TWO PHOTON RESONANT PICOSECOND PULSE PROPAGATION IN LITHIUM VAPOR

DISSERTATION

Presented to the Graduate Council of the North Texas State University in Partial Fulfillment of the Requirements

For the Degree of

DOCTOR OF PHILOSOPHY

By

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August, 1987
Coherent effects of two photon resonant (TPR) picosecond pulses through lithium vapor have been studied in this work. Tunable picosecond pulses from a synchronously pumped dye laser were amplified at 10 Hz to mJ levels. The amplified pulses were sent through a pulse sequencer. The continuously phased and time delayed pair of pulses interacted with a lithium vapor column in a concentric heat pipe. The incident and transmitted signals as well as the third harmonic energy were recorded as a function of phase and delay between the pulses. These measurements were repeated for several tunings to study the effect of ac Stark shift (induced by the picosecond pulse) on the resonance.

The anomalous transmission of "90° phase shifted" pulse pair through a TPR medium (2s → 4S transition in lithium) has been experimentally demonstrated. It was seen that the dynamic Stark shift (being proportional to the intensity) played a crucial role in the tuning of a short pulse in two photon resonant interaction. Over a scan of about typically 0.2 nm in the average wavelength of the pulse, the resonance (Stark tuned) could be swept from the peak
field (maximum Stark shift) to zero field (zero Stark shift). For a detuning, the resonance condition was satisfied over only a little portion (temporal) of the pulse, where the intensity of the field generates just enough Stark shift to compensate the detuning from zero field resonance.

The work of this dissertation has been to prove that the coherence of multiphoton excitation can be studied by an appropriately phased and time delayed sequence of pulses. An application of this fundamental study of coherence has been made for the enhancement of third harmonic generation. The coherent recovery of the energy lost to the two photon absorption process enabled a larger propagation distance for the fundamental than in an interaction which is incoherent or coherent, but not using a 90° phase shifted pulse pair. Phase matching over this longer propagation distance gave an enhancement of third harmonic generation.
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CHAPTER I

INTRODUCTION

The purpose of this work is to investigate nonlinear resonant interaction of radiation with matter, in a time scale such that a phase relationship is maintained between the resonance and the radiation. This situation is referred to as "coherent resonant interaction" and it requires that no collision occurs during the optical excitation, since even elastic collision will produce random phase shifts in the excited atoms. A broad frequency distribution of resonances, such as caused by a "Maxwellian velocity distribution," will also contribute to a time decay of the mutual coherence between excited atoms. By restricting the interaction (duration) time to a few tens of picoseconds and using low pressure (several torr) vapors as media, we ensure that both collisions and Doppler broadening are negligible.

A basic coherent experiment consists of exciting a system with one pulse and "interrogating" it with a second one. The interaction is by definition "coherent" if the result of the measurement depends on the relative phase of the two pulses. In single photon coherent resonant interaction, the physical picture is fairly simple. A first pulse induces an electric dipole, of which the the field opposes or enhances that of
subsequent pulses. Such effects have been studied since 1966.

The case of two photon resonance is interesting for several reasons. Being a forbidden dipole transition, it cannot reradiate, unless in the presence of a strong field (stimulated two photon emission). A two photon absorption is known to enhance the third order susceptibility responsible for third harmonic generation (THG). Hence one might wonder if coherent interaction effects can be exploited to enhance THG. This work shows that this is indeed the case. As compared to higher order processes two photon resonances are a very special case, having energy as the main parameter (e.g., transition rate and Stark shift are both proportional to $|\mathcal{E}|^2$).

Coherent effects of the interaction of an ultrashort light pulse with a two photon resonant medium has been studied theoretically and experimentally. Various two photon analogues of one photon coherent effects have been reported in the literature for almost a decade. With the availability of amplified picosecond and femtosecond laser pulses nowadays, we can study the exciting field of Coherent Multiphoton Resonant Interactions. The coherence here refers to the coherence of the excitation and not of the light field.

The energy density is the main parameter characterizing the interaction. The lithium atom has an ideal level structure for this study. Transition $2S - 4S$ is pure two photon with $2S$
- 2P transition far off resonance as opposed to Na atom where the two photon resonance is at the same time a single photon resonance for picosecond (ps) pulse interaction. The two photon resonant (TPR) wavelength is conveniently available from tunable dye lasers. When a picosecond pulse having sufficient energy density (i.e., more than 50 mJ/cm$^2$) excites a TPR transition (here the 2S-4S transition in lithium vapor), many events occur simultaneously. The overall views are

1. Dominant two photon absorption,

2. Third Harmonic Generation occurs due to Raman effect, and

3. Loss due to ionisation, proportional to the intensity of the pulse and the total loss is proportional to the pulse energy density.

The nonlinear susceptibility (here $\chi^{(3)}$) is enhanced due to the presence of multiphoton resonance (here TPR). The conversion efficiency for harmonic generation, however, is limited by the presence of loss mechanisms such as resonant absorption, ionisation, non-uniform phase matching, etc., out of which the resonant absorption loss can be avoided by using the coherent interaction of properly phase shifted and time delayed pulse sequences propagating through several absorption lengths. This has been theoretically predicted by Diels & Georges and experimentally verified by Mukherjee et al.$^{19,13}$

The experimental and theoretical investigations reported
in this thesis are a part of an ongoing effort to understand Coherent Effects in Multiphoton Resonant Interaction in atoms and molecules by the use of properly phase shifted and time delayed sequence of pulses.

The demonstration of coherent effects of TPR interaction in lithium vapor by controlling phased and time delayed pair of pulses in the visible has initiated a theoretical and later possible experimental verification of coherent Four Photon Resonant (FPR) Interaction in Mercury Vapor. The detailed study of the FPR coherent pulse propagation in the PhD dissertation of Mukherjee gives us for the first time a close look at a real physical system under FPR conditions. Due to the numerous nonlinear couplings involved in FPR coherent pulse propagation, a wealth of new information can be obtained which were not present in the lower order processes.

In the experiments described in this thesis pulses of duration 6ps having an energy of 2 nJ per pulse from a synchronously pumped dye laser are amplified at 10 Hz to mJ levels. The amplified pulses are then sent through a Macht-Zender (MZ) Interferometer which produced the phase shifted and time delayed pair of pulses. The output of the Interferometer is passed through a spectroscopic heat pipe which produces a uniform (number density) vapor column of an adjustable length. As the pair of phase shifted and time delayed pulses (as monitored by the MZ interferometer)
propagate through the lithium vapor the dynamics of TPR absorption, THG and ionisation are modulated. Two factors govern this modulation, namely the resonance condition and the relative phase between the two pulses. It is important to remember that the resonance condition depends on the AC Stark shift of the exciting field (being proportional to the intensity) which changes over the envelope of the pulse. In principle, when the pulses are propagating on resonance and are phase shifted by 90°, we should observe an anomalous transmission similar to the Zero Area Pulse Propagation in one photon case. As opposed to the case of TPR, the lossless propagation condition $\Theta = 0 \text{(AREA)}$ can be obtained through late emission in the single photon case. After interacting with the lithium vapor in the heat pipe, the transmitted fundamental signal and the third harmonic energy was recorded as a function of the relative phase and delay between the pair of pulses.

The thesis is organised in the following manner. In Chapter II the density matrix formalism will be used to describe TPR coherent pulse propagation. After presenting an overview and illustrating the two photon vector model, pulse propagation effects and THG will be discussed. In Chapter III, a theoretical description of TPR pulse propagation and THG through lithium vapor (2S-4S) will be given. The relevant experimental conditions will be defined.
Chapter IV elaborates on the experimental methods and procedures. In Chapter V, the experimental and theoretical results will be presented. Finally in Chapter VI, the conclusion will be discussed.
Classically, the interaction of light with matter is viewed as an incident electromagnetic field perturbing the electronic charge cloud of the atom or molecule setting it into oscillation, which may not always be simple. The complicated oscillation of the induced electric dipole moment can be thought of a superposition of the harmonics of the fundamental field only if its oscillation is stationary (i.e., directly related to the field). If the transient effects can be neglected, the total polarisation $P$ can be written as,

$$ P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots $$

The transient response of the medium is imbedded in the time dependence of the induced polarisation $P$ which is again dependent on the state of polarisation the atom was in, before being excited. This polarisation $P$, which can be calculated semiclassically (i.e., treating the electromagnetic field
classically and the atom or molecule quantum mechanically), becomes the source term in Maxwell's equations, giving the generated field.

Coherent effects arise out of the correlation between the phases of incident electric field of the pulse and of the induced electric dipoles of the medium. In the coherent regime one can no longer average over the phases to calculate the net rate of transition. When optical pulses are very short and the total duration of the interaction is much smaller than any relaxation times and inverse detunings, then the interaction may be almost completely phase preserving and coherent. The first complete study of this interaction of an isolated two level system with radiation pertained to spins aligned in a magnetic field. The system of equations describing the precession of the resultant magnetisation vector in a radio frequency field is referred to as Bloch's equations. The same set of equations have been shown to apply to a single photon interaction. Similar sets of differential equations describing two and n-photon interactions are also referred to as Bloch's equations. These "optical Bloch's equations" were given a vector model interpretation by Feynman, Vernon and Hellwarth, geometrically as a pseudopolarisation vector rotating about the incident electric field.

The first work on coherent two photon effects was done by Hartmann where the Raman Echo for a three level was
developed. He factored out the fast time dependence in the Hamiltonian by making an Unitary Transformation to go to a "Doubly Rotating Frame." After this, using several time independent transformations the equations of motion for the density matrix were solved.

Later Belenov and Poluektov and Takatsuji used Heitler's method of renormalising the unperturbed Hamiltonian to absorb the contribution from the off resonant intermediate states. This was done by using a time dependent Unitary Transformation which broke the total Hamiltonian to an unperturbed, nonresonant, energy shifted part and a completely resonant (with 2-levels) part. Then through the Heisenberg's equation of motion for the Pauli Spin Operators, the optical Bloch equation were solved for the density matrix elements. Thereafter the polarisations were found and introduced into the Maxwell's equations to analyse the propagation effects. A comparison of the two methods of treating TPR pulse propagation is elegantly outlined in the paper by Grischkowsky et al.

Brewer and Hahn gave an exact solution for a three level system under transient or steady state conditions. In their vector model, Brewer and Hahn assumed two states to be resonantly coupled by a two photon transition. However this model did not impose any restriction on the intermediate level, which can be off resonance, or coupled by a single photon.
resonance to the other levels. In the approximate vector model of Takatsuji$^3$ the two photon resonant states may be exactly resonant or near resonant with the input field. There is no restriction to the number of intermediate states, but they have to be adiabatically coupled to the input field. When the intermediate levels are sufficiently off resonant, then their coupling with the input field is described by an adiabatic approximation. The interaction equations become very much simplified under this approximation. Grischkowsky et al.$^5$ discussed the adiabatic following model in two photon processes emphasizing mixing and pulse reshaping due to nonlinear pulse velocity and self phase modulation (SPM) in propagation effects.

Georges et al.$^7$ described the Third Harmonic Generation (THG) under two photon resonance (TPR) conditions in metal vapors. Diels and Georges gave a complete theoretical analysis of coherent two photon resonant third and fifth harmonic generation in metal vapors. They described coupled Maxwell-Bloch equations taking into account the optical Stark shifts, multiphoton ionisation, Doppler broadening, two photon absorption, the reaction of the third harmonic field on the fundamental and the nonlinear index of refraction. A numerical calculation done on pulse propagation under 2S-4S transition in lithium vapor for 90$^\circ$ phase shifted pulses (two photon analogue of zero area pulse propagation) predicted an energy conversion
efficiency of several percent in the third harmonic.

The theoretical calculations done in this thesis follow
the spirit of Diels et al. to explain the experimental results.
The equations describing TPR transition

In the case of resonant excitation with short pulses one can describe the atomic system using the density matrix formalism in order to explain the transient effects. The equation of motion of the density matrix $\rho$ is:

$$i\hbar \frac{\partial \rho}{\partial t} = [H, \rho] \quad (\text{II.1})$$

where $H$ is the total Hamiltonian of the atomic system in the presence of an electromagnetic field. The total Hamiltonian $H$ is approximated as usual by

$$H = H_0 - \vec{\mu} \cdot \vec{E} \quad (\text{II.2})$$

where $H_0$ is the unperturbed part and $\vec{\mu} \cdot \vec{E}$ is the major contribution to the perturbation originating from the electric dipole moment $\vec{\mu}$ induced by the external field $\vec{E}$ of the pulse. This can be reduced to a two level model under adiabatic approximation of the off resonant levels. The level structure corresponding to the unperturbed Hamiltonian $H$ is sketched in Fig 1. We will assume that no two levels are coupled resonantly by a single photon of the applied electromagnetic field and that the state $12\rangle$ ($4S$ in lithium) is the only one coupled to the ground state $11\rangle$ ($2S$ in lithium) by a "near resonant" two photon transition. In this context, the
Expression "near resonant" means that the detuning from the TPR cannot be considered to be larger compared to the transition rates to and from level \( I_2 \rangle \) and to the inverse of the duration. All states other than \( I_1 \rangle \) and \( I_2 \rangle \) are labelled \( I \ell \rangle \). Because these levels are off resonant, the amplitudes of the corresponding density matrix elements \( \rho_{i\ell} \) and \( \rho_{\ell i} \) can be neglected and the whole interaction can be described by a 4 X 4 matrix with only the terms \( \rho_{11} \), \( \rho_{22} \) and \( \rho_{21} \). The "adiabatic approximation" mentioned in the introduction will be applied to transitions involving levels \( \{ \ell \} \). The total interacting field \( \vec{E}(z,t) \) propagating along the Z-direction is,

\[
\vec{E}(z,t) = \vec{E}_1(z,t) e^{i(\omega t-kz)} + \vec{E}_3(z,t) e^{i3(\omega t-kz)} + c.c. \quad (II.3)
\]

where the electric field of the incident pulse \( \vec{E}_1(z,t) \) and of the third harmonic generated pulse \( \vec{E}_3(z,t) \) are the slowly varying complex amplitudes given by,

\[
\vec{E}_{1,3} = \left| E_{1,3}(z,t) \right| e^{i \phi_{1,3}(z,t)} \quad (II.4)
\]

where \( \phi_{1,3}(z,t) \) are the phases of \( \vec{E}_1 \) and \( \vec{E}_3 \) respectively.

We can define the atomic density matrix elements as,

\[
\rho_{12} = \text{atomic density matrix element coupling the resonant levels } 2S-4S.
\]

With \( N \) being the number density of atoms we have,

\[
N \rho_{11} = \text{population of atoms in } 2S \text{ state}
\]

\[
N \rho_{22} = \text{population of atoms in } 4S \text{ state}
\]
We define an amplitude $\sigma_{12}$ of the off diagonal matrix elements $\rho_{12}$ by:

$$\rho_{12}(t) = \sigma_{12}(t)e^{-i\omega t}$$  \hspace{1cm} (II.5)

where $\sigma_{12}(t)$ is slowly varying amplitude satisfying

$$\dot{\sigma}_{12}(t) \ll 2\omega \sigma_{12}(t)$$ \hspace{1cm} (II.6)

A derivation for the equations for $\sigma_{12}, \sigma_{22}$ and $(\sigma_{11} + \sigma_{22})$ is given in Appendix C, where the reaction of the third harmonic field is neglected. Using equations (II.3), (II.5) and (II.6) in equation (II.1) we have:

$$\frac{\partial \sigma_{12}}{\partial t} + i \left(2\omega - \omega_{21} - \delta \omega_{21}\right) \sigma_{12} + \left(\frac{1}{T_{2}} + \frac{\gamma_{1} + \gamma_{2}}{2}\right) \sigma_{12}$$

$$\hspace{3cm} = i (\sigma_{22} - \sigma_{11}) \left(\frac{\gamma_{2}}{\hbar^2} \mathcal{E}_{1}^2 + \frac{E_{21}^*}{\hbar^2} \mathcal{E}_{1} \mathcal{E}_{3}\right)$$  \hspace{1cm} (II.7)

and with the population terms $\rho_{11} = \sigma_{11}$ and $\rho_{22} = \sigma_{22}$ we have

$$\frac{\partial \sigma_{22}}{\partial t} = -\frac{\sigma_{22}}{T_{1}} - \gamma_{2} \sigma_{22} - 2 \text{Im} \left[\left(\frac{\gamma_{2}}{\hbar^2} \mathcal{E}_{1}^2 + \frac{E_{21}^*}{\hbar^2} \mathcal{E}_{1} \mathcal{E}_{3}\right) \sigma_{12}\right]$$  \hspace{1cm} (II.8)

and

$$\frac{\partial}{\partial t} (\sigma_{11} + \sigma_{22}) = -\gamma_{1} \sigma_{11} - \gamma_{2} \sigma_{22}$$  \hspace{1cm} (II.9)
where

\[ \omega_{21} = \text{Unperturbed two photon transition frequency} \]

\[ \delta \omega_{21} = \text{A.C. Stark Shift} \]

\[ \delta \omega_{21} = \frac{1}{\hbar} \left[ \alpha_j(\omega) - \alpha_j'(\omega) \right] |E_1|^2 \]

\[ + \frac{1}{\hbar} \left[ \alpha_j(3\omega) - \alpha_j'(3\omega) \right] |E_3|^2 \]

(II.10)

\[ \gamma_{1,2} = \text{Ionisation rates from 2S and 4S states} \]

\[ \gamma_1 = \gamma_{3\omega}^{3\omega} = \frac{2}{\hbar} \alpha_j''(3\omega) |E_3|^2 \]

(II.11)

\[ \gamma_2 = \gamma_{3\omega}^{3\omega} + \gamma_2^{3\omega} = \frac{2}{\hbar} \left[ \alpha_j''(\omega) |E_1|^2 + \alpha_j''(3\omega) |E_3|^2 \right] \]

(II.12)

where the complex polarisabilities of levels \( I_1 \) and \( I_2 \) are correspondingly with \( j = 1, 2 \)

\[ \alpha_j'(m\omega) = \alpha_j'(m\omega) - i \alpha_j''(m\omega) \]

\[ = \frac{1}{\hbar} \sum_{\ell} \left( \frac{|\mu_{\ell j}|^2}{\omega_{\ell j} - m\omega} + \frac{|\mu_{\ell j}|^2}{\omega_{\ell j} + m\omega} \right) \]

(II.13)

where \( \mu_{\ell j} \) and \( \omega_{\ell j} \) are the dipole matrix elements and the transition frequency of the transition \( I_1 \) \( \leftrightarrow \) \( I_j \). The summation is over all intermediate levels, bound or free.
The matrix elements responsible for two photon absorption is \( r_{12} \) given by

\[
\gamma_{12} = \sum_{\ell} \frac{\mu_{1\ell} \mu_{2\ell}}{\omega_{2\ell} + \omega}
\]  

(II.14)

and the matrix element responsible for Stimulated Raman Scattering (resulting the third harmonic generation) is given by

\[
\gamma_{21} = \sum_{\ell} \left( \frac{\mu_{2\ell} \mu_{1\ell}}{\omega_{1\ell} - 3\omega} - \frac{\mu_{2\ell} \mu_{1\ell}}{\omega_{2\ell} + 3\omega} \right)
\]  

(II.15)

where again the summation is over all intermediate levels bound and free. Note that to carry out the summation

\[
\sum_{\ell} \frac{\mu_{2\ell} \mu_{1\ell}}{\omega_{1\ell} - 3\omega}
\]

in the continuum one has to use Cauchy Integral for \( \omega_{1\ell} = 3\omega \) i.e. a resonance in the continuum and so becomes complex.
Two Photon Vector Model

The two photon vector model had been developed in many papers (Ref 1,3,4,5,10). Analogous to the one photon vector model of Feynman, Vernon and Hellwarth (Ref17) one can construct a two photon vector model, where a pseudopolarisation vector \( \vec{r} \) representing the coherent two photon interaction, rotates about the effective angular velocity vector \( \vec{\Omega} \). The latter can be decomposed into two orthogonal components namely the detuning (along the "population" axis) and the Rabi rate (defined in Eqn.II.18 below) for two photon transition.

Although in real situation the solution of the density matrix equations is very complex & difficult, we can consider some fictitious situation in which the motion of the vector can be easily visualised. We will write equations (II.7), (II.8), (II.9) under the following assumptions:

1. Detuning \( \Delta \omega = 0 \)
2. Interacting pulse width is much smaller than the relaxation times \( T_1 \) and \( T_2 \).
3. Neglect ionisation, which is true when peak field is not very large and \( \chi''_2(\omega) \) is not appreciable.
4. Neglect the reaction of the generated third harmonic field.
5. Doppler broadening is much smaller than the inverse pulse width.
6. Real electric field \( \vec{E} \), this is true when the self
phase modulation is small.

Under these assumptions the equations (II.7), (II.8) and (II.9) can be written as,

$$\dot{\sigma}_{12} = i (\sigma_{22} - \sigma_{11}) \frac{\chi_{12}}{\hbar^2} E^2$$  \hspace{1cm} (II.16)

and

$$\dot{\sigma}_{22} - \dot{\sigma}_{11} = -4 \text{Im} \left( \frac{\gamma_2}{\hbar^2} E^2 \sigma_{12}^{*2} \right)$$  \hspace{1cm} (II.17)

From equations (II.16), (II.17) and defining the Two Photon Rabi Rate of transition as

$$\Omega = \frac{2 \chi_{12}}{\hbar^2} E^2$$ \hspace{1cm} (II.18)

we have,

$$\dot{\sigma}_{12} = i \left( \sigma_{22} - \sigma_{11} \right) \frac{\Omega}{2}$$

$$\dot{\sigma}_{22} - \dot{\sigma}_{11} = -4 \text{Im} \left( \frac{\Omega}{2} \sigma_{12}^{*} \right)$$

Let us define the polarisation as $Q = i \sigma_{12}$ and the population difference as $W = (\sigma_{22} - \sigma_{11})/2$

Substituting these in the above equations we have,

$$\dot{Q} = - \Omega W$$

$$\dot{W} = \Omega Q$$

$$\ddot{Q} = - \Omega^2 Q$$

So

$$Q = \sin \Omega t$$ \hspace{1cm} (II.19)

and

$$W = \cos \Omega t$$ \hspace{1cm} (II.20)

We see that $Q$ and $W$ constitutes a circular motion with angular speed $\Omega$. So the pseudopolarisation vector $\vec{r}$ with the
components as \( r_1 = 0, r_2 = 0 \) and \( r_3 = \omega \) rotates with the angular velocity \( \vec{\Omega} = -\frac{\gamma_2}{\hbar^2} \vec{E}_1^2 \). This is shown in Fig. 2.

The real picture, however, is complex and making the only assumption of no ionisation we get the components of \( \vec{r} \) and \( \vec{\Omega} \) as,

\[
\begin{align*}
  r_1 &= \sigma_{12} + \sigma_{21} = \text{Re} \, \sigma_{12} \\
  r_2 &= i \left( \sigma_{12} - \sigma_{21} \right) = -\text{Im} \, \sigma_{12} \\
  r_3 &= -\left( \sigma_{22} - \sigma_{11} \right)
\end{align*}
\]

and

\[
\begin{align*}
  \Omega_1 &= -\Omega = -2 \frac{\gamma_2}{\hbar^2} \vec{E}^2 \\
  \Omega_2 &= 0 \\
  \Omega_3 &= \Delta \Omega
\end{align*}
\]

The effect of ionisation would be to reduce the length of the pseudopolarisation vector \( \vec{r} \) and also cause loss of coherence. The vector \( \vec{r} \) loses its phase memory if the system (atomic or molecular) undergoes a phase relaxation (i.e., observed over a time greater than \( T_2 \)). This loss of phase memory reduces the length of its projection in the plane 1 - 2.
Two Photon Resonant Pulse Propagation
and Harmonic Generation

When matter-radiation interactions are in transient regime there are many novel manifestations of the coherent interchange of energy like Photon Echo, Self-Induced Transparency etc. Most of these one photon effects have their two photon analogues. The proper use of the coherent transient interaction has many applications like Isotope Separation, Harmonic Generation etc. As a result of the finite build up time of the polarisation if an ultrashort light pulse (pulse width < $T_2$ and $T_1$, the phase and population relaxation times) is propagated off resonance through an absorbing medium, there will be spectral broadening, as well as phase modulation and frequency shifting.

The possibility of complete population inversion by transient excitation has been utilised to demonstrate various coherent phenomena. A method of isotope separation had been suggested utilising the spectral dependence of the transient excitation. The lossless slow propagation of "2$\pi$ secant hyperbolic" pulses called self induced transparency is a proof of complete population inversion.

Before an induced dipole has lost the phase memory, its coherent oscillation is determined by its previous excitation. In the "Zero Area Pulse Propagation" (i.e. in one photon case) the
phase of the electric field is suddenly reversed in the middle of the exciting pulse. The Rabi rate of transition is E for the first half of the pulse and \(-E\) for the second half, after the phase reversal. This results in transient amplification in the second half of the pulse recovering the energy that was lost to the matter due to one photon absorption in the first half, giving rise to an anomalous propagation.

The propagation of 90° phase shifted pulse pair which is the two photon analogue of "Zero Area Pulse Propagation" will be discussed here with reference to THG.

To study the TPR coherent pulse propagation and harmonic generation we need the expressions for the polarisations oscillating in the medium at the harmonics of interest induced by the fields present. The polarisation per atom oscillating at the fundamental and third harmonic frequencies, neglecting Doppler averaging (in our experiment \(T^* \approx \text{Pulse Width}\)) are given by,

\[
P_\perp(z,t) = \mathcal{P}_\perp(z,t) \ \text{e}^{i(\omega t-kz)} + \text{c.c.}
\]

where

\[
\mathcal{P}_\perp(z,t) = \left[ \alpha_1(\omega) \mathcal{E}_1 + \alpha_2(\omega) \mathcal{E}_2 \right] \mathcal{E}_1 + 2 \left( \frac{r_{12}}{\hbar} \right) \mathcal{E}_2^* \]

\[
+ \left( \frac{\omega_{21}}{\pi} \right) \mathcal{E}_2^* \mathcal{E}_3
\]  \hspace{1cm} (11.21)
\[ P_3(z,t) = P_3(z,t) e^{i \omega t - kz} + c.c. \]

where \( P_3(z,t) = \left[ \alpha_1(3\omega)\sigma_{11} + \alpha_2(3\omega)\sigma_{22} \right] E_3 + \left( \frac{e_{21}}{n} \right) \sigma_{12} E_1 \]  

These will become the source terms in Maxwell's equations for generating the corresponding harmonic fields. Under plane wave and slowly varying amplitude approximation the second order Maxwell's equations reduce to first order equations for complex amplitudes of the harmonic in the laboratory frame as

\[ \frac{\partial \xi_m(z,t)}{\partial z} + \frac{i}{\varepsilon_0} \frac{\partial \xi_m(z,t)}{\partial t} = -\frac{i \omega_m N}{2e \varepsilon_0} P_m \]

where \( N = \text{Number density of atoms} \)
\( \varepsilon_0 = \text{Vacuum permittivity} \)

In the time frame of the pulse i.e. the retarded time \( t_r = t - z/c \) we can rewrite the above equation as

\[ \frac{\partial \xi_m(z,t_r)}{\partial z} = -\frac{i \omega_m N}{2e \varepsilon_0} P_m(z,t_r) \]  

The total field seen by an observer at a reduced time slice \( t_r \) is:
A detailed theory of propagation effect taking into account the optical Stark shift, ionisation, reaction of the third harmonic field, Doppler broadening, two photon absorption has been elegantly reported in Ref 10. In this dissertation however, an attempt is made to describe analytically the essential physical phenomena involved in coherent pulse propagation and harmonic generation, taking only two photon absorption into account. The effect of the neglected factors will be described qualitatively. A theoretical simulation, with parameters very close to that of the experiment performed, will be presented in Chapter III.

In the following discussion we assume as before:

1. Detuning $\Delta \omega = 0$
2. Coherent excitation i.e. Pulse Width $< T_2, T_1$
3. No Ionisations
4. No reaction of third harmonic field
5. Neglect doppler broadening

Under these conditions the complex polarisation amplitudes $\rho_i$ and $\rho_3$ from equations (II.21), (II.22) may be written as

$$\rho_i = [\alpha_1(\omega) \sigma_{11} + \alpha_2(\omega) \sigma_{22}] e^{i\omega t} + 2 (r_{12}/\hbar) \sigma_{12} \mathcal{E}^*$$

and
The net two photon transition undergone by an atom after a pulse has gone is given by a quantity known as the "AREA" of the pulse $\Theta$ as

$$\Theta = \int \Omega \, dt$$

where $\Omega$ is the two photon Rabi frequency given in equation (II.18). Pulse propagation effect is described in two cases depending on the magnitude of $\Omega$. 

\[ R^3 = \left( \frac{\tilde{\gamma}_{21}}{\pi} \right) \bar{\Omega} \mathcal{E} \]  

(II.25)
Case I  Weak Field (\( \Omega \) is small enough to cause no sufficient two photon absorption)

Then \( \delta_{12} \approx 0 \) and \( \rho_i \approx \alpha_i(\omega,z,t) \). Putting these in equation (II.23), we have

\[
\frac{\partial \mathcal{E}}{\partial z} = -i \frac{\omega N}{2c \varepsilon_0} \alpha_i(\omega,z,t) \mathcal{E}
\]

\[
\mathcal{E}(z,t) = \mathcal{E}(0,t) e^{-i \frac{\omega N}{2c \varepsilon_0} \alpha_i(\omega,z,t) z} = \mathcal{E}(0,t) e^{-i \phi}
\]

Note that "t" here is really the reduced time \( t_r \). Depending on the proximity of an intermediate state with respect to the fundamental \( \alpha_i(\omega,z,t) \) will be different for different \( \omega \)'s in the pulse spectrum. So the phase \( \phi = \frac{\omega N}{2c \varepsilon_0} \alpha_i z = k_{med} z \) will have an \( \omega \) dependence in \( k_{med} \) (contribution of the medium to the wavevector). Hence there will be dispersion. In our case the intermediate state 2P in lithium is quite off resonant so that the pulse will propagate with negligible absorption and small dispersion.

CASE II  Strong Field (\( \Omega \) is enough to cause sufficient two photon absorption)

Putting equation (II.24), in (II.23) with \( m = 1 \) gives

\[
\frac{\partial \mathcal{E}}{\partial z} = -i \left( \frac{\omega N}{2c \varepsilon_0} \right) \left[ \alpha_1(\omega) \delta_{11} + \alpha_2(\omega) \delta_{22} \right] \mathcal{E}
\]

\[
-2i \left( \frac{\omega N}{2c \varepsilon_0} \right) \left( r_{12} / m \right) \delta_{12} \mathcal{E}^* 
\]
\[
\left( \frac{\partial E}{\partial z} \right) = \left( \frac{\partial E}{\partial z} \right)_{\text{NP}} + \left( \frac{\partial E}{\partial z} \right)_{\text{TPA}}
\]

where the first term is the effective Nonlinear Polarisability

\[
\chi(\omega,z,t) = \chi_1(\omega,z,t) \sigma_{11} + \chi_2(\omega,z,t) \sigma_{22}
\]

and the second term is due to the Two Photon Absorption (TPA). These two terms will be described separately.

\[
\left( \frac{\partial E}{\partial z} \right)_{\text{NP}} = -i (\omega N/2c \varepsilon_0) \chi(\omega,z,t) E
\]

\[
E_{\text{NP}}(z,t) = E(0,t) e^{-i(\omega N/2c \varepsilon_0) \chi(\omega,z,t) z}
\]

\[
\varphi(\omega,z,t) = -i \varphi(\omega,z,t)
\]

\[
E_{\text{NP}}(z,t) = E(0,t) e^{-i \varphi(\omega,z,t)}
\]

(II.26)

The time dependence in \( \varphi(\omega,z,t) \) comes from the strong modulation of \( \sigma_{11}(t) \) and \( \sigma_{22}(t) \) due to heavy two photon absorption. The quantity \( \varphi(\omega,z,t) \) will give additional frequency contribution to the pulse spectrum and is called the Self Phase Modulation (SPM) or Chirp. \( \varphi(\omega,z,t) \) will also give dispersion as in case 1.

The other term is
\[
\left( \frac{\partial \mathcal{E}}{\partial z} \right)_{TPA} = -2i \left( \frac{\omega \hbar / 2c \varepsilon_0}{r_{12} / \hbar} \right) \sigma_{12} \mathcal{E}^* \tag{II.27}
\]

Assuming square pulse excitation of width $\tau$ we can solve equation (II.16) as

\[
\sigma_{12} = i \left( \sigma_{22} - \sigma_{11} \right) \frac{\kappa_{12}}{\kappa_1^2} \mathcal{E}^2 \tau \tag{II.28}
\]

Using this in the above equation we have,

\[
\left( \frac{\partial \mathcal{E}}{\partial z} \right)_{TPA} = 2 \left( \frac{\omega \hbar}{2c \varepsilon_0} \right) \left( \frac{\kappa_{12}}{\kappa_1^2} \right) \left( \sigma_{22} - \sigma_{11} \right) |\mathcal{E}|^2 \tau \tag{II.29}
\]

now,

\[
\frac{\partial |\mathcal{E}|^2}{\partial z} = \mathcal{E} \frac{\partial \mathcal{E}^*}{\partial z} + \mathcal{E}^* \frac{\partial \mathcal{E}}{\partial z} \tag{II.30}
\]

Putting $\frac{\partial \mathcal{E}}{\partial z}$ from above,

\[
\frac{\partial |\mathcal{E}|^2}{\partial z} = -2 \left( \frac{\omega \hbar / c \varepsilon_0}{r_{12} / \hbar} \right)^2 \left( \sigma_{11} - \sigma_{22} \right) \tau \text{ Re} \{ |\mathcal{E}|^4 \} \tag{II.31}
\]

Equation II.31 is the Two Photon Beer's law as, valid only for the distance over which the pulse shape is conserved:

\[
\frac{\partial I}{\partial z} = -\beta I \tag{II.31}
\]

the two photon absorption coefficient $\beta$ is,
\[ \beta = 2 \left( \omega \frac{N}{e \epsilon_0} \left( \frac{r_{12}}{\hbar^2} \right)^2 \left( \sigma_{11} - \sigma_{22} \right) \right) \tau \]

The energy density of the light fields at \( \omega \) and \( 3\omega \) (neglecting fifth harmonic) is dissipated in various ways as they propagate through the medium. The total energy density of the light fields at \( \omega \) and \( 3\omega \) is given by,

\[ U = U_1 + U_3 = 2c \epsilon_0 \int \left( |\varepsilon_1|^2 + |\varepsilon_3|^2 \right) dt \]

Finding \( \frac{\partial |\varepsilon_1|^2}{\partial z} \) and \( \frac{\partial |\varepsilon_3|^2}{\partial z} \) from Maxwell's equations (11.23) and using the polarisation components from equations (11.21) and (11.22) the change of the total energy density with propagation distance is,

\[ \frac{\partial U}{\partial z} = -N \hbar \omega \left[ 2\sigma_{22} + \int \left( 2 \sigma_{22} T_1 + 3 \sigma_{22} \sigma_{22} + 5 \gamma_2 \sigma_{22} + 3 \gamma_1 \sigma_{11} \right) dt \right] \]

Where the first term is due to two photon absorption, the second term is due to spontaneous emission and the last three terms are due to ionisation. The dominant loss will be due to the two photon absorption. By a careful choice of the coherent excitation, the depletion of the fundamental due to the two photon absorption process may be minimised. This is done by a pair of "90° phase shifted pulses" which is a pulse sequence
with a $90^\circ$ phase shift between them.

The amount of third harmonic generation is determined by the ratio $\frac{\gamma_{21}}{\tau_{12}}$. A theoretical simulation of the two photon absorption and third harmonic generation of "$90^\circ$ phase shifted pair" of picosecond pulses is given in the next chapter. There it will be shown that the detuning can be made zero only over a little portion of the real pulse. This is due to the intensity dependent A.C. Stark Shift induced by the electric field of the pulse.

The effect of ionisation is an irrecoverable loss. Strong ionisation gives rise to loss of coherence too. If the energy relaxation time $T_1$ or the phase relaxation time $T_2$ are comparable to the total interaction time then there is loss of coherence and "$90^\circ$ phase shifted pair" of pulses will no longer result in lossless propagation. Doppler broadening can be neglected if the inverse doppler width is much longer than the pulse duration. If they are comparable, then because of the line broadening, the pulse will interact more strongly with one velocity group of atoms than with another. This will effectively contribute to losses and the absorption length will decrease. A plot, demonstrating the influence of Doppler broadening on two photon coherent pulse propagation is given by Diels et al in Ref.10, where they have shown that the presence of Doppler Broadening doubles the absorption loss. They have discussed the Third Harmonic Generation for both thin samples as well as for long propagation distances which can be
drastically different depending on the phase matching achieved in thick samples. In the short pulse approximation (\( t_p \ll T_1, T_2 \) and the inverse Doppler width \( T_2^* = \sqrt{\pi/\sigma_D} \) ) the efficiency for harmonic generation is the same for pulses of same energy and shape but different widths.

The increase of pressure by a factor two doubles the number density of the atoms and doubles the collisional broadening. The change in temperature involved in doubling the pressure brings no significant change to the Doppler broadening. The efficiency of harmonic generation is unaffected by a change in pressure.

Phase matching plays the most important role in pulse propagation and harmonic generation over long distances. The phase matching in steady state is described as a spatial periodic shuttling of energy between the input field and the generated wave (Refs 26,27). The length of this periodicity is called "Coherence Length". The phase matching in our coherent transient case however is more complicated since different frequency components of the pulse spectrum (both the fundamental as well as third harmonic) travel at different speeds because of the dispersion described before. Different temporal portions of the pulse (in the time domain) also travel at different velocities, because the susceptibilities vary with the population along the pulse. The "ideal" coherent propagation condition sought is not only that where the maximum energy is transmitted, but also such that, in the retarded
frame, the susceptibility remains constant in time and space for a portion of the signal.
CHAPTER III

TWO PHOTON RESONANT PULSE PROPAGATION
THROUGH LITHIUM VAPOR

In this chapter the theory of TPR pulse propagation and harmonic generation (outlined in chapter II) will be applied to the 2S-4S transition in lithium vapor. The term diagram of lithium will be presented in the first section. The second section will describe the theoretical simulation, while the theoretical results will be given in section three. Experimental conditions needed to see this effect will be outlined in the last section.

Term diagram of Lithium

The term diagram is given in Fig 3. The two photon resonance transition under study is 2S - 4S as marked in the figure. Since $\Delta \ell = 0$, two photon transition is allowed. The unstarkshifted resonance is at $\lambda = 571.2$ nm. The presence of the intermediate off resonant state 2P is the dominant term in $r_{12}$ and helps the two photon transition. Absorption of another photon from 4S level takes the transition to the continuum and a third harmonic photon is generated. Besides the
THG, transition from the 4S level contribute substantially to ionisation loss.

Theory applied to 2S - 4S transition in Lithium

The experimental set up described in Chapter IV provides data on energy absorption and harmonic generation for a pair of gaussian pulses, as their relative phase is continually scanned between zero and $2\pi$, and their relative delay is incremented (continuously) from zero to two pulse duration (FWHM). In this section we produce a set of theoretical curves simulating the experimental situation. Comparison of the measured and calculated data will not only confirm the adequacy of the theoretical model, but in addition enable us to determine the exact value of various physical parameters of lithium. It is appropriate at this point to discuss briefly the type of excitation applied to the lithium vapor. The laser pulses used to generate the sequence are gaussian in shape of fixed duration. Since the signal is created by a coherent superposition of two pulses separated by less than the pulse duration, the energy of the applied excitation is also a function of their relative phase and delay. For instance the average frequency of a $90^\circ$ phase shifted pulse pair is higher than that of the original laser pulses while that of a $270^\circ$ phase shifted pulse is lower.

Though the optical table was "vibration free", there is
always a very slight (of the order of $\lambda$) movement of the mechanical parts. This is enough to throw the relative phase between the pulses out of $90^\circ$ randomly anywhere between $0^\circ$ to $360^\circ$. So instead of trying to fix the delay arm of the interferometer, we translated it so all the delay (positive as well as negative) and the phase could be scanned. In other words an autocorrelation of the pulse was taken. During the autocorrelation scan, the two pulses (one from the fixed arm and one from the delay arm) would sweep through a phase cycle between their electric fields for every delay of a wavelength. For every one of this cycle the pair of pulses pass instantly through the two phase points $90^\circ$ and $270^\circ$ during which the pulses behave like a "90$^\circ$ phase shifted pair", as far as the two photon transition is concerned. The pulses in the experiment are gaussian type and the well known interferometric autocorrelation maxima and minima (Ref 28) will be determined by the delay of the pulses. Due to interference, the superposed electric field of the two pulses swing through maxima and minima and in doing so generates a similar oscillating stark shift. The excitation is exactly on resonance when the fundamental frequency $\omega$ is exactly half of the stark shifted $2S - 4S$ transition in lithium. During this resonance condition if the pair of pulses have a relative phase delay of $90^\circ$ or $270^\circ$, a coherent recovery of the two photon absorption takes place and an anomalous transmission results. Maximum third harmonic generation is possible in this condition and if
phase matching is possible (done with Mg vapor) then an energy conversion efficiency of several percent is predicted (Ref 10).

In the theoretical simulation we considered a thin sample of lithium vapor, at a pressure of 10 Torr. Pulse energy given by the "Area" of the pulse was taken such that it sufficiently excites the two photon transition. The detuning was scanned by varying the fundamental frequency $\omega$ of the incident pulses to observe the effect of Stark shift on the resonance condition. A numerical computation is made of the interferometric second order autocorrelation function $F(\tau)$ where,

$$ F(\tau) = \frac{\int \left[ E(t) + E(t-\tau) e^{-2\pi i \nu \tau} \right]^2 dt}{2 \int |E(t)|^4 dt} $$

Excluding the rapidly varying terms $\cos(2\pi \nu \tau)$ and $\cos(4\pi \nu \tau)$ we have the intensity autocorrelation function $I(\tau)$ as,

$$ I(\tau) = \frac{1 + 2 \int |E(t)|^2 |E(t-\tau)|^2 dt}{\int |E(t)|^4 dt} $$

The results of the computation appear in the next section.
Theoretical Result

In the experiment a microcomputer stores the data of both interferometric as well as intensity autocorrelations. The theoretical results showing the characteristics of several Stark tuning in nonlinear transmission are illustrated in Fig. 4. The second order interferometric autocorrelation of the transmitted fundamental pulses (bold trace) and third harmonic signals (light trace) are plotted. In both sets of curves the upper trace is the envelope of the maximum intensity points and the lower trace is the envelope of the minimum intensity points of each individual fringe in the interferogram. The pulse area which is proportional to the pulse energy was equal to 1. Far below resonance (right side of Fig. 4) where detuning is

\[ \Delta \omega = 12.10 \times 10^{-1} \] s , the pulse does not interact with the line and the autocorrelation is unchanged from the one without lithium. At a detuning of \( \Delta \omega = 9.10 \times 10^{-1} \) s (which corresponds to six inverse pulse widths) below resonance, the top of the autocorrelation is truncated. This is because near zero delay, the Stark shift corresponding to the coherent superposition of the two pulses is sufficiently large to bring the resulting peak electric field into resonance.
With increasing laser frequency (i.e., decreasing detuning) the "hole" burnt by the resonance line in the autocorrelations move towards larger delays. In the last case, the coherence region extends very far in the wings of the autocorrelation, far beyond the limits of the pulse overlap. Near zero delay the second harmonic signal can be larger than that of the incident pulses, because of pulse reshaping (shortening). The computer program that was used to solve the Maxwell - Bloch equations is given in Appendix B.

The third harmonic intensity varies also with delay, frequency and phase. The trace of the maxima and minima of each phase cycle is shown by the light traces of Fig 4. The envelope of the third harmonic versus delay far below

\[ \Delta \omega = 12.10 \text{ s} \]

resonance is a third order interferometric autocorrelation, with a peak to background ratio of 32 to 1. In general the shape of the upper envelope of the third harmonic versus delay follows qualitatively that of the second harmonic of the fundamental signal versus delay. The situation is different in the case of propagation through thick samples. It will be shown in the experimental section that utilising coherent propagation effect it is possible to enhance harmonic generation from that of a single pulse generation.

To observe the anomalous transmission effect and enhancement of third harmonic utilising atomic coherence induced by a pair of "90 phase shifted" pulses, the
The experimental requirements are rather stringent. The source has to be able to generate high power tunable picosecond pulses with an uniform wavefront— an essential condition in order to be able to define the phase. One needs to be able to adjust the relative phase and delay between the two pulses of the sequence. The phase or frequency modulation of the pulses should be small, otherwise it is not possible to define a phase for these pulses. Finally resonant coherent interaction is strongly dependent upon the pulse shape and frequency which are not particularly easy to determine for picosecond pulses. The experimental apparatus developed to demonstrate the coherent effect is discussed in the following chapter.
CHAPTER IV

EXPERIMENTAL APPARATUS

This chapter describes the experimental apparatus that was used to observe anomalous transmission and enhancement of harmonic generation by a sequence of "90° phase shifted" pair of pulses propagating under TPR (2S - 4S) condition through lithium vapor. Some of the experimental difficulties that had to be overcome are,

1. Generation of pulses of several picosecond (we had 6 psec pulses) duration, about 1 mJ energy per pulse, with a flat wavefront, Gaussian beam profile, without significant self phase modulation (SPM). This is described in sections 2 and 3.

2. Splitting of these pulses into a sequence of two identical pulses of well defined relative delay and phase and designing a mechanism for monitoring the Phase Difference between them. This is described in the fourth section.

3. The data acquisition system by which even at an extremely slow repetition rate (10 Hz) of the amplified pulses, a fringe resolved second order autocorrelation could be recorded. This is described in the fifth & sixth sections.

4. Production of a homogeneous mixture of lithium and magnesium (for phase matching) vapors near 1100°K. This is
described in seventh section.

The Schematic

The set up of the experiment is shown in Fig 5. Pulses of duration 6 ps (FWHM) whose center frequency is tuned to the TPR transition 2S–4S of lithium (wavelength of 571 nm) are produced by a synchronously pumped dye laser. These pulses are then sent through a three stage amplifier (Ref 29) to be amplified to millijoule energy levels. The repetition rate of the amplified pulses is 10 pps, set by the rate at which the ND:YAG laser pumped the three stage amplifier. About 1% of the amplified pulse energy is coupled out by a beam splitter (a flat glass mounted near Brewster’s angle) to be passed through a KDP crystal for second harmonic generation (SHG) and detected by detector D1 after passing through several visible blocking, UV transmitting filters (SCHOTT UG11). The signal of D1 monitors the energy of the amplified pulses and is used as an energy normalising factor. The main beam after the beam splitter is sent through a Mach Zender Interferometer which monitors the phase coherence of the pulses, as well as the uniformity of the wavefront, and serves as a pulse sequencer. One of the two complementary outputs of the interferometric delay line is sent through the interaction region (the heat pipe), while the other is detected by detector D2 after SHG and several cut off filters. To adequately define the relative
phase of the pulses transmitted by the interferometer, their
spacing is monitored with an accuracy better than 1/20 of the
wavelength, by sending a He-Ne laser beam through the same
interferometer and measuring the successive constructive-
destructive interferences as detected by detector D4. The same
beam splitter used to sample the amplified pulse is used to
divert the He-Ne interference fringe into detector D4 (Fig 5).

As mentioned above, one of the two outputs of the
interferometric delay line is a two pulse sequence sent
through the resonant medium: a heat pipe containing the
resonant medium, lithium vapor and nonresonant medium,
magnesium vapor for phase matching. After transmission from the
heat pipe the third harmonic and fundamental beams are split
by a couple of interference filters (Corion 190nm with 20nm
bandwidth) which transmits the third harmonic and reflect the
fundamental (at near normal incidence). The third harmonic is
detected by a photomultiplier (PMT) tube (Hamamatsu tube R928).
The fundamental beam after SHG and filters is detected by
detector D3 whose signal gives the transmission.

For obvious reason of clarity, the data acquisition
system is not shown in the schematic. All the analog signals
from the detectors D1, D2, D3, D4 and PMT are passed through a
home built "hold and digit" circuit to a "Smoke Signal
Broadcasting" microcomputer with two 8 floppy disk drives,
which is our data station. The software needed for data
acquisition and signal averaging, done by the program called
"test" is given in Appendix A.

The Laser Source

The source of tunable 6 psec pulses were generated by a Spectra Physics Synchronously Pumped dye laser system. An acousto-optically modelocked Ar-ion laser (Spectra Physics model no. 171) pumped a dye laser (Spectra Physics model no. 375) synchronously. By careful modelocking of the Ar laser at 514.5 nm we were able to obtain very stable pulses of duration 120 psec (no adjustments were needed for at least a couple of months). These pulses were detected by a very fast detector (Spectra Physics model no. 403B ), sampled by a sampling unit (Tektronix sampling head s - 6 and trigger unit S-53,) and displayed on a Tektronix Oscilloscope (model no.7603). The amplitude fluctuation was seen to be about 2% and the temporal jitter was 5 psec. The tube current corresponding to this modelocking was 42 amperes and the average power out was 1.05 Watts. These modelocked pulses at 514.5 nm with a repetition rate of 82 MHz. pumped the dye laser synchronously; i.e., the cavity length of the dye laser was made slightly shorter than that of the Ar laser to within a micron. By this the longitudinal mode spacing \(\frac{c}{2\ell}\) of the argon laser pulses were matched nearly with the longitudinal mode spacing of the dye laser. The dye that was used in the dye laser was Rhodamine 6G (concentration 2.7 milli mole) dissolved in ethylene glycol.
A triple plate Birefringent Filter (Coherent) was used to smoothly tune the center frequency of the picosecond pulses. The filter introduces also a bandwidth limitation, resulting in larger (6 ps) but more stable pulses than generally available from a synchronously pumped system. At the output of the dye laser we had 6 psec pulses tunable around 571 nm with repetition rate of 82 MHz, and average power of 200 mW. The beam diameter at the output was only 1 mm. Since the absorption length at 1 Torr of lithium vapor is 13.5 cm (Ref 10) we had to amplify the pulses to maintain about 50 mJ/cm² over several characteristic absorption lengths.

An interferometric second order autocorrelation of the picosecond pulses after it leaves the amplifier is given in Fig.6.

Picosecond Pulse Amplifier

We have developed a picosecond laser oscillator-amplifier system (Ref 29), capable of producing single pulses of 1 mJ energy, at a repetition rate of 10 Hz, tunable around 571 nm. The amplifier is sketched in Fig.7, with three stages of amplification. The amplifying element at each stage is a flow-through dye cell (Quartz). The first two stages were built from ordinary spectrophotometric cuvettes with a crosssection of 1 cm by 1 cm square (Hellma Cells). The third dye cell was a cylindrical cell (Hellma Cell) of length 5 cms. and diameter
2.5 cms. The first two stages are pumped transversely and the third one longitudinally. Coupling between the first and second stages is minimized by synchronising the first stage at the leading edge, while the second stage is synchronised at the trailing edge of the pump pulse.

Several dyes and solvents have been investigated to search for the maximum gain and minimum absorption at 571 nm. The combination of Hexafluoroisopropanol (HFI) with Rhodamine 6G shifts sufficiently the absorption and fluorescence spectra of Rhodamine 6G to meet our condition. In view of the high costs of HFI, mixtures of water and HFI were investigated, which was believed to combine the spectral shift of HFI and thermal conductivity properties of water. Unfortunately, the desired spectral shift was not found in the water solutions, which separated into two nonmiscible components after a few days. For operation at 571 nm, the choice of dye solutions in the three stages are,

First Stage: 0.18 mM concentration of Rhodamine 6G in Ethanol
Second Stage: 0.14 mM concentration of Rhodamine 6G in Ethanol
Third Stage: 0.01 mM concentration of Rhodamine 6G in water and ammonix

Beam Profile: Careful positioning of the pump beams relative to the amplified beam in the last two stages provide
complete control over the spatial intensity distribution. A gaussian spatial beam profile of the amplified beam is shown in Fig 8.

**Spatial Coherence:** The demonstration of achieving uniform extinction of the beam in the Mach Zender Interferometer proved the spatial coherence of the amplified pulse.

**Temporal Coherence:** This is crucial to the experiments of coherent propagation and has been used to test the laser sources, the scanning technique and the data acquisition. The amplified pulse (shown in the autocorrelation trace in Fig 6) can best be represented by a Gaussian temporal profile with a phase modulation proportional to the pulse intensity (\(=1.43 E^2\)). The small phase modulation corresponds to an increase of bandwidth duration product of only 10%, hence too small to be determined by conventional spectral measurements.

The Interferometric Delay Line

The pulse sequencer or the interferometric delay line is sketched in Fig 9. It was built entirely of prisms instead of dielectric beam splitters and mirrors for the following reasons:

1. High peak power may damage the dielectric coatings.
2. It can be used for all wavelengths
transparent through the glass used.

3. The beam splitting ratios can be continuously adjusted for any polarisation or wavelength.

The 50 - 50 beam splitting is achieved by two air-gap prisms with pressure adjustment by thumb screws as shown. At the input, the pulses are split into beams of equal energy. One of the beams is sent to the adjustable delay consisting of a prism on a translation stage. The other beam is sent through a fixed optical delay. Both beams recombine at a similar beam splitter as in the input side and exit the interferometer via two complementary outputs. The two outputs are complementary in the sense that when one output gives maximum energy the other gives minimum and vice versa, the total energy always remaining the same. The periodicity of the oscillation of energy in each of the two outputs is determined by the speed of the translation stage, i.e., the rate at which the delay is scanned.
The rate at which the data can be acquired is limited by the amplifier to 10 Hz. At that acquisition rate in order to achieve a phase resolution of \( \frac{2\pi}{20} \) the translation stage has to move at an uniform rate of 0.1 micron per second! Even at that translation speed, the required phase resolution will not be achieved because of the pulse to pulse intensity fluctuation of the amplifier. There is in addition a minimum feasible speed at which an uniform translation is possible. Reducing the average speed below 0.1 micron per second resulted in a motion consisting of "jumps". The larger static friction was sufficient to stop the motion, until it was overcome by the driving force, resulting in an advance of several wavelengths under the smaller dynamic friction, followed by another immobility.

At larger speeds, the translational motion of the delay arm was uniform, but the speed showed periodic fluctuation representative of the inaccuracy of the individual wheels of the reducing gear.

The "minimum" speed problem imposed an averaging method over several wavelengths to extract the required phase resolution. The speed fluctuation due to inaccuracies in the gear boxes called for a continuous monitoring of the optical delay. This absolute delay measurement was made by monitoring the constructive-destructive interferences of a He-Ne beam retracing the dye laser beam path through the interferometer (Fig 5). The delay between successive maxima (or minima) as
recorded by D4 (Fig 5) corresponds to a distance of 632.8 nm, the He-Ne laser wavelength. Since the CW He-Ne laser beam has a much larger coherence length than that of the picosecond pulses, the distance calibration remained valid even when the pulses were completely separated. In this experiment, the He-Ne laser serves as a wavelength standard to measure the optical delay and determine the wavelength of the dye laser. This is the basis on which our data acquisition program ("Test") was built. A new method ("Wavemeter") for finding the average wavelength of a picosecond pulse can be found in Ref 14.

The Detectors & The Microprocessor

Detectors: The detectors D1, D2, D3 consisted of a frequency doubling KDP crystal, several SCHOTT UG11 filters and a detector box. The detector box had a broad area (of uniform active area of 1 cm square) UV, EG & G detectors and a preamplifier circuit. The signal from the detectors is proportional to the energy incident on the photodiode. The detectors were shown to be linear over the dynamic range used in these experiments. The visible detector D4 was a p-i-n diode from Coherent whose signal is also suitably amplified to give an appreciable signal in the 0-10V range. The photomultiplier tube (Hamamatsu R928) gave current signals proportional to the third harmonic (190 nm) energy. This was
converted into a voltage pulse in the range 0-10V.

The analog signals from all the five detectors were fed into five channels of a home built peak detect and hold circuit. Then the signals were digitized and fed into the microcomputer. For triggering the computer to enable it to read the digitized signals from all the detectors, we used the pulsed signal going from the divider clock (which synchronises the RF modelocker output with the YAG laser) to the Q-Switch trigger.

The Microcomputer: The microcomputer that was used for data handling, performing statistics and storing in floppy diskettes was a "Smoke Signal Broadcasting" computer (model: Chieftain). It had a Motorola 6800 microprocessor with 64 K bytes of storage. The software that was built to handle the data acquisition is described in the next section.

The Data Acquisition Program

The hardware of our data acquisition system is schematically represented in Fig.10. About 1% of an amplified pulse is used as a reference beam, while the remaining part is sent through the interferometer. The detectors D1, D2 & D3 detects the second harmonic of the fundamental pulses. This second harmonic scheme serves two purposes. It discriminates the amplified pulses from the non amplified pulse train (which has higher average power) and it
provides a more accurate transmission measurements. The PMT detects the third harmonic energy and D4 monitors the He-Ne fringes. The signals from the five detectors are amplified, digitised and fed into the computer as shown in the figure.

The software "Test" which runs the experiment uses a "discriminator" in the signal (D1) of the reference beam which rejects data from pulses with either too low or too high amplified pulse energies. In addition to throwing out data with extreme pulse energies the "discriminator" partitions the data into "energy windows" (typically three), whose limits are set by the user depending on the energy fluctuation of the amplified dye pulses. This had to be done since coherent pulse propagation effect depends very strongly on the pulse energy.

It was pointed out in the last section that in order to trace out the individual fringes in the autocorrelation curve the maximum speed of the translation stage can be as slow as 0.1 micron per second which is incompatible with smooth motion and the He-Ne fringes as monitored by D4 gave the distance calibration for determining the delay and phase with interferometric accuracy.

The computer program "Test", given in Appendix A runs the computer to take data, partition them into three "energy windows", does the averaging and finally stores them in the floppy diskettes. The errors due to amplitude fluctuation of the pulses are eliminated by energy normalizing the signals of D2, D3 and PMT (third harmonic) i.e. dividing these signals by
the signal of D1. While varying the delay in the autocorrelator the He-Ne fringes detected by D4 are tracked, so that phases and delays could be calculated for the dye pulses (at $\lambda = 571$ nm) using the ratio of the two wavelengths. The resulting dye fringes are partitioned over large number of sets (each set has typically 100 fringes). Per set two different averaging processes are performed. On one hand all the data in each set are averaged out, thereby washing out the fringes completely. In the absence of sample cell, this averaging yields the conventional 3:1 intensity autocorrelation curve. On the other hand an average fringe per set is constructed, by averaging out the data on a single periodicity over the whole set. This averaging process makes it possible to track small phase shifts over consecutive sets of fringes. In the absence of sample cell, the maxima and minima of each fringe, as a function of delay, retraces the interferometric autocorrelation curve (peak to background ratio of 8:1). In this way one obtains, during a single experiment, both the phase and intensity information.

The amount of averaging that can be done is directly related to the duration and coherence of the pulses. The criteria are those of slowly varying envelope approximation. The averaging can only be done over the number of cycles over which the pulse amplitude and phase can be considered to be constant. With our 6 psec pulses, the maximum number of periods over which the data could be averaged is at most $N=300$. 
As the delay is being continuously scanned, the data per set are accumulated for a total delay increment of $X_{max} = N \lambda$ where $\lambda$ is the wavelength of the pulse and sorted according to their relative phase:

$$\Delta \phi = 2\pi \left( X - N \lambda \right) / \lambda$$

The accuracy of the averaging process is directly related to the accuracy in the determination of the wavelength of the dye laser pulses. If for instance we choose to divide the $2\pi$ phase interval into 25 segments and average the data over $N=100$ periods, the maximum allowable error in the wavelength specification is $\lambda/2500$. The example in Fig.17 shows such an averaging over 100 periods of the second harmonic signal as recorded by detector D1, D3, and the PMT after energy normalisation. In that particular recording, the data points were sorted in 25 phase sectors. The accuracy of this averaging process, however, can be improved by computing, for each phase sector, the average phase as well as the average signal. This procedure is particularly useful in the cases where the signal has a complex dependence on the phase difference.

The Heat Pipe

Three successive prototypes had to be constructed to evolve from the basic heat pipe design of Vidal & Cooper (30) to an instrument which confines a homogeneous, uniform and
transparent mixture of Lithium and Magnesium vapors and that can be operated continuously for several days at 1000 °C. The final working heat pipe is sketched in Fig. 11. It consists of two stainless steel concentric pipes which are isolated from each other. The inner pipe contains the homogeneous mixture of Lithium and Magnesium whose total pressure is determined by the buffer gas (He) pressure which confines the vapors to the central region of the pipe. The inner pipe also contain two wicks (fine steel mesh) attached to the inner wall which are separated by 10 cms, leaving the central zone free. The wicks draw by capillary action the molten lithium towards the central part, where it vaporises and by convection current goes to the cooler end of the vapor zone where it condenses on the wicks. This way it acts both like a differential pump and a purifying process.

The outer pipe is isolated from the inner one and has a wicks connected to the inner wall of the outer pipe and the outer wall of the inner pipe and an interconnecting wick to avoid hot spots and provide uniform flow of lithium vapor in the region. The outer pipe contains lithium vapor at a pressure set by its buffer gas (He) pressure and acts like a thermal bath for heating the inner pipe with very high thermal conductivity. The buffer gas pressure $P_b$ in the inner pipe is always kept greater than the buffer gas pressure $P_a$ in the outer pipe. From the vapor pressure curves of lithium and magnesium given in Fig. 12 we see that the outer pipe with
pressure $P_a$, temperature $T_a$ tries to heat up the inner pipe to the same temperature. But at temperature $T_a$ the vapor pressure of magnesium being much larger than lithium, some of it will escape to the outer region. The central portion of the inner pipe has thus saturated vapor of lithium at pressure $P_a$, mixed with unsaturated vapor of magnesium at a pressure $P_b - P_a$. This central region constitutes our interaction region. The distribution of vapor pressure and temperature along the inside of the inner pipe, is sketched in Fig. 13, when the heat pipe oven operates ideally. The central region of length $L$, with no wick, has a temperature $T_a$ (same as the outer pipe temperature) confining saturated lithium with partial pressure $P_a$, and unsaturated magnesium vapor at a pressure $(P_b - P_a)$. Outside this central region the temperature drops to $T_b$ determined by the vapor pressure curve (Fig 12) of magnesium at a pressure $P_b$. There is no partial pressure of lithium in this region. The length of the vapor zone at temperature $T_b$ is determined by the input power. Outside this region the magnesium condenses and the inert gas (He) fills this space with pressure $P_b$. This buffer gas prevents any chemical reaction of the metal vapors with the windows.

The power consideration for heat pipes is very important and a careful balance of power input and power dissipated ensures a perfect operation. The outer pipe wants to operate at a temperature $T_a$ while the inner pipe wants to operate at $T_b$ ($T_a > T_b$). As a result the inner pipe cools the outer pipe. A
stable equilibrium is achieved dominantly through the cooling chambers of both pipes. The ideal input power of the heaters would be equal to the power required to maintain the heated central portion of length L at a temperature $T_a$. A detailed account of similar heat pipes is given in Refs 30 & 31.

Not only has the heat pipe to be perfectly sealed, but in addition, the outgassing of the walls has to be continuously and selectively (without pumping the He buffer gas) eliminated during the experiment. The selective pumping was achieved with cryogenically cooled molecular shives.

Another problem is the accumulation (frost) of solid lithium at the edges of the hot zone. The resulting accumulation over several hours of an annular ridge of solid lithium has been sufficient to block the laser beam in the first two prototypes. This problem was solved by a larger diameter of the inner pipe in the final version of the heat pipe, but it still limits its continuous operation to roughly one week. After complete clogging of the inner pipe it was found possible to completely clean the pipe by flowing water through it continuously for a couple of days. By this all the lithium, magnesium and their oxides could be cleaned and the pipe reused.

Measurements of phase matched third harmonic generation have been made in homogeneous mixtures of lithium & magnesium. Because of the high sublimation vapor pressure of Mg, a sizeable fraction is lost and enters the vacuum system during the heating process. A possible solution to this problem would
have been to introduce magnesium in the hot heat pipe (after the lithium had melted) which was abandoned since it was too hazardous. Anyway, the useful life span of the heat pipe between refills, appeared to be more limited by lithium accumulation than by magnesium depletion.
CHAPTER V
INTRODUCTION

The data that were taken with the 2S - 4S transition in lithium will be reviewed in this chapter. At first the 30% fluctuation of the second harmonic of the amplified pulse energies as recorded by the reference beam detector D1 was carefully examined. Three "energy windows" were selected judiciously so that only the very extreme pulse energies were left out. Keeping in mind that the theoretical simulation was done only for monoenergetic pulses and coherent effect of pulse propagation is very sensitive to pulse energies, we had to narrow the width of the "energy windows" as much as possible. But narrowing the window too much means that there will be a smaller number of data depending upon the standard deviation of the fluctuation of the pulse energies. However, to be able to trace out individual fringes a minimum number (say 18 points per period at least) of data points are needed. Clearly, there was a trade off in the choice of "energy window" width, so that it does not average over the effects of a broad range of pulse energies and not lose too many data points. Usually, the pulse energies are sharply peaked about a mean energy and the middle "energy window" could be chosen quite narrowly and still have about 65% of the total data points
falling in its range. The averaging process was therefore more reliable for the middle window than for the other two. Almost all the data points presented in this dissertation are taken from the middle "energy window".

The ratio of the signals of detectors D3 and D1 versus delay give a recording of the second harmonic energy of the transmitted (through lithium heat pipe) signal, versus relative phase and delay of the incident pulses. The autocorrelation of the incident pulse is given by the ratio of the signals of detectors D2 and D1. The signal from the PMT divided by the signal from D1 gives the third harmonic signal.

The data acquisition system produces a pair of maximum (upper trace) and minimum (lower trace) points for each delay, which represents the maximum and minimum signal (second or third harmonic) for the average fringe (made of an accumulation of data for typically 100 fringes). Each of these average fringe is stored in the computer to investigate the relative phase dependence between the incident, transmitted and the third harmonic signals. Experimental results are presented in the next section, where the first subsection will illustrate the anomalous transmission and coherent transient pulse propagation effect in two photon resonance conditions that was experimentally observed for the first time. The subsequent subsection will present data on nonlinear transmission and harmonic generation. The phase dependence between the third harmonic and the incident signals
will be illustrated. Lastly, data on enhancement of third harmonic generation under phase matched conditions will be presented.

This chapter will end with a discussion section elucidating the several limiting factors of this experiment.
Experimental Results

Anomalous Transmission

The observed anomalous transmission is shown in Fig. 14. There the interferometric autocorrelation of the incident pulses (circles) and the signal of the transmitted pulses (triangles) are shown. The "Dip" representing the two photon resonance of the incident field with the Stark shifted 2S - 4S transition in lithium vapor appears at a delay of 2.5 ps. This experimental recording can be correlated to the theoretical situation of $\Delta \omega = 3.10 \text{ s}$ in Fig. 4. The coherent recovery or "hump" appears right after the "Dip" and continues for a few hundred fringes after which it dies due to off resonance excitation (lack of sufficient Stark shift to bring the incident field into resonance).

The phase dependence of the incident and the transmitted signals of Fig. 14 are shown for a three psec delay in Fig. 15. The dotted curve represents incident signal (D2/D1). The transmitted signal is given by the solid line. At zero delay (in Fig 14), the incident and the transmitted signals are in phase and have the same periodicity of $2 \pi \text{.}$ This is a characteristic of an off resonant process where the Stark shift produced by the superposed electric field of the pulses
(at zero delay) has sufficiently shifted the 4S level to be off resonant with the two photon incident field. It is only the intensity of the superposed field at a delay of about 2.5 psec that causes sufficient Stark shift of 4S level to bring it into two photon resonance with the incident field. This resonance condition gradually tapers off towards large delays. So at a delay of 3 psec, the coherent nature of two photon anomalous transmission of "90° phase shifted" pulses appeared as shown in Fig.15 where the periodicity of the incident signal (dotted curve) is seen to be 2π. The transmitted signal however, has a periodicity of π as predicted by the theory for "90 phase shifted" pulses. This is the signature of the coherence of the atomic polarisation in two photon resonant pulse propagation (Ref 5). When the incident pulse pairs have a zero phase difference, the two photon absorption occurs equally for both pulses, and there is an attenuation of the incident signal as seen from Fig.15. When the pulse pairs have a relative phase of 90°, however the two photon absorption by this pair of pulses will be proportional to $E^2$ for the first pulse and $-E^2$ for the second pulse which corresponds to a stimulated two photon emission. The energy that was lost to the matter by the first pulse through two photon absorption has actually been coherently recovered by the second pulse. As a result we have a propagation for the second pulse with less loss. Effectively, the two photon absorption length increases in the case of "90° phase shifted" pair of pulses. Hence
there occurs less attenuation at a phase value of 90° between the pulses in the pair. At a phase difference of 180° i.e. minimum energy incident (ideally there should be no energy at all) transmitted signal was minimum. Again at a phase difference of 270°, provided the pulse overlap is negligible, the pulses behaves exactly the same way as the "90° phase shifted" pair as far as two photon transition is concerned and so another anomalous transmission was observed.

This periodicity of \( \pi \) or a "double fringe" in a 2 \( \pi \) phase cycle was observed along the strong resonance region and the effect became less prominent as the incident field was going out of resonance due to lack of sufficient Stark shift.

**Nonlinear Transmission & Harmonic Generation**

The experimental recording of Fig.16 presents the second order interferometric autocorrelations of incident (triangles) and transmitted (circles) signals. The trace of boxes with dotted lines are the maxima and minima of fringes of the third harmonic signal. The tuning was made for a longer wavelength (than that corresponding to "zero field" resonance) of the incident field. Resonance is thus satisfied for a larger amount of Stark shift i.e. greater intensity, which is reached at a smaller delay. The resonance occurred at 2 psec. This particular recording can be assimilated to the theoretical
simulation with

\[ 1 \pm \frac{1}{11} \]

detuning \[ \Delta \omega = 6.10 \text{ s} \] of Fig. 4.

**Phase Dependence:**

A very interesting relative phase dependence of the third harmonic and transmitted signals was observed. The third harmonic signal was phase shifted from the transmitted signal by a phase angle \(-\pi/6\). This can be seen in Fig. 17 where the maxima (as well as the minima) of the third harmonic signal (dotted curve) is shifted to the left of the corresponding maxima (or minima) of the transmitted signal (solid curve) by one phase sector. Since the phase cycle of \(2\pi\) was divided into twelve phase sectors (data points), the minimum phase that could be resolved was \(\pi/6 \approx 0.5\). This relative phase shift of the third harmonic and the transmitted signals was clearly observed for all the fringes in the autocorrelation trace only where the resonance condition was satisfied. Fig. 17 illustrates the fringes at a delay of 2 psec which shows the onset of resonance.

The experimentally found phase shift of \(\pi/6\) of the third harmonic signal relative to that of the transmitted may be ascribed to be the phase angle of the third order susceptibility.

**Enhancement of Third Harmonic Generation**

The experimental recording shown in Fig. 18 represents the envelope (maxima and minima of the fringes) of the third
harmonic signal versus delay, in a phase matched mixture of lithium and magnesium (10 Torr of lithium and 15 Torr of magnesium). The wavelength of the incident field was longer than that of in Fig. 16, so that the resonance dip occurred at the zero delay. This corresponds qualitatively to the theoretical result shown in Fig. 4 for a detuning of $\Delta \omega = 9 \times 10^8$.

The "hump" in the third harmonic at 2 psec delay indicates that there has been an enhancement of harmonic generation from the value at zero delay (i.e. single pulse). The incident and transmitted signals for this recording is shown in Fig. 19. The transmitted signal in this data showed a similar "hump" representing coherent recovery. The energy conversion efficiency that was recorded in this data was 1% at the peak. Since a maximum energy conversion efficiency of 6% was predicted for a plane wave (Ref 10), the maximum of 1% for a gaussian beam is close to the theoretical limit. It should be noted that, in the case of incoherent interaction, the third harmonic energy (from the third order interferometric autocorrelation) should be 32 times larger at zero delay than at 2 psec delay. The peak efficiency of 1% corresponds thus to a forty fold enhancement by coherent interaction.
Discussion of the Technical Problems

In this experiment, the total run time for just one recording of data versus phase and delay is approximately seven hours. During this time it is a formidable task to keep all the components stabilized. A change of room temperature by one degree is enough to cause both amplitude and pulsewidth fluctuation in the synchronously pumped dye laser. Little noise will be amplified a million times after it leaves the amplifier! There is the thermal lensing effect in the laser rods of the YAG laser. As the temperature of the external cooling water (which takes the heat out of the heat exchanger of the closed loop internal cooling) changes substantially it affects the cooling of the YAG rods. This results in fluctuations of the spatial profile of the YAG beam which is very detrimental to the stability of the amplified dye laser pulses. There are various factors besides the optical set up like the ambient temperature and pressure change that affects or limits the data acquisition.

The furnace of the heat pipe (at 1100 K) had to be very well insulated from the surrounding, since all the sophisticated optical equipments were very temperature sensitive. In fact we sealed it so much that heaters melted several times due to hot spots in the furnace. The heat pipe
and the associated furnace was mounted on a 5 feet by 5 feet table. The manometer gauges for pressure monitoring with the molecular shieves and the cryogenic cold traps were attached on a panel to the side of the table. Because of the space requirement of the pump station the interaction region had to be located about seven feet away from the Mach Zender Interferometer which did the pulse sequencing. Extreme care had to be taken to align the beams from the fixed and the delayed arms of the interferometer to see that they overlapped all along the way. The overlap was quite well satisfied as shown by the recording of the transmitted signal measured by the detector D3, which is situated fifteen feet away from the interferometer.

The pressure adjustable beam splitters in the interferometer posed another technical difficulty. After the pressure is adjusted for 50-50 beam splitting, elastic relaxation would cause the beam splitting ratio to change. Sometimes it would take nearly a day to stabilise to proper ratio. In addition there was a long term variation in the beam splitting ratio and almost every week it needed readjustments.

The most difficult part in the alignment of the interferometer was in being able to translate the delay arm over about 1 cm to scan the autocorrelation of the 6 psec pulses. The problem was to overlap and exactly match the wavefront of the two beams all along the travel. Over two or three mm travel the overlap was very good, but for a larger travel the
wavefronts started interfering at an angle and there appeared a horizontal fringe, shifting vertically across the spatial profile of the beam instead of the dimming and brightening of the beam in the wavefront matched situation. This is because, as the translation stage (Klinger -Micro Control) moves up the threads of the drive screw, the inclination of the stage with the horizontal plane keeps changing giving a vertical tilt to the wavefront of the delay arm and so it matches with the wavefront of the fixed arm only along a horizontal line resulting the horizontal fringe shift. This problem is very severe since the atoms would really interact with partially overlapping (wavefront) beams and would give rise to an extremely complicated situation and no clear coherent effect could be seen. Careful tilt adjustment of the coupling to the reduction gear was made and an iris was used to take only the central part of the gaussian spatial profile. This combination did improve the situation though not totally. The ideal solution may be to translate the delay arm on air cushion.

The synchronously pumped dye laser was mounted on a 4 feet by 12 feet optical table and another similar table holding the amplifier, pulse sequencer and associated optics was attached to it forming an L shape. This L shaped table configuration was laid on a table of fine sand for isolating the table from high frequency vibration. The tray was supported by a heavy metal frame. All this was kept floating on a set of six inflated tubes (donut shaped), one of which
(kept below the corner of the L shaped tables) could be inflated separately from the rest of the five tubes (which were interconnected) and is kept generally at a different pressure. By being able to adjust the pressure of the single tube separately, the table could be suitably tilted (to compensate any uneven distribution of load) to keep it horizontal. This is an inexpensive way of making the tables vibration free. The disadvantage of this set up is that when there is a big change in the atmospheric pressure (like before a big storm), the relative alignment of the floating table with respect to the heat pipe table would be lost.
CHAPTER IV

CONCLUSION

Coherent effects of TPR picosecond pulses through lithium vapor have been studied in this work. The anomalous transmission of "90° phase shifted" pulse pair through a TPR medium (2s → 4S transition in lithium) has been experimentally demonstrated. It was seen that the dynamic Stark shift (being proportional to the intensity) played a crucial role in the tuning of a short pulse in two photon resonant interaction. Over a scan of about typically 0.2 nm in the average wavelength of the pulse, the resonance (Stark tuned) could be swept from the peak field (maximum Stark shift) to zero field (zero Stark shift). Thus over only a little portion (temporal) of the pulse can the resonance condition be fulfilled, where the intensity of the field generates just enough Stark shift to compensate the detuning from zero field resonance. As one examines the coherence of higher order processes for which ultrashort pulses are suitable, the dynamic Stark shift plays a very important role in the tuning characteristics. The effect of dynamic Stark shift on four photon resonant interaction in Hg vapor has been theoretically studied in Ref 14.

The work of this dissertation has been to prove that the coherence of multiphoton excitation can be studied by
appropriately phased and time delayed sequence of pulses. An application of this fundamental study of coherence has been made for the enhancement of third harmonic generation. The coherent recovery of the energy lost to the two photon absorption process enabled the total interaction to be longer (than an incoherent interaction or coherent, but not 90° phase shifted pulse pair) and by proper phase matching gave an enhancement of third harmonic generation.
Figure 1: The interacting levels of lithium. The ground state $|2S\rangle$, the two photon excited state $|4S\rangle$ and the continuum resonance are shown. The set of off resonant states are represented by $\{\ell\}$. 
LITHIUM

FIG 1
Figure 2: The two photon vector model. (a) With zero detuning. (b) With finite detuning.
Figure 3: The term diagram of Lithium.
Figure 4: Theoretical result showing the envelope of the interferometric autocorrelation of the transmitted pulse (shown as second harmonic) and the corresponding third harmonic signals for various detunings.
Figure 5: The Schematic of the experiment. I.F. stands for the interference filter transmitting the third harmonic beam. The inset shows the relevant energy levels.
Figure 6: The interferometric autocorrelation of the amplified dye pulse.
--- Experimental Recording

Theory:

Gaussian Pulse, FWHM 6.2 ps:

\[ E = e^{-\left(\frac{t}{5.27}\right)^2} e^{i\phi} \]

- \[ \phi = 1.43 \, E^2 \]
- \[ \phi = 1.5\left(\frac{t}{5.27}\right)^2 \]
Figure 7: The Picosecond Amplifier chain giving mJ energy pulses.
Figure 8: The spatial profile of the amplified dye beam.
Figure 9: The Pulse sequencer.
Figure 10: Data Acquisition System.
Figure 11: The sketch of the heat pipe.
Figure 12: Vapor pressure curves of magnesium and lithium.
FIG 12

TEMPERATURE (°K)

PRESSURE (TORR)

Mg

Li

P

T

800 1000 1200 1400
Figure 13: The temperature and partial pressure distribution along the inside of the inner heat pipe.
Figure 14: The interferometric autocorrelation of the incident pulse and transmitted signal tuned at the wing of the trace where the pulses are just separated. Note the anomalous transmission (hump) signifying coherent recovery due to propagation of "90° phase shifted pair" of pulses.
TRANSMITTED SIGNAL VERSUS DELAY

- Incident
- Transmitted

SECOND HARMONIC SIGNAL

DELAY (psec)
Figure 15: The phase dependence of the incident and transmitted signals of figure 14 at a delay of three picoseconds. Note that the observed periodicity of the incident signal is $2\pi$ while that of the transmitted is, $\pi$ as expected from the propagation of "90° phase shifted pair" of pulses.
TRANSMITTED SIGNAL VERSUS PHASE

DELAY = 3 psec

--- Incident

Transmitted

SECOND HARMONIC SIGNAL

PHASE

FIG 15
Figure 16: Second order interferometric autocorrelation of incident (triangles) pulse and transmitted (circles) signal. The trace of boxes with dotted lines represent the maxima and minima of the fringes of third harmonic signal.
TRANSMITTED SIGNAL AND THIRD HARMONIC VERSUS DELAY

FIG 16
Figure 17: The incident, transmitted, and the third harmonic signals versus phase corresponding to a delay of 2 psec in the recording of figure 16. Note the phase shift of $-\pi/6$ in the third harmonic signal from that of the transmitted one.
TRANSMITTED SIGNAL AND THIRD HARMONIC VERSUS PHASE

DELