exponential the gain is a strong function of the pump pulse and the amplified signal will be much narrower than the pump. For purposes of this calculation the shape of the nitrogen pulse can be approximated by

\[ g(t) = K t e^{-\alpha t} \]  

(3-14)

where \( K = 6.30 \times 10^8 \) sec\(^{-1}\), \( \alpha = 2.5 \times 10^8 \) sec\(^{-1}\) and \( g(t_{\text{max}}) = 1. \)
In this approximation \( t_{\text{max}} \), the time when the pump pulse is at its maximum, is 4.0 nanoseconds and the half power points occur at \( t = 0.9 \) and 10.7 nanoseconds.

For the amplified pulse the half power points can be determined from the definition

\[
G(t_{1/2}) = G(t_{\text{max}}) \tag{3-15}
\]

Using this definition and \( G(t_{\text{max}}) = 10^3 \) then \( \Delta t_{1/2} \), the pulse length of the amplified signal, is 3.4 nanoseconds which corresponds to the time when the pump pulse is within 10% of its maximum output. For \( G(t_{\text{max}}) = 10^4 \), \( \Delta t_{1/2} = 3.0 \) nanoseconds and the pump pulse is within 7.5% of its maximum. Finally for \( G(t_{\text{max}}) = 10^5 \) the values are \( \Delta t_{1/2} = 2.8 \) nanoseconds and 6.5%. From this it is also clear that the amplified pulse length is a function of the gain and the shape of the nitrogen pulse when it is near its peak.

If the gain of the amplifier is too high then it will saturate on its own amplified spontaneous emission and become useless. To analyze this behavior consider a gain region with length \( L \) and cross-sectional area \( A' \) where \( A' \ll L^2 \) as shown in Figure 17. The number of spontaneously emitted photons leaving the volume element \( A' \text{d}t \) per second is

\[
\beta_0 \text{d}z = \frac{n_0 A'}{A} \text{d}z \tag{3-16}
\]

If \( \gamma \) is the number of photons per second passing through the gain region at \( z = L \) then

\[
\gamma = \beta_0 \int_0^L dz G(L-z) \frac{\Omega(z)}{4\pi} \tag{3-17}
\]

where \( G(L-z) = \exp \{a(L-z)\} \), \( a = n_0 \sigma \), and \( \Omega(z)/4\pi \) is the fraction of
Figure 17. Diagram for Determining the Amplified Spontaneous Emission Flux.
the spontaneous emission that will pass through the gain region at 
\( z = L \). For \( z \ll L \),
\[
\frac{\Omega(z)}{4\pi} = \frac{\Lambda'}{4\pi(L-z)^2}
\]  
(3-18)
and the integral can be written as
\[
\gamma = \frac{\Lambda'}{4\pi} \beta_0 \int_0^{L-\delta'} dz' e^{\alpha(L-z')/(L-z')^2} + \beta_0 \int_{z'}^{L} d\zeta e^{\alpha(L-z)\Omega(\zeta)/4\pi}
\]  
(3-19)
where \( 0 < z' < L \). The first integral in the above becomes approximately
\[
\int_0^{L-\delta'} dz' \frac{e^\alpha L e^{-\alpha z}}{L^2(1 - z/L)^2}
\]  
\[
\approx \frac{e^\alpha L}{\alpha L^2} (1 - e^{-\alpha z'})
\]  
(3-20)
and
\[
\gamma \approx (1/\alpha)\beta_0 \frac{G(L)\Omega(\delta)}{4\pi}.
\]  
(3-21)
The contribution from the second integral in Equation (3-19) is small compared to the first integral and is ignored. The characteristic 
length \( \delta \) is defined as \( \alpha^{-1} \) and corresponds to the length of the gain
region from \( z = 0 \) that determines \( \gamma \) at \( z = L \).

At saturation the increase of photons in the amplified pulse
per second as the pulse travels a short distance, \( \zeta \), is equal to the
number of pump photons absorbed over the same distance or
\[
(1/\alpha)\beta_0 \frac{\Omega}{4\pi} \{ e^{\alpha(L+\zeta)} - e^{\alpha L} \} = \gamma_p \zeta/L.
\]  
(3-22)
\( \gamma_p \) is the total number of pump photons absorbed per second and
\( \gamma_p = \beta_0 L \). With this the condition which must be met in order to
avoid self-saturation of the dye amplifier is
\[
G \frac{\Omega}{4\pi} \leq 1.
\]  
(3-23)
When \( d = 0.5 \) millimeter and \( L = 25 \) millimeters the gain limit is \( G = 4 \times 10^4 \) where \( aL = 10.6 \). When \( P_p \) is \( 2 \times 10^5 \) watts the maximum allowed concentration of the Rhodamine 6G dye is \( 2.2 \times 10^{-3} \text{kg-mole/m}^3 \).

The amplified pulse length was shortened even further by extending the length of the dye amplifier cavity to 0.70m. This caused the first pass through the dye cell to return to the cell 5 nanoseconds later. The trailing edge of the first pulse arrives at the cell after the pump power has passed its peak and is decreasing rapidly. This causes the trailing edge to have less amplification and hence shortens the pulse.

In this experiment \( c_d = 1.3 \times 10^{-3} \text{kg-mole/m}^3 \) which predicts a single pass gain of 500 or a double pass gain of \( 2.5 \times 10^4 \). The actual gain was \( 10^4 \) corresponding to an input of 5 milliwatts and an output of 50 watts. The penetration depth was 1.1 millimeters which allowed reflection of the pump beam off the back of the cell which gave a more uniform gain region.

**Alignment of the Dye Amplifier**

The output of the dye laser passed through a beamsplitter (\( R = 94\% \)), through a 1 meter focal length lens located just after the beamsplitter and came to focus at the dye cell located 0.85 centimeters from the lens. See Figure 4. The lens and beamsplitter were mounted on an X-Y translator. A 13X telescope expanded the beam which was then reflected back through the telescope by a 2380 line/millimeter grating. The grating had a blaze angle corresponding to 500 nanometers and was used
in the Littrow configuration. After passing through the dye cell for the second time, it passed through the lens and was reflected by the beamsplitter towards a 60° prism. As it transversed the prism, the spontaneous emission noise, which had a predominately shorter wavelength than the amplified beam, was angularly separated from the output. The spatial filter allowed the amplified signal to pass but blocked most of the spontaneous emission. When measured by the high speed detector (discussed later), the signal-to-noise ratio was over 100. Finally, the beam passed through a 1 meter focal length collimating lens and entered the beam delay line.

To align the amplifier, the focusing lens behind the beamsplitter was removed and the dye cell adjusted until the dye laser beam passed through the cell. The lens was inserted and transversely adjusted until the beam again passed through the cell. The position of the beam could be seen as it entered the dye. The beam should be in the center of the holes and not reflecting off the glass. Adjustments were made by three adjusting screws on the platform that held the dye cell, telescope, and diffraction grating assembly. Next, the telescope was removed and the beam centered in the diffraction grating. Replacing the telescope should not have moved the spot. When it did, the three adjusting screws on the dye amplifier platform were again adjusted. Next, the grating was adjusted until the beam passed back through the cell and was reflected off the beamsplitter. If a small intense dot did not form at a distance from the beamsplitter equal to the distance from the beamsplitter to the dye laser, the telescope was adjusted.
It was also convenient to allow the output to fall on a screen a few meters away to inspect the beam quality. Beam quality could also be checked by observing the pattern on the diffraction grating or the reflection off the grating which hit the ceiling. The beam striking the grating should be collimated.

The N\textsubscript{2} laser was adjusted by use of the three adjustable feet on the N\textsubscript{2} laser platform. The height of the beam was adjusted by raising or lowering the front "foot". The N\textsubscript{2} beam could be rotated slightly by rotating the cylindrical lens which focused the output into a thin line. The 6 inch focal length cylindrical lens was placed 14.3 millimeters in front of the dye in the dye cell causing a beam width of 1/2 millimeter. Scribe marks on the front of the dye cell located the position of the N\textsubscript{2} beam. The high speed detector could also be used to verify alignment.

Alignment of the prism and spatial filter was straightforward. The spatial filter was 1 millimeter in diameter.

**Beam Delay Line and Focusing Optics**

Figure 4 shows the beam delay line which was used to divide a single incoming pulse into two non-synchronous collinear pulses with a 1:4 intensity ratio. The beamsplitters used in the delay line were antireflectively coated windows for 1.15 micrometer radiation incident at 45°. At the Na D lines the reflectivity of these coatings is a strong function of the incident angle which allowed the intensity ratio of the pulses to be easily adjusted. This was accomplished by translation of the second mirror in the
delayed beam path which changed the incident angle of the second beamsplitter.

The beams which are about 2 millimeters in diameter at this point were expanded by the lens combinations $L_1 L_3$ and $L_2 L_3$. $L_1$ and $L_2$ were translated along the beam path to make the respective pulses convergent, divergent, or plane wave. For near plane wave pulses $L_1$ and $L_2$ were $+10$ diopters lens and $L_3$ was $+2$ diopters. Shorter radii of curvature pulses were created by using $+5$ to $+40$ diopter lens for $L_1$ and $L_2$.

As the beam expanders were defocused the spot size and hence the intensity of the radiation as it entered the cell varied. It was determined by numerous trials that a pulse ratio of $1:2$ to $1:8$ was normally capable of producing echoes which allowed minor adjustments of $L_1$ and $L_2$. For larger adjustments neutral density filters with transmission coefficients of $12.5$, $25$, and $50\%$ were used to maintain a good pulse ratio.

Finally the overall intensity was adjusted by using crossed polaroids located just before the Na cell. The first polaroid was vertically oriented and the second allowed to rotate which provided a large range of pulse intensities.

Alignment of the two beams was not difficult particularly when the pulses were not collinear. To align the beam delay line, the lenses $L_1$ and $L_2$ were removed and the beam adjusted so that the first pulse passed through the center of both beam splitters. The first beam splitter was adjusted so the reflected beam struck the center of
the first mirror. The mirrors were adjusted so the beam passed through the second beamsplitter and superimposed on the first pulse which was reflected by the second beam splitter. This was checked two ways. First, a card was placed 5-10 centimeters from the beamsplitter and the spots overlapped by adjusting the last mirror. Second, the card was replaced with a mirror and the beam reflected on the wall several meters away. The second beamsplitter was adjusted to overlap the spots. To help alignment in both cases, the second beam was blocked and unblocked while adjustments were made until the two spots were overlapped. If the image appeared to move, the spots were not overlapped. A more accurate method, which was not necessary for this experiment, was to place a long focal length lens (generally 1 meter) several meters from the second beamsplitter. When poorly aligned, this procedure produced two spots in the focal plane. Adjustment of the second beam splitter until the spots were overlapped yielded an angular separation of beams less than $10^{-4}$ radians.

$L_1$ and $L_3$ were inserted into the system and adjusted so that the unexpanded beam was collinear with the expanded first pulse. $L_2$ was inserted and adjusted for collinearity.

Circular polarization was produced by placing a Mica $\lambda/4$ plate directly in front of the Na oven. This plate was designed for 623.9 nanometers which is close enough to the Na D lines to produce a polarization very close to circular.
Sodium Oven and Cell

The operation of sodium oven and cell were relatively free of difficulties. It was noticed that the cell could be brought up to operating levels easily by initially overheating the cell until near total absorption of the beams occurred. Allowing the cell to cool to about 170°C yielded about 10-20% transmission which gave good echo formations. The evacuated oven was pumped out at the start of each day and the leak rate of 3 torr/hour did not seem to cause trouble during the day.

Trouble Shooting

When the system appeared to be working properly but no echoes appeared there were several causes that were checked which normally corrected the difficulty. Based on these experiences the following troubleshooting guide was written.

1) Insure that both beams are properly aligned and pass through the cell. Place a card after the sodium cell and see if both beams fill the aperature in the oven leaving round spots on the card. This is most easily checked by blocking one beam then the other. With a little practice large variations in the pulse ratios can also be detected in this way.
2) Although originally centered on the proper sodium line the dye laser may have drifted off resonance. To easily check this hold a card just after the sodium oven and adjust the etalon control on the dye laser for minimum transmitted intensity. This should be less than 1-2 turns.

3) If there is no change in intensity or if it was very close to resonance check the sodium cell visually via the slit on the side of the oven. The cell should be bright and uniformly glowing from the spontaneous emission. If it is brighter at the entrance of the cell then at the exit or if the cell is dim then the cell temperature needs adjustment.

4) When the first three steps have been taken and still no echo is seen the most probable cause is low intensity of the initial pulse from the amplifier. When the intensity is high a flickering should be easily noticed in the focused spots of the pulses. This is due to nonlinear effects in the sodium and uneven amplification of the laser signal. An absence of this flickering usually indicates the nitrogen laser needs adjustment. The high speed detector is useful at this point.
Echo Measurements

To measure the radii of curvature, \( R \), of the pulses in a straightforward manner a 1 meter focal length lens, \( L_4 \), was placed directly behind the Na cell oven. The focal points were then located by moving a white card along the beam until the best focus was found. A tape measure was used and then the image distances, \( q \), were recorded. From these measured \( q \)'s the corresponding \( R \)'s were calculated from

\[
R = \frac{1}{1/q} - 1 + \Delta z
\]  

(3-18)

where \( \Delta z \) was the distance from the Na cell to \( L_4 \).

To measure the diameter and angular separation of the beams a screen constructed from 2 millimeter per division graph paper was employed. The diameters were determined by comparing the size of the spots to the known width of the graph lines. The angular separation was calculated by dividing the spatial separation of the spots on the screen by the image distance, \( q \).

The high speed detector and 7904 oscilloscope was used to analyze the pulse structure and measure the formation time of the echo. Triggering of the oscilloscope was caused by a second high speed detector placed in the beam delay line as shown in Figure 4. By moving the detector slightly the oscilloscope could be triggered on either pulse. When triggered in this manner the nonsynchronous initial pulses and echo always appeared at the same respective position on the oscilloscope screen and thus served to identify them. This was necessary because only one pulse was usually seen on the
screen at a time and an easy means of identification and making occurrence time measurements was needed.
CHAPTER IV
EXPERIMENTAL RESULTS

Visual Radii of Curvature Measurements

The first photon echoes were discovered with the use of the high speed detector. The initial pulses were plane waves at an angle of 3-5 milliradians and overlapped as they passed through the Na cell. With $L_4$ in place, the focal points of the pulses were located and the sensitive area of the detector positioned at the expected focal point of the echo. Various parameters such as tuning, cell temperature, and pulse intensity were varied until a small signal, which occurred 8 nanoseconds after the second pulse, was detected. A white card was placed in front of the detector and a third dot appeared as shown in Plate 9. When either of the initial pulses were blocked, the third pulse disappeared which confirmed it was an echo. Finally, $L_4$ was removed, the first pulse made divergent, and the echo came to focus in a manner expected of a phase conjugated beam as shown in Figure 2.

Subsequently, photon echoes were observed with focal lengths measuring from 0.86 meters to greater than 6 meters. With $L_4$ in place, both positive and negative radii of curvatures were investigated and found to obey the general relation

$$\frac{1}{R_E} = \frac{2}{R_2} - \frac{1}{R_1}.$$  \hspace{1cm} (1-45)
Plate IX. Photograph of Non-Collinear Plane Wave Initial and Echo Pulses Focused by a One Meter Focal Length Lens. The First Pulse is on Top, the Second is in the Middle, and the Echo is on the Bottom.
The radii of curvature calculated from these measurements are found in Tables 1-4. The data tables are separated according to the polarization of the initial pulses and the Na D line used. Data within the tables are arranged according to the magnitude and sign of \( R_1 \) and \( R_2 \). Radii of curvatures which were measured directly without the use of \( L_4 \) are labeled as \( R_{\text{ED}} \).

The computed values of \( R_E \) were calculated from the measured values of \( R_1 \) and \( R_2 \) and are listed under \( R_{\text{EC}} \). Variations between the values \( R_E \) and \( R_{\text{EC}} \) are listed under \( \eta \) where

\[
\eta = \frac{q_E - q_{\text{EC}}}{q_E} \times 100 .
\]

(4-1)

\( q_E \) and \( q_{\text{EC}} \) are the respective focal distances with \( L_4 \) in place and \( q_{\text{EC}} \) was calculated from Equation 3-18. \( \eta \) was calculated in terms of \( q \) instead of \( R \) to avoid the singularity when \( q \) was close to the focal length of \( L_4 \).

The radii of curvature of the initial pulses varied from infinity to slightly less than one meter, a limit imposed by the geometrical arrangement of the beam delay line. The corresponding radii of curvature of the echoes varied from infinity to the shortest recorded at \(-0.77 \) meters. Echoes with shorter radii were observed but the proximity of the second pulse made accurate measurements difficult. Values of \( R_{\text{ED}} \) greater than 3 meters were also observed but not recorded because of the larger uncertainties in the focal lengths.
Table 1. Radii of Curvature Measurement for Linear Polarization and the $^2S_{1/2} - ^2P_{3/2}$ Transition. $R_1$, $R_2$, and $R_E$ are the Radii for the First, Second, and Echo Pulses, Respectfully. $R_{ED}$ is the Directly Measured Radii of the Echo, and $R_{EC}$ is the Radii of Echo Calculated from $R_1$ and $R_2$. $\eta$ is as defined in Equation (4-1) and Represents the Variation between $R_E$ and $R_{EC}$.

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Table 2. Radii of Curvature Measurement for Linear Polarization and the $^2S_{1/2} - ^2P_{3/2}$ Transition. $R_1$, $R_2$, and $R_E$ are the Radii for the First, Second, and Echo Pulses, Respectfully. $R_{ED}$ is the Directly Measured Radii of the Echo, and $R_{EC}$ is the Radii of the Echo Calculated from $R_1$ and $R_2$. $\eta$ is as defined in Equation (4-1) and Represents the Variation between $R_E$ and $R_{EC}$.

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Table 3. Radii of Curvature Measurement for Circular Polarization and the \(^2S_{1/2} \rightarrow ^2P_{1/2}\) Transition. \(R_1\), \(R_2\) and \(R_E\) are the Radii for the First, Second, and Echo Pulses, Respectfully. \(R_{ED}\) is the Directly Measured Radii of the Echo, and \(R_{EC}\) is the Radii of the Echo Calculated from \(R_1\) and \(R_2\). \(\eta\) is as defined in Equation (4-1) and Represents the Variation between \(R_E\) and \(R_{EC}\).

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Table 4. Radii of Curvature Measurements for Circular Polarization and the $^2S_{1/2} - ^2P_{3/2}$ Transition. $R_1$, $R_2$, and $R_E$ are the Radii for the First, Second, and Echo Pulses, Respectively. $R_{ED}$ is the Directly Measured Radii of the Echo, and $R_{EC}$ is the Radii of the Echo Calculated from $R_1$ and $R_2$. $\eta$ is as defined in Equation (4-1) and Represents the Variation between $R_E$ and $R_{EC}$.

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The quality of the focusing action of the Na vapor was also investigated by measuring the sizes of the focused pulses. With \( L_4 \) in place a screen was translated along the beam until the minimum diameter or focal point was found. Comparing the spot size to the known linewidth of the 2mm/div graph paper, which served as the screen, gave a rough measurement of the spot size.

Using this technique for the case of plane wave pulses the typical minimum diameter of the initial pulses and echo were between 150 and 250 micrometers. This is much larger than the minimum possible spot size of a few micrometers and reflects the imperfections in the beam quality of the initial pulses and the effects of the distortions in the cell windows. The important effect to be noted here is that the echo size was comparable to the size of the initial pulses which indicates that the focusing action of the sodium was at least as good as the beam quality of the initial pulses. The spot sizes of different radii of curvature pulses were also measured. The spot size depended on the focal length of the respective beam being measured but again the spot size of the echo was comparable to the spot size of one of the initial pulses with the same focal length.

**Effect of Polarization and D Line Selection**

As shown in Tables 1-4, photon echoes were produced on both D lines and with linear and circular polarization. When the initial
pulses were linearly polarized the echo was linearly polarized in the same direction. When circular polarization was used, the echo was also circularly polarized. These parameters had no apparent effect on the focusing of the echoes which is in agreement with Chapter I and the previous work done in this laboratory with SF$_6$ and SiF$_4$.$^{7,8,21}$

**Measurements Using the High Speed Detector**

The high speed detector and 7904 Tektronix oscilloscope, which was triggered by the second pulse, was used to analyse the shape, occurrence time, and relative heights of the pulses. A detector risetime of less than 2 nanoseconds allowed good pulse resolution and pulse shapes for all but the weakest echoes. These could be seen visibly but were too weak to register on the oscilloscope.

The low average intensity of the pulses were below the threshold intensity of available power meters but could be measured by the high speed detector. With $L_4$ in place the echo and second pulse were alternately focused on the photo diode and the resulting pulse heights compared. Assuming the response of the detector was linear, the resulting intensity ratios were typically 1:100 to 1:120 for well formed echoes. The largest intensity ratio recorded was 1:80 corresponding to 25 milliwatts/cm$^2$.

Photographs of the oscilloscope traces were taken with a polaroid camera. Because of the slow nitrogen laser repetition rate, exposure
times of 20 seconds were used. The resulting photographs then represent an average of 400 traces and are a good measure of pulse repeatability.

Traces of the initial pulses are shown in Plate X. In the configuration used in this experiment the pulse lengths were 5 nanoseconds long when measured from the half power points and the pulse heights were in a 1:4 ratio. The pulses were not completely separated as the first pulse had only dropped to about 10% of its maximum value when the second pulse began. In this photograph the time scale per division is 10 nanoseconds which implies peak-to-peak pulse separation of 8 nanoseconds.

Because of the comparatively low intensity of the echo it was necessary to separate it from the initial pulses in order to study it in detail. In Plate XI a small fraction of the second pulse was allowed to overlap the echo in order to measure the occurrence time of the echo easily. From this photograph, which is a representative case, the peak-to-peak time separation between the second pulse and echo is 8 nanoseconds. Plate XII shows the echo by itself. In general it was difficult to achieve a good pulse-to-pulse repeatability as shown here. A pulse height fluctuation of 10-20% was normal. In some cases, particularly for high Na cell absorptivity, the peak heights varied by a factor of 2-3.
Plate X. Oscilloscope Trace of the Initial Pulses. The Time Scale is 10 Nanoseconds per Division.
Plate XI. Oscilloscope Trace of Second Pulse and Echo. Second Pulse is Approximately 1/100 of Its Normal Size. The Time Scale is 10 Nanoseconds per Division.
Plate XII. Oscilloscope Trace of the Echo. The Time Scale is 10 Nanoseconds per Division.
Angular Position of the Echo
and Phase Matching Results

As expected \(^2\) the angular position of the echo was determined by the relation \( \hat{k}_E = 2\hat{k}_2 - \hat{k}_1 \). In particular when the second pulse was at an angle \( \theta \) to the first, then the echo appeared at an angle of \( 2\theta \) as shown in Plate IX. This positional relationship was often used to locate the echo when it was weak.

The angular separations of the beams was accurately measured for the case of near-plane wave pulses. \( L_4 \) was left in place and the beams were brought to focus on 2 millimeter/division graph paper which served as the screen. As the angle between the pulses was increased, the intensity of the echo decreased. When this experiment was repeated many times, an upper limit of 7 millimeters or 7 milliradians was consistently observed. Beyond this the echo was very faint or non-existent.

The angular spread is about twice that calculated in Chapter I, which can be accounted for by the high absorptivity of the Na vapor that was used to generate the echoes. This was often around 90\% which indicates that the initial pulse intensity had dropped by 50\% when the pulse had propagated through the first quarter of the cell. If this intensity is used as the cutoff point for echo production, then the effective length of the cell is 6.4 millimeters. This corresponds to a theoretical \( \theta_{\text{max}} \) from Equation 1-81 of 6.8 milliradians which is very close to the experimental value of 7 milliradians.
Auxiliary Pulses

During the course of the experiment the beam attenuator located just before the Na oven was removed which increased the intensity of the pulses entering the Na cell by a factor of 3-4. The result was the formation of two auxiliary pulses or spots as shown in Plate XIII. Also, an oscilloscope trace of the echo pulse showed the existence of an additional pulse superimposed over the echo which occurred 10 nanoseconds earlier as shown in Plate XIV. Plate XII, the echo alone at normal intensity, and Plate XIV were taken within 3 minutes of each other with the only change being the removal of the beam attenuator. The oscilloscope was triggered by the second pulse so the time positions on the screen are comparable. Subsequent investigations revealed that the relative heights of the pulses could be changed by alternating the intensity ratios of the initial pulses.

A proposal for explaining the appearance of the auxiliary pulses at high initial pulse intensity was sum-frequency mixing due to the timewise overlapping of the initial pulses and the second pulse and the echo. The overlap between the initial pulses was discussed earlier while the overlap between the second pulse and echo is evident from Plate XIV. This photograph shows the echo and a small fraction of the second pulse. If the second pulse were displayed at its true intensity, the tail would easily overlap the echo.
Plate XIII. Photograph of Non-Collinear Plane Wave Initial, Echo, and Auxiliary Pulses Focused by a One Meter Focal Length Lens. The Bright Center Dot is the Second Pulse while the First is on the Right and the Echo is on the Left. The Outside Dots are the Auxiliary Pulses.
Plate XIV. Oscilloscope Trace of Echo Pulse When Initial Pulses are Overly Intense. The First Pulse is the Auxiliary Pulse and the Second is the Echo. The Time Scale is 10 Nanoseconds per Division.
A further indication of sum-frequency mixing is shown in Plate XV. This is an oscilloscope trace of the auxiliary pulse which is outside of the echo pulse. The pulse occurs at the same time or slightly later than the echo. A corresponding trace of the auxiliary pulse located outside the first pulse could not be made because the pulse was too weak. An additional pulse located farther out than the pulse outside the echo was also seen but was even weaker. The only data recorded on this auxiliary pulse was its position. Its angular separation was the same as for any of the other adjoining pulses thus resulting in five equally spaced spots.

To test this sum-frequency proposal the time delay between the pulses was reduced to 1 nanosecond which effectively overlapped the pulses timewise. The effect was dramatic as five new pulses appeared as shown in Plate XVI! When either of the initial pulses was blocked, the pattern disappeared. Further, the radii of curvature of the spots was a function of the curvature of the initial pulses with the greatest effect seen in the pulses farthest out.

Based on the above results and Plates XIV and XV, it was concluded that the auxiliary pulses were due to sum-frequency mixing. In particular, the pulse which overlapped the echo and the pulse located outside the first pulse were due to the overlap of the initial pulses. Further, the pulse located outside the echo was due to the overlap of the second pulse and the echo.
Plate XV. Oscilloscope Trace of the Auxiliary Pulse Located Outside the Echo. The Time Scale is 10 Nanoseconds per Division.
Plate XVI. Photograph of Simultaneous, Non-Collinear Plane Wave Initial Pulses and Resultant Auxiliary Pulses Focused by a One Meter Focal Length Lens. The Bright Center Dot and the Dot Immediately to the Right are the Initial Pulses.
CHAPTER V
COMPUTER SIMULATION: DAMPING EFFECTS

In Chapter I the approach taken to describe the focusing of photon echoes yielded the proper phase and intensity information in a straightforward manner but did not include damping effects. In this chapter an alternate approach using phenomenological equations is used to describe the production of photon echoes as a function of the degree of off-resonance of the pulses and the lifetimes of the states involved. The results are compared with those of Chapter I and for the conditions for which the photon echoes were produced in this experiment. This development is similar to that presented in Reference 27.

Phenomenological Approach to Echo Production

The response of a two-level, non-degenerate atom or molecule with upper level a and lower level b can be studied by using the density matrix representation of the Schrödinger equation,

\[ \text{i}h \dot{\sigma} = H \sigma - \sigma H + \dot{\sigma}_r \]  

(5-1)

where \( \sigma \) is the density matrix, \( \dot{\sigma} \) is the total time derivative, and \( \dot{\sigma}_r \) is the partial time derivative of \( \sigma \). The perturbation is expressed in the dipole approximation as
\[ v = -\mathbf{P} \cdot \mathbf{E} \]  \hspace{1cm} (5-2)  

where \( \mathbf{P} \) is the polarization operator and the electric field \( \mathbf{E} \) is 
\[ \mathbf{E} = \mathbf{\hat{u}} \mathbf{E}(y, t) e^{i(ky - \omega t)} e^{i\phi} + c.c. \]  \hspace{1cm} (5-3)  

where 
\[ \psi = \phi + ky - \omega t \]  \hspace{1cm} (5-4)  

and 
\[ \phi = \phi_0 + \dot{\phi} t . \]  \hspace{1cm} (5-5)  

In this analysis only plane waves are considered and \( \phi_0 \) represents the initial phase. For convenience, let \( \mathbf{\hat{u}} = \mathbf{\hat{z}} \) and \( \Delta J = 1 \), then \( E = E^* \) and the matrix elements \( (b|\overrightarrow{\mathbf{P}} \cdot \mathbf{\hat{u}}|a) = (a|\overrightarrow{\mathbf{P}} \cdot \mathbf{\hat{u}}^*|b) \) are real. The phenomenological lifetimes are defined as 
\[ \frac{(b|\sigma_r|b) - (a|\sigma_r|a)}{(b|\sigma_0|b) - (a|\sigma_0|a)} = \frac{\left\{ (b|\sigma|b) - (a|\sigma|a) \right\} - (b|\sigma_0|b) - (a|\sigma_0|a)}{T_1} \]  \hspace{1cm} (5-6)  

and 
\[ (a|\sigma|b) = -\frac{(a|\sigma|b)}{T_2} \]  \hspace{1cm} (5-7)  

where \( \sigma_0 \) is the initial density matrix. Since this system is non-generate, the contraction \( \sigma_{ab} = (a|\sigma|b) \) is also used. Substituting the above into the Schrödinger Equation yields
\[
\dot{\sigma}_{ab} = -i\omega_{ab}\sigma_{ab} + \frac{E}{i\hbar}(a|\vec{P}\cdot\vec{u}|b)\{e^{i\psi} - e^{-i\psi}\}(\sigma_{bb} - \sigma_{aa}) - \frac{\sigma_{ab}}{T_2} \tag{5-8}
\]

In the rotating wave approximation, the substitution

\[
e^{i\psi} - e^{-i\psi} = e^{-i\psi}(e^{2i\psi} - 1) \tag{5-9}
\]

is used and the rapidly oscillating term, exp (2i\psi), is assumed to have a small effect and is ignored. With this approximation \(\dot{\sigma}_{ba}\) is written as

\[
\dot{\sigma}_{ab} = -(i\omega_{ab} + \gamma_2)\sigma_{ab} + \frac{iKE}{2} e^{-i\psi}(\sigma_{bb} - \sigma_{aa}) \tag{5-10}
\]

where

\[
\gamma_2 \equiv 1/T_2 \text{ and } \frac{K}{2} \equiv \hbar^{-1}(a|\vec{P}\cdot\vec{u}|b).
\]

The difference \(\sigma_{aa} - \sigma_{bb}\) can also be evaluated using the Schrodinger equation with the result

\[
\dot{\sigma}_{aa} - \dot{\sigma}_{bb} = iKE[e^{i\psi}\sigma_{ba} - \sigma_{ab}e^{-i\psi}] - \frac{(\sigma_{aa} - \sigma_{bb}) - (\sigma_{aa} - \sigma_{bb})}{T_1}. \tag{5-11}
\]

The conservation of probability requires

\[
\sigma_{aa} + \sigma_{bb} = 1 \tag{5-12}
\]

and the definition of \(\sigma_{ba}\) implies

\[
\sigma_{ab}^* = \sigma_{ba} \tag{5-13}
\]

These are the general equations which must be solved in order to gener-
ate photon echoes.

To solve the equations the phase dependence is removed by introducing the following substitutions

\[ A_1 = \sigma_{ab} e^{-i\psi}, \]
\[ A_2 = A_1^* = \sigma_{ba} e^{i\psi}, \]
\[ A_3 = \sigma_{aa} - \sigma_{bb}, \]  \(5-14\)

and

\[ \Delta = \omega_{ab} - \omega + \phi. \]

The result may be put into matrix form as

\[
\begin{pmatrix}
\dot{A}_1 \\
\dot{A}_2 \\
\dot{A}_3
\end{pmatrix} =
\begin{pmatrix}
i\Delta + \gamma_2 & 0 & iKE/2 \\
0 & -i\Delta - \gamma_2 & -iKE/2 \\
iKE & -iKE & \gamma_1
\end{pmatrix}
\begin{pmatrix}
A_1 \\
A_2 \\
A_3
\end{pmatrix}
\]

\[ + \gamma_1 A_3(t_0) \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \] \(5-16\)

where \( \gamma_1 \equiv 1/T_1. \)

Finally, this may be put into dimensionless form by using the following definitions:

\[ X \equiv \Delta/KE, \]
\[ Y \equiv \gamma_1/KE, \]
\[ Z_1 \equiv \gamma_2 / KE , \]

and

\[ KEt \equiv T . \]  \hspace{1cm} (5-17)

The result is

\[
\begin{pmatrix}
    A_1 \\
    A_2 \\
    A_3
\end{pmatrix}
\frac{d}{dT}
\begin{pmatrix}
    A_1 \\
    A_2 \\
    A_3
\end{pmatrix}
=
\begin{pmatrix}
    iX+Z & 0 & i/2 \\
    0 & -iX+Z & -i/2 \\
i & -i & Y
\end{pmatrix}
\begin{pmatrix}
    A_1 \\
    A_2 \\
    A_3
\end{pmatrix}
+ Y A_3(T)
\begin{pmatrix}
    0 \\
    0 \\
    1
\end{pmatrix} \hspace{1cm} (5-18)
\]

or simply

\[
\frac{d}{dT} A = -BA + C . \]  \hspace{1cm} (5-19)

A solution to this first order differential equation is

\[
A(T-T_0) = A_s + e^{-B(T-T_0)} A(T_0) - A_s \]  \hspace{1cm} (5-20)

where \( E \) is assumed constant. \( A_s \) is the steady state solution when

\[
dA/dT = 0 \text{ and } \]

\[
A_s = B^{-1} [Y A_3(T_0) \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}] . \]  \hspace{1cm} (5-21)

By direct calculation of the above

\[
A_{s1} = \frac{Y(X+iZ)}{Z+Y(X^2+Z^2)} , \]

\[
A_{s2} = A_{s1}^* = \frac{Y(X-iZ)}{Z+Y(X^2+Z^2)} , \]
Figure 18. The Photon Echo Sequence.
and
\[ A_{3} = \frac{X^{2} + Z^{2}}{Z + (X^{2} + Z^{2})} \] (5-22)

where the subscripts refer to the steady state solution for \( A_1 \), \( A_2 \), and \( A_3 \), respectfully.

At this point an expression for \( A(T) \) has been developed for \( T > T_0 \) and \( E \) constant. Specifically, this is

\[ m (i) - b (t - T_q) \]

\[ A (T-T^*_1) = A + e U \] (5-23)

where

\[ A_1 (T_0) = A_2 (T_0) = 0 \]

and

\[ A_3 (T_0) = -1 \] (5-24)

This corresponds to the time during the first pulse of the photon echo sequence as shown in Figure 18. The superscripts identify the time interval of the sequence for which the expression of \( A \) is valid.

As \( T \) increases beyond the first pulse and \( T_1 < T < T_2 \), \( A(t) \) becomes

\[ A^{(0)} (T-T_1) = A_s^{(0)} e^{-B_0 (T-T_1)} [A^{(0)} (T_1) - A_s^{(0)}] \] (5-25)

where

\[ A^{(0)} (T_1) = C_1 A^{(1)} (T_1 - T_0) \] (5-26)

Note \( T_1 \) and \( T_2 \) now represent points in time while the lifetimes \( T_1 \) and \( T_2 \) will be given in terms of their reciprocals, \( \gamma_1 \) and \( \gamma_2 \). \( C_1 \) is a 3 \( \times \) 3 matrix which is needed to preserve the continuity of \( \sigma_{ab} (T) \) at
the discontinuity at $T = T_1$. Specifically,

$$\mathbf{c}_1 = \begin{pmatrix} i[\psi_1(T_1) - \psi_0(T_1)] & 0 & 0 \\ 0 & e^{-i[\psi_1(T_1) - \psi_0(T_1)]} & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (5-27)$$

where $\psi$ is as previously defined. Also from Equation (5-26) it is clear that $A$ may not be continuous across the discontinuities in $E$, but $\sigma_{ab}(T)$ is continuous for all $T$.

Continuing through the pulse sequence in a similar manner yields:

**$T_2 < T < T_3$**

$$A(2)(T-T_2) = A_s(2) + e^{-B_2(T-T_2)} [A(2)(T_2) - A_s(2)] \quad (5-28)$$

$$A(2)(T_2) = C_2 A(0)(T_2 - T_1) \quad (5-29)$$

$$\mathbf{c}_2 = \begin{pmatrix} e^{i[\psi_0(T_2) - \psi_2(T_2)]} & 0 & 0 \\ 0 & e^{-i[\psi_0(T_2) - \psi_2(T_2)]} & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (5-30)$$

**$T > T_3$**

$$A(0)(T-T_3) = A_s(0) + e^{-B_0(T-T_3)} [A(0)(T_3) - A_s(0)] \quad (5-31)$$

$$A(0)(T_3) = C_3 A(2)(T_3 - T_2) \quad (5-32)$$
\[ C_3 = \begin{pmatrix} \exp(i[\psi_2(T_3) - \psi_0(T_3)]) & 0 & 0 \\ 0 & \exp(-i[\psi_2(T_3) - \psi_0(T_3)]) & 0 \\ 0 & 0 & 1 \end{pmatrix} \] (5-33)

Combining the expressions for \( A(T) \) and solving for \( \sigma_{ab}(T) \) yields

\[
\sigma_{ab}(T) = A_1(T-T_3)e^{-i\psi_0(T)}
\]

\[
= \begin{vmatrix} e^{-B_0(T-T_3)} \\ e^{-B_2(T_3-T_2)} \end{vmatrix}_{11} C_3 \begin{vmatrix} e^{-B_2(T_3-T_2)} \\ e^{-B_1(T_1-T_0)} \end{vmatrix}_{1r}
\]

\[
\times \begin{vmatrix} e^{-B_0(T_2-T_1)} \\ e^{-A_1(T_0)-A_s(T_1)} \end{vmatrix}_{rr} \begin{vmatrix} C_2 \\ C_1 \end{vmatrix}_{rr}
\]

\[
\times \begin{vmatrix} A(1)(T_0) \end{vmatrix}_{r'} e^{i\psi_0(T)}.
\]

(5-34)

In the above, the bracket subscripts refer to the matrix element.

Also, the terms \( A(0)(T_1) \), \( A(2)(T_2) \), and \( A(0)(T_3) \) were considered small and ignored.

From Equation (1-36), the phase of the echo is related to those of the initial pulses by \( \phi_E = 2\phi_2 - \phi_1 + \text{constant} \). Using this in Equation (5-34), shows echoes are formed when \( r = 2 \) and \( r' = 1, 2, 3 \). Other values of \( r \) yield non-echo terms which are not considered here.

A method for calculating the matrix elements of \( \exp(-BT) \) is
shown in Appendix A. Also shown is the case when the lifetimes, $T_1$ and $T_2$, are equal or equivalently when $Y = Z$. Finally, these results are used to show that the time shift predicted by the phenomenological equations for the non-damped case is the same as shown in Equation (1-72).

**Density Matrix Calculations**

An IBM 360 computer using FORTRAN IV language was used to calculate $\sigma_{ab}(T)$ which was accomplished in two steps. First, $\sigma_{ab}(T)$ was written as

$$\sigma_{ab}(X,Y,Z,T_p,T_G,T) = \alpha(X,Y,Z,T_p,T_G)e^{-(iX+Z)(T-T_3)}$$

where $\alpha$ is the computer generated product of the matrix elements in Equation (5-34) and

$$e^{-(iX+Z)(T-T_3)}$$

was computed. $T_p$ is the pulse length, $T_G$ is the time gap between the pulses, $X = \Delta/KE$, $Y = \gamma_1/KE$, $Z = \gamma_2/KE$, and $T = KEt$. The $\exp \{-i\psi_0(T)\}$ has been suppressed in Equation (5-35).

In this form with $\alpha$ calculated by the computer, the second step of multiplying $\alpha$ by $\exp \{-i\psi_0(T)\}$ can be easily performed. Appendix B contains the computer program used to calculate $\alpha$ and $\sigma_{ab}(T-T_3)$. Appendix C lists $\alpha(X,Y,Z,T_p,T_G)$ for a number of cases.

In Appendix D, it is shown that the sum of the amplitudes of the
electric fields of the radiating dipoles in a small sample at a point far from the sample is proportional to $W$ where

$$W = i \sigma_{ab}(T) + \text{c.c.}$$

Since this is the term of interest, the following discussion will be in terms of $W$ as it is affected by the various parameters.

**Results of Calculations**

The first case to be considered is when the lifetimes are infinite or when $Y = Z = 0$. Here the steady state terms are $A_{s1}^{(1)} = A_{s2}^{(1)} = A_{s3}^{(1)} = 0$. For these and subsequent calculations $T_p = 0.40$ and $T_G = 0.64$. In Figure 18, $W$ is plotted against $T - T_3$ for various values of $X = \Delta/KE$. $\Delta = \omega_{ab} - \omega + \phi$ and is a measure of the degree of off-resonance of $\omega$. The echo will form for positive $W$ and the intensity will be proportional to $W^2$.

Several important features are evident in Figure 19. The time of maximum echo intensity is the same for all $X$'s although the curves become sharper as $X$ increases. The degree of off-resonance as measured by $X$ indicates that $W$ for values of $X$ greater than 3.0 will have little effect on the formation of the echo. This trend was also noted in Equation (1-66).

The absence of damping terms is particularly evident in the curves for the higher $X$'s. For each $X$ the succeeding peaks show no loss in
Figure 19. Plot of $W = \sigma_{ab} (t) + C.C.$ Versus $(T-T_3)$ for $X = \Delta/KE$
Ranging from 0.4 to 2.8 and No Damping, $Y = \gamma_1/KE = Z = \gamma_2/KE$. 
Figure 20. Plot of $\bar{W}$, the Average for X Ranging from 0.0 to 3.0 in Steps of 0.2 Versus $(T-T_3)$. There are No Damping Effects as $Y = Z = 0$. 
amplitude as T increases. The time of echo formation is $T-T_3 = 0.89$. This is in agreement with $T-T_3 = T_G + (2/\pi)T_p = 0.64 + (2/\pi)(0.40) = 0.89$ from Equations (1-73) and (1-74).

The average of these curves with X ranging from 0.0 to 3.0 in steps of 0.2 is plotted in Figure 19. The result is a symmetrically shaped echo with a peak at $T-T_3 = 0.89$.

The next case to be considered is when the lifetimes correspond to those of the Na D lines and when the initial pulse width is 5 nanoseconds, then $Y = 2Z = 0.2$. Figure 21 is a plot of W versus $T-T_3$ for this case. Two new effects can be easily seen. There is a negative time shift in the echo formation which decreases as X increases. A second effect is the exponential damping which is evidenced by the lower peak values of W and by the decrease in the maximum values of W as T increases. This latter effect was expected but the first represents a new feature.

The average of W versus $T-T_3$ is plotted in Figure 22. The resultant echo forms earlier and its intensity is much lower than in the previous case.

The large time shift for small X seen in Figure 21 can be shown to be primarily a function of Y and not of Z by considering two further cases. First, let $Y = \gamma_1/KE = 0$ and $Z = \gamma_2/KE = 0.1$ as shown in Figures 23 and 24. From Figure 23 the shift at low X is much smaller than for the previous case. The resultant average of W over X
Figure 21. Plot of $W = f_{ab}^c(T) + C.C.$ Versus $(T-T_3)$ for $X = \Delta/KE$
Ranging from 0.4 to 2.8, where $Y = \gamma_1/KE = 0.2$ and $Z = \gamma_2/KE = 0.1$. These Lifetimes Correspond to the Na D Lines.
Figure 22. Plot of $\bar{W}$, the Average for $X$ Ranging from 0.0 to 3.0 in Steps of 0.2 Versus $(T-T_3)$. Also, $Y = \gamma_1 / KE = 0.2$ and $Z = \gamma_2 / KE = 0.1$. 
Figure 23. Plot of $W = \sigma_{ab}(T) + C.C.$ Versus $(T-T_3)$ for $X = \Delta/KE$ Ranging from 0.4 to 2.8, where $Y = \gamma_1/KE = 0$ and $Z = \gamma_2/KE = 0.1$. 
Figure 24. Plot of $\bar{W}$, the Average of $W$ for $X$ Ranging from 0.0 to 3.0 in Steps of 0.2 Versus $(T-T_3)$. Also, $Y = \gamma_1/KE = 0$ and $Z = \gamma_2/KE = 0.1$. 
plotted in Figure 24 shows an echo shape similar to that of the undamped case, $Y = Z = 0$.

With $Y = Z = 0.1$ the shift is large for small $X$ and the resultant echo is distorted as shown in Figures 25 and 26.

The cause for this shift is the steady state terms $A_s$ as given in Equation (5-17). As $Y$ approaches zero, the $A_s$'s also approach zero. This implies that from the expression for $\sigma_{ab}(T)$ given in Equation (5-34), the matrix element of interest is $\{e^{-B_1(T-T_0)}\}_{23}$ which is a complex number. As shown in Appendix A, this element gives rise to a time shift of $T = 1$ for small $X$. This same feature was also seen in Equations (1-73) and (1-74).

As $Y$ increases from zero, the $A_s$'s increase causing the complex matrix elements $\{e^{-B_1(T_1-T_0)}\}_{21}$ and $\{e^{-B_1(T_1-T_0)}\}_{22}$ to become important. These elements cause a further shift in the echo formation time which is shown by comparing Figures 24 and 26.

Thus, the steady state terms, $A_s$, determine the effect of the lifetimes on the production of the echo. For long lifetimes the $A_s$'s approach zero and the results are similar to those found in Chapter I. As the lifetimes become shorter, the echo intensity drops and there is a shift in the echo formation time which is a function of $\Delta$, the degree of off-resonance of the initial pulses. Further, this shift appears to be a stronger function of the lifetime $T_1$ than of $T_2$. 
Figure 25. Plot of $W = i\sigma_{ab}(T) + C.C.$ Versus $(T - T_3)$ for $X = \Delta/KE$ ranging from 0.4 to 2.8, where $Y = \gamma_1/KE = Z = \gamma_2/KE = 0.1.$
Figure 26. Plot of $\bar{W}$, the Average for $W$ for $X$ Ranging from 0.0 to 3.0 in Steps of 0.2
Versus $(T-T_3)$. Also, $Y = \gamma_1/KE = Z = \gamma_2/KE = 0.1$. 
APPENDIX A

Exponential Matrix Calculations

In the development of the phenomenological approach to the production of photon echoes it was necessary to solve the differential equation

$$\frac{dA}{dT} = -BA + C$$

where

$$A = \begin{pmatrix} A_1 \\ A_2 \\ A_3 \end{pmatrix}$$

$$B = \begin{pmatrix} iX+Z & 0 & i/2 \\ 0 & -iX+Z & -i/2 \\ i & -i & Y \end{pmatrix}$$

and

$$C = YA_3(T_0) \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}.$$  

The parameters $X$, $Y$, and $Z$ were defined as

$$X \equiv \Delta/KE,$$

$$Y \equiv \gamma_1/KE,$$

and

$$Z \equiv \gamma_2/KE.$$  

$K$, $E$, $\Delta$, $\gamma_1$, $\gamma_2$, and $T$ are as defined in Chapter V.
The solution was

\[ A(T-T_0) = A_s + e^{-B(T-T_0)} \left[ A(T_0) - A_s \right] \]

where

\[ A_s^1 = A_s^2 = \frac{Y(X+1Z)}{Z+Y(X^2+Z^2)} \]

and

\[ A_s^3 = \frac{-Y(X^2+Z^2)}{Z+Y(X^2+Z^2)} \]

The matrix elements of \( \exp(-BT) \) can be calculated from

\[ e^{-BT} = \sum_r e^{-\lambda_r T} P_r \]

where \( \lambda_r \) is the \( r^{th} \) eigenvalue of \( B \). \( P_r \) is a projection operator and may be calculated from

\[ P_r = \prod_{r \neq s} \frac{B-\lambda_s}{\lambda_r-\lambda_s} \] .

In general there is no simplification to ease the calculations. The complex eigenvalues of \( B \) are calculated and used to determine the projection operators and finally the matrix elements of \( \exp(-BT) \). If the lifetimes are equal, \( Y=Z \), then the eigenvalues of \( B \) take on a simple form which allows \( P_r \) to be easily calculated.

For \( Z=Y \), \( B \) becomes
\[
B = \begin{pmatrix}
iX+Z & 0 & i/2 \\
0 & -iX+Z & -i/2 \\
i & -i & Z
\end{pmatrix}
\]

with the eigenvalues

\[\lambda_0 = Z,\]

and

\[\lambda_{\pm} = Z \pm i\sqrt{1+X^2}.\]

For $X=0$, the resulting projection operators are

\[
P_0 = \begin{pmatrix}
1/2 & 1/2 & 0 \\
1/2 & 1/2 & 0 \\
0 & 0 & 0
\end{pmatrix},
\]

and

\[
P_{\pm} = \begin{pmatrix}
1/4 & -1/4 & \pm 1/4 \\
-1/4 & 1/4 & \mp 1/4 \\
\pm 1/2 & \mp 1/2 & 1/2
\end{pmatrix},
\]

If $X \neq 0$, then

\[
P_0 = \frac{1}{1+X^2} \begin{pmatrix}
1/2 & 1/2 & -X/2 \\
1/2 & 1/2 & -X/2 \\
-X & -X & X^2
\end{pmatrix},
\]

and
\[ P_\pm = \frac{1}{2(1+X^2)} \begin{pmatrix} 1/2 + X^2 & \sqrt{1+X^2} & -1/2 & 1/2(X \pm \sqrt{1+X^2}) \\ -1/2 & 1/2 + X^2 & \sqrt{1+X^2} & 1/2(X \mp \sqrt{1+X^2}) \\ X \pm \sqrt{1+X^2} & X \mp \sqrt{1+X^2} & 1 & \end{pmatrix} \]

Using these expressions for \( P_r \) and \( \lambda_r \) the matrix elements of \( \exp(-BT) \) needed to generate photon echoes are

\[ \begin{align*}
\{ e^{\frac{-B_0(T_T - T_3)}{2}} \} &= -e^{(iX+Z)(T_T - T_3)} , \\
\{ e^{\frac{-B_2(T_3 - T_2)}{2}} \} &= \frac{1}{2(1+X^2)} e^{-Z(T_3 - T_2)} [1 - \cos(\sqrt{1+X^2}(T_3 - T_2))] , \\
\{ e^{\frac{-B_1(T_2 - T_1)}{2}} \} &= -e^{-(iX+Z)(T_2 - T_1)} , \\
\{ e^{\frac{-B_1(T_1 - T_0)}{2}} \} &= \frac{1}{2(1+X^2)} e^{-Z(T_1 - T_0)} [1 - \cos(\sqrt{1+X^2}(T_1 - T_0))] , \\
\{ e^{\frac{-B_1(T_1 - T_0)}{2}} \} &= \frac{1}{2(1+X^2)} e^{-Z(T_1 - T_0)} [1 + (1+2X^2) \cos(\sqrt{1+X^2}(T_1 - T_0))] + iX\sqrt{1+X^2} \sin(\sqrt{1+X^2}(T_1 - T_0)) , \\
\{ e^{\frac{-B_1(T_1 - T_0)}{2}} \} &= \frac{1}{2(1+X^2)} e^{-Z(T_1 - T_0)} [-X + X \cos(\sqrt{1+X^2}(T_1 - T_0))] + i\sqrt{1+X^2} \sin(\sqrt{1+X^2}(T_1 - T_0)) .
\end{align*} \]
In the case of infinite lifetimes, \( Y = Z = 0 \), the \( A_s \)'s are zero and according to Equation (5-34), \( \sigma_{ab}(T) \) becomes

\[
\sigma_{ab}(T) = e^{-i\alpha(T-T_0)} e^{i\alpha(T_2-T_1)} \frac{1}{(2(1+\alpha^2))^2} \times [1-\cos(\sqrt{1+\alpha^2}(T_3-T_2))] \\
\times [-\alpha + \alpha \cos(\sqrt{1+\alpha^2}(T_1-T_0))] \\
+ i\sqrt{1+\alpha^2} \sin(\sqrt{1+\alpha^2}(T_1-T_0))].
\]

The last square bracket can be written as a real number times \( \exp(i\alpha T_s') \) where

\[
\alpha T_s' = \arctan \frac{\sqrt{1+\alpha^2} \sin(\sqrt{1+\alpha^2}(T_1-T_0))}{\sqrt{1+\alpha^2} \sin(\sqrt{1+\alpha^2}(T_1-T_0))}. 
\]

From Equations (1-56) and (5-17) it follows that \( T_1-T_0 \), the pulse width of the first pulse, is just \( \pi/2 \). Thus by use of the power series expansion

\[
\arctan u = \frac{\pi}{2} - \frac{1}{u} + \frac{1}{3u^3} - \frac{1}{5u^5}
\]
valid for \( u^2 > 1 \), \( \alpha T_s' \) to first order is \( \frac{\pi}{2} + \alpha \). With this, \( \exp(i\alpha T_s') \) becomes \( i \exp(\alpha T_s) \) and \( T_s = 1 \). Using the definition of \( T \) and Equation (1-56), the equation \( T_s = 1 \) becomes \( ts = \frac{2\pi}{\pi} \). This last expression is Equation (1-72), the time shift predicted in Chapter I.
APPENDIX B

The Computer Program

The computer program which was used to calculate $\alpha(X,Y,Z,T_p,T_G)$ in Equation (5-35) and $\sigma_{ab}(T)$ in Equation (5-36) is presented in this appendix. The program language is FORTRAN IV Extended and the H compiler was used. The general method of calculation is as outlined in Appendix A. Comment cards within the text of the program relate the program variables with those given in Appendix A.
COMPUTER PROGRAM FOR COMPUTING THE DENSITY MATRIX SIGMA

/*I09PARM LINES=5000
//PK0CLI9 DD OSN=HEP*PROCLIB,DISP=SHR
// EXEC FTXCG,LIB=*SYS1.IMSL,DUE* DOUBLE*
//FURT,SYSTN DD *
COMPLEX*16 UE,Du,RT,EB1(3,3),EBP0(3,3)
COMPLEX*16 b0(3,3),DLT,ELFD,T1INV,T2INV,CAP,EIG(3),
COMPLEX*16 AB(3,3),K(6),PR01B(3,3),EB01(3,3)
COMPLEX*16 0(3,3),A(3,3),n(3,3),FUD,PR01A(3,3)
COMPLEX*16 EB02(3,3),EB03(3,3),TL(3,3),TIME,TIMG
COMPLEX*16 PRO3A(3,3),PRO3B(3,3),PRO3(3,3),EB03(3,3)
COMPLEX*16 PRO1(3,3),PRO2A(3,3),PRO2B(3,3),PRO2(3,3)
COMPLEX*16 EBIPRD(3,3),EB(3,3)
COMPLEX*16 AO(3,3),AS(3),EF(3),EHI,EBF
COMPLEX*16 TM,SIGMA(96),SUM(96),SUM0(98)
COMPLEX*16 XI,ETA,ZETA

S#1(1)=0.0,0.0)
DE=(0.0,0.0)

100 READ(5,10,END=999)T1INV,T2INV,CAP
READ(5,11)DLT,ELFD,TIME

ELFD IS THE ELECTRIC FIELD AMPLITUDE
C DT IS THE DEGREE OFF-RESONANCE DELTA
C CAP IS THE CONSTANT K, TIME IS THE PULSE LENGTH
DE=DE+(1.0,0.0)
ELFD=ELFD/(4.0,0.0)
T1INV=(1.0-50.0,0.0)
WRITE(6,25) T1INV,T2INV
WRITE(6,27) CAP,TIME
WRITE(6,17) DLTD,ELFD
XI=DLT/(CAP*ELFD)
ETA=(0.0,1.0)*T2INV/(CAP*ELFD)
ZETA=(0.0,1.0)*T1INV/(CAP*ELFD)

XI IS THE VARIABLE X, ETA IS Z, AND ZETA IS Y
WRITE(5,29)
WRITE(6,16) XI,ETA,ZETA
WRITE(6,17)

B0(1,1)=DLT+T2INV
B0(2,1)=(0.0,0.0)
B0(3,1)=CAP*ELFD
B0(1,2)=(0.0,0.0)
B0(2,2)=-(DLT+T2INV
B0(3,2)=-B0(3,1)
B0(1,3)=0.05*CAP*ELFD
B0(2,3)=-B0(1,3)
B0(3,3)=T1INV

B3 IS THE MATRIX B
CALL EIGCC(A0,3,3,O,EIG,A,B,3,WK,IER)

C EIG ARE THE EIGENVALUES OF B

C PRO1 IS THE PROJECTION OPERATOR CORRESPONDING TO EIG(1)

C

DO 4 J = 1,3
PRO1(I,J) = (0.0,0.0)
DO 5 K = 1,3
PRO1(I,J) = PRO1(I,J) * PRO1A(I,K) * PRO1B(K,J)

3 CONTINUE

4 CONTINUE

5 CONTINUE

DO 4 J = 1,3
PRO2A(I,J) = (BC(I+1,1)-EIG(J))/(EIG(J)-EIG(K))
PRO2B(1,1) = (BG(1,1) - EIG(1)) / (EIG(2) - EIG(1))
PRO2B(2,1) = (BG(2,1)) / (EIG(2) - EIG(1))
PRO2B(3,1) = (BG(3,1)) / (EIG(2) - EIG(1))
PRO2B(1,2) = (BG(1,2)) / (EIG(2) - EIG(1))
PRO2B(2,2) = (BG(2,2) - EIG(1)) / (EIG(2) - EIG(1))
PRO2B(3,2) = (BG(3,2)) / (EIG(2) - EIG(1))
PRO2B(1,3) = (BG(1,3)) / (EIG(2) - EIG(1))
PRO2B(2,3) = (BG(2,3)) / (EIG(2) - EIG(1))
PRO2B(3,3) = (BG(3,3) - EIG(1)) / (EIG(2) - EIG(1))

DO 33 I = 1, 3
DO 34 J = 1, 3
PRO2(I,J) = (B0(0,0,0,0)
DO 35 K = 1, 3
PRO2(I,K) = PRO2(I,K) * PRO2B(K,J)
C PRO2 IS THE PROJECTION OPERATOR CORRESPONDING TO EIG(2)
35 CONTINUE
34 CONTINUE
33 CONTINUE
C
PRO3A(1,1) = (BG(1,1) - EIG(1)) / (EIG(3) - EIG(1))
PRO3A(2,1) = (BG(2,1)) / (EIG(3) - EIG(1))
PRO3A(3,1) = (BG(3,1)) / (EIG(3) - EIG(1))
PRO3A(1,2) = (BG(1,2)) / (EIG(3) - EIG(1))
PRO3A(2,2) = (BG(2,2) - EIG(1)) / (EIG(3) - EIG(1))
PRO3A(3,2) = (BG(3,2)) / (EIG(3) - EIG(1))
PRO3A(1,3) = (BG(1,3)) / (EIG(3) - EIG(1))
PRO3A(2,3) = (BG(2,3)) / (EIG(3) - EIG(1))
PRO3A(3,3) = (BG(3,3) - EIG(1)) / (EIG(3) - EIG(1))
PRO3B(1,1) = (BG(1,1) - EIG(2)) / (EIG(3) - EIG(2))
PRO3B(2,1) = (BG(2,1)) / (EIG(3) - EIG(2))
PRO3B(3,1) = (BG(3,1)) / (EIG(3) - EIG(2))
PRO3B(1,2) = (BG(1,2)) / (EIG(3) - EIG(2))
PRO3B(2,2) = (BG(2,2) - EIG(2)) / (EIG(3) - EIG(2))
PRO3B(3,2) = (BG(3,2)) / (EIG(3) - EIG(2))
PRO3B(1,3) = (BG(1,3)) / (EIG(3) - EIG(2))
PRO3B(2,3) = (BG(2,3)) / (EIG(3) - EIG(2))
PRO3B(3,3) = (BG(3,3) - EIG(2)) / (EIG(3) - EIG(2))

DO 43 I = 1, 3
DO 44 J = 1, 3
PRO3(I,J) = (B0(0,0,0,0)
DO 45 K = 1, 3
PRO3(I,K) = PRO3(I,K) * PRO3B(K,J)
C PRO3 IS THE PROJECTION OPERATOR CORRESPONDING TO EIG(3)
45 CONTINUE
44 CONTINUE
43 CONTINUE
C
TL(1,1) = C*EXP(-EIG(1)*TIME)
TL(2,2) = TL(1,1)
TL(3,3) = TL(1,1)
CALL EICL(40,3,EICL,EICL,EICL,EICL)

CALL EICL(40,3,EICL,EICL,EICL,EICL)

CALL EICL(40,3,EICL,EICL,EICL,EICL)

CALL EICL(40,3,EICL,EICL,EICL,EICL)

CALL EICL(40,3,EICL,EICL,EICL,EICL)

CALL EICL(40,3,EICL,EICL,EICL,EICL)
DO 114 J=1,3
PRO1(I,J)=(0.0,0.0)
DO 115 K=1,3
PRO1(I,J)=PRO1(I,J)*PRO1A(I,K)*PRO1B(K,J)
115 CONTINUE
114 CONTINUE
113 CONTINUE
C
PRO2A(I,1)=(BO(I,1)-EIG(3))/(EIG(2)-EIG(3))
PRO2A(2,1)=(BO(2,1))/(EIG(2)-EIG(3))
PRO2A(3,1)=(BO(3,1))/(EIG(2)-EIG(3))
PRO2A(I,2)=(BO(I,2))/(EIG(2)-EIG(3))
PRO2A(2,2)=(BO(2,2)-EIG(3))/(EIG(2)-EIG(3))
PRO2A(3,2)=(BO(3,2))/(EIG(2)-EIG(3))
PRO2A(I,3)=(BO(I,3))/(EIG(2)-EIG(3))
PRO2A(2,3)=(BO(2,3))/(EIG(2)-EIG(3))
PRO2A(3,3)=(BO(3,3)-EIG(3))/(EIG(2)-EIG(3))
PRO2B(I,1)=(BO(I,1)-EIG(1))/(EIG(2)-EIG(1))
PRO2B(2,1)=(BO(2,1))/(EIG(2)-EIG(1))
PRO2B(3,1)=(BO(3,1))/(EIG(2)-EIG(1))
PRO2B(2,2)=(BO(2,2)-EIG(1))/(EIG(2)-EIG(1))
PRO2B(3,2)=(BO(3,2))/(EIG(2)-EIG(1))
PRO2B(I,3)=(BO(I,3))/(EIG(2)-EIG(1))
PRO2B(2,3)=(BO(2,3))/(EIG(2)-EIG(1))
PRO2B(3,3)=(BO(3,3)-EIG(1))/(EIG(2)-EIG(1))
DO 133 I=1,3
DO 134 J=1,3
PRO2(I,J)=(0.0,0.0)
DO 135 K=1,3
PRO2(I,J)=PRO2(I,J)*PRO2A(I,K)*PRO2B(K,J)
135 CONTINUE
134 CONTINUE
133 CONTINUE
C
PRO3A(I,1)=(BO(I,1)-EIG(1))/(EIG(3)-EIG(1))
PRO3A(2,1)=(BO(2,1))/(EIG(3)-EIG(1))
PRO3A(3,1)=(BO(3,1))/(EIG(3)-EIG(1))
PRO3A(I,2)=(BO(I,2))/(EIG(3)-EIG(1))
PRO3A(2,2)=(BO(2,2)-EIG(1))/(EIG(3)-EIG(1))
PRO3A(3,2)=(BO(3,2))/(EIG(3)-EIG(1))
PRO3A(I,3)=(BO(I,3))/(EIG(3)-EIG(1))
PRO3A(2,3)=(BO(2,3))/(EIG(3)-EIG(1))
PRO3A(3,3)=(BO(3,3)-EIG(1))/(EIG(3)-EIG(1))
PRO3B(I,1)=(BO(I,1)-EIG(2))/(EIG(3)-EIG(2))
PRO3B(2,1)=(BO(2,1))/(EIG(3)-EIG(2))
PRO3B(3,1)=(BO(3,1))/(EIG(3)-EIG(2))
PRO3B(I,2)=(BO(I,2))/(EIG(3)-EIG(2))
PRO3B(2,2)=(BO(2,2)-EIG(2))/(EIG(3)-EIG(2))
PRO3B(3,2)=(BO(3,2))/(EIG(3)-EIG(2))

\[ \text{PROC} 3(1,3) = \frac{(B(1,3))/(E(3)-E(2))}{(E(3)-E(2))} \]
\[ \text{PROC} 1(2,3) = \frac{(B(2,3))/(E(3)-E(2))}{(E(3)-E(2))} \]
\[ \text{PROC} 2(3,3) = \frac{(B(3,3))/(E(3)-E(2))}{(E(3)-E(2))} \]
\[ \text{DO } 143 \text{ I}=1,3 \]
\[ \text{DO } 144 \text{ J}=1,3 \]
\[ \text{PROC}(I,J) = \{0.0,0.0,0.0\} \]
\[ \text{DO } 145 \text{ K}=1,3 \]
\[ \text{PROC}(I,J) = \text{PROC}(I,K) \times \text{PROC}(K,J) \]
\[ \text{CONTINUE} \]
\[ \text{CONTINUE} \]
\[ \text{CONTINUE} \]

\[ \text{TL}(1,1) = \text{CDEXP}(\text{-E}(1) \times \text{TIME}) \]
\[ \text{TL}(2,2) = \text{TL}(1,1) \]
\[ \text{TL}(3,3) = \text{TL}(1,1) \]
\[ \text{TL}(1,2) = \{0.0,0.0,0.0\} \]
\[ \text{TL}(1,3) = \{0.0,0.0,0.0\} \]
\[ \text{TL}(2,1) = \{0.0,0.0,0.0\} \]
\[ \text{TL}(2,3) = \{0.0,0.0,0.0\} \]
\[ \text{TL}(3,1) = \{0.0,0.0,0.0\} \]
\[ \text{TL}(3,2) = \{0.0,0.0,0.0\} \]
\[ \text{DO } 155 \text{ I}=1,3 \]
\[ \text{DO } 156 \text{ J}=1,3 \]
\[ \text{EB01}(I,J) = \{0.0,0.0,0.0\} \]
\[ \text{DO } 157 \text{ K}=1,3 \]
\[ \text{EB01}(I,J) = \text{EB01}(I,K) \times \text{TL}(I,K) = \text{PROC}(K,J) \]
\[ \text{CONTINUE} \]
\[ \text{CONTINUE} \]
\[ \text{CONTINUE} \]

\[ \text{TL}(1,1) = \text{CDEXP}(\text{-E}(2) \times \text{TIME}) \]
\[ \text{TL}(2,2) = \text{TL}(1,1) \]
\[ \text{TL}(3,3) = \text{TL}(1,1) \]
\[ \text{DO } 165 \text{ I}=1,3 \]
\[ \text{DO } 166 \text{ J}=1,3 \]
\[ \text{EB02}(I,J) = \{0.0,0.0,0.0\} \]
\[ \text{DO } 167 \text{ K}=1,3 \]
\[ \text{EB02}(I,J) = \text{EB02}(I,K) \times \text{TL}(I,K) = \text{PROC}(K,J) \]
\[ \text{CONTINUE} \]
\[ \text{CONTINUE} \]
\[ \text{CONTINUE} \]

\[ \text{TL}(1,1) = \text{CDEXP}(\text{-E}(3) \times \text{TIME}) \]
\[ \text{TL}(2,2) = \text{TL}(1,1) \]
\[ \text{TL}(3,3) = \text{TL}(1,1) \]
\[ \text{DO } 175 \text{ I}=1,3 \]
\[ \text{DO } 176 \text{ J}=1,3 \]
\[ \text{EB03}(I,J) = \{0.0,0.0,0.0\} \]
\[ \text{DO } 177 \text{ K}=1,3 \]
\[ \text{EB03}(I,J) = \text{EB03}(I,K) \times \text{TL}(I,K) = \text{PROC}(K,J) \]
\[ \text{CONTINUE} \]
\[ \text{CONTINUE} \]
\[ \text{CONTINUE} \]
CONTINUE

\[ E_B(1,1) = E_B(1,1) \times E_B(2,1) \times E_B(3,1) \]
\[ E_B(2,1) = E_B(2,1) \times E_B(2,2) \times E_B(2,3) \]
\[ E_B(3,1) = E_B(3,1) \times E_B(3,2) \times E_B(3,3) \]
\[ E_B(1,2) = E_B(1,1) \times E_B(2,1) \times E_B(3,1) \]
\[ E_B(2,2) = E_B(2,1) \times E_B(2,2) \times E_B(2,3) \]
\[ E_B(3,2) = E_B(3,1) \times E_B(3,2) \times E_B(3,3) \]
\[ E_B(1,3) = E_B(1,1) \times E_B(2,1) \times E_B(3,1) \]
\[ E_B(2,3) = E_B(2,1) \times E_B(2,2) \times E_B(2,3) \]
\[ E_B(3,3) = E_B(3,1) \times E_B(3,2) \times E_B(3,3) \]

C \ E_B \ is \ the \ matrix \ for \ pulse \ 2.

\[ T_I M = (0.00-09,0.00-00) \]

C \ T_I M \ is \ the \ time \ between \ the \ pulses.

\[ E_B(1,1) = C D E X P \left( -(T_1-I N V) \times T_I M \right) \]
\[ E_B(2,2) = C D E X P \left( -(T_1-I N V) \times T_I M \right) \]
\[ E_B(3,3) = C D E X P \left( -(T_1-I N V) \times T_I M \right) \]
\[ E_B(1,2) = (0.0,0.0,0.0) \]
\[ E_B(2,1) = (0.0,0.0,0.0) \]
\[ E_B(2,3) = (0.0,0.0,0.0) \]
\[ E_B(3,1) = (0.0,0.0,0.0) \]
\[ E_B(3,2) = (0.0,0.0,0.0) \]

C \ E_B \ is \ the \ matrix \ for \ the \ gap \ between \ pulse \ 1 \ & \ 2.

\[ E_B = E_B \times E_B \times E_B \times E_B \]
\[ E_B = E_B \times E_B \times E_B \times E_B \]
\[ E_B = E_B \times E_B \times E_B \times E_B \]
\[ E_B = E_B \times E_B \times E_B \times E_B \]
\[ E_B = E_B \times E_B \times E_B \times E_B \]

C \ E_B \ is \ the \ constant \ alpha.

\[ T_M = (0.0,0.0) \]
\[ D O \ 190 \ J = 1,61 \]
\[ E_B = E_B \times E_B \times E_B \times E_B \]
\[ E_B = E_B \times E_B \times E_B \times E_B \]
\[ E_B = E_B \times E_B \times E_B \times E_B \]

C \ E_B \ is \ the \ final \ density \ matrix.

C \ T_M \ is \ the \ time \ (T-T_3).

\[ \text{SUM}(J) = \text{SUM}(J) \times E_B \]
\[ \text{SUM}(J) = \text{SUM}(J) \times E_B \]
\[ \text{SUM}(J) = \text{SUM}(J) \times E_B \]
\[ \text{SUM}(J) = \text{SUM}(J) \times E_B \]

C \ \text{SUM} \ \text{D} \ is \ the \ average \ of \ \text{E_B} \ over \ \text{X} \ from \ 0 \ to \ \text{X}.

\[ \text{WRITE} \left( 6,30 \right) \ \text{SUM} \left( J \right) \]
\[ \text{TM} = \text{TM} \times (0.50-09,0.00-00) \]

190 \ \text{CONTINUE}

\[ \text{GO TO 100} \]

999 \ \text{STOP}
C
10 FORMAT(6D13.6)
11 FORMAT(6D13.6)
12 FORMAT(' ODELTA=' ,2D20.6,5X, ' ELECTRIC FIELD=' ,2D20.6)
13 FORMAT(' EBI=' ,2D20.6)
14 FORMAT(' EBIPRD=' ,2D20.6)
15 FORMAT(' E3PRD=*, 2D20.6)
16 FORMAT(' OXI=' ,2D15.6,5X, ' ETA=' ,2D15.6,5X, ' ZETA=*, 12D15.6)
17 FORMAT(' O*)
18 FORMAT(' AS(1)=*,2D15.6,5X, AS(2)=*,2D15.6,5X, AS(3)=*,2D15.6)
19 FORMAT(' A VECTOR =*,2D15.6,5X,2D15.6,5X,2D15.6)
20 FORMAT(' EB=*,2D20.6)
21 FORMAT(' OSIGMA=*,2D20.6,5X, TM=*,2D20.6)
22 FORMAT(' SIGMA=*,2D20.6,5X, TIME=*,2D20.6)
23 FORMAT(' IT1INV=*,2D20.6,5X, T2INV=*,2D20.6)
24 FORMAT(' EBD=*,2D20.6)
25 FORMAT(' OCAP=*,2D20.6,5X, TIME=*,2D20.6)
26 FORMAT(' EBI=*,2D20.6)
27 FORMAT(' OXI=DELTA/CAP*ELF0, ETA=1/CAP*ELF0*T2, ZETA=1/CAP*ELF0*T1*)
28 FORMAT(' SUM=*,2D20.6)
END

/*
//GO.SYSLIB DD OSN=SYS1.IMSL.DOUBLE,DISP=SHR
// DD OSN=SYS1.IMSL.SINGLE,DISP=SHR
// DD OSN=SYS1.FORTLIB,DISP=SHR
// DD OSN=SYS2.FORTSSP,DISP=SHR
//GO.SYSIN DD *
C DATA CARDS ARE INSERTED HERE. TWO EACH ARE NEEDED FOR C THE CALCULATION OF EACH SIGMA,
/*
//
APPENDIX C

Computed Values of $a(X, Y, Z, T_1, T_2)$

This appendix contains a list of $a(X, Y, Z, T_p, T_G)$ for different values of $Y$ and $Z$. Recall from Chapter V that $X = \Delta/KE$, $\Delta = \omega_{ab} - \omega + \phi$, $Y = 1/(KET_1)$, $Z = 1/(KET_2)$, $T_1$ and $T_2$ are the lifetimes, $T_p$ is the initial pulse width, and $T_G$ is the time gap between the pulses. In all cases $X$ varies from 0.4 to 2.8 in steps of 0.4. Also $T_p = 0.40$ and $T_G = 0.64$. 
Table 5. Computed Values of $a(X,Y,Z,T_p,T_G)$ where $X = \Delta/KE$, $Y = \gamma_1^2/KE$, $Z = \gamma_2/KE$,
$T_p$ = the Pulse Length of the Initial Pulses, and $T_G$ = Time Between the Pulses.

<table>
<thead>
<tr>
<th>$X$</th>
<th>$a(X,0.0,0.0,0.40,0.64)$</th>
<th>$a(X,0.025,0.025,0.40,0.64)$</th>
<th>$a(X,0.1,0.1,0.40,0.64)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.4</td>
<td>0.47323</td>
<td>-10.9804856</td>
<td>0.228892</td>
</tr>
<tr>
<td>0.8</td>
<td>0.145560</td>
<td>+10.395190</td>
<td>0.422684</td>
</tr>
<tr>
<td>1.2</td>
<td>-0.279307</td>
<td>+10.175878</td>
<td>-0.0514369</td>
</tr>
<tr>
<td>1.6</td>
<td>-0.159770</td>
<td>-10.156454</td>
<td>-0.111919</td>
</tr>
<tr>
<td>2.0</td>
<td>0.0621958</td>
<td>-10.108408</td>
<td>0.0614502</td>
</tr>
<tr>
<td>2.4</td>
<td>0.0514987</td>
<td>+10.0133171</td>
<td>0.0320415</td>
</tr>
<tr>
<td>2.8</td>
<td>0.000322627</td>
<td>+10.0143756</td>
<td>-0.0114124</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$X$</th>
<th>$a(X,0.25,0.25,0.40,0.64)$</th>
<th>$a(X,0.2,0.1,0.40,0.64)$</th>
<th>$a(X,0.0,0.0,0.1,0.40,0.64)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.4</td>
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APPENDIX D

The Electric Field Intensity
as a Function of $\sigma_{ab}(t)$, the Density Matrix

The induced electric dipole for the two level atom discussed in Chapters I and V can be calculated from

\[
\hat{d}_\alpha = \text{Tr} \hat{P}_\alpha \sigma(t) = \{\sigma_{ab}(t)(a|\hat{P}|b) + \text{c.c.}\}_\alpha = \hat{\hbar} \hat{K}\{\sigma_{ab}(t) + \text{c.c.}\}_\alpha
\]

where $\alpha$ identifies the $\alpha$th atom or molecule in the sample. The resulting electric field from this oscillating dipole is

\[
E_\alpha(\hat{r},t) = \frac{-\pi r_x \hat{p}_x + \pi r_y \hat{p}_y}{\varepsilon_0 \lambda^2} e^{-i\omega(t-t'/c)} + \text{c.c.} (1-19)
\]

In Reference 12 it was shown that when the sum over $\alpha$ is taken the resultant magnitude of the total electric field was proportional to $i\hat{p}_\alpha + \text{c.c.}$ where $p_\alpha$ is the coefficient of $\exp(-i\omega t)$ in Equation (1-18). $p_\alpha$ was also assumed to be constant in magnitude and direction. Thus, with $\hat{u}$ and $K$ real, the magnitude of the electric field is proportional to $W$ where

\[
W \equiv i\sigma_{ab}(t) + \text{c.c.} \quad (D-1)
\]
REFERENCES


27. Lecture Notes, Professor C.V. Heer, The Ohio State University, Columbus, OH 1977.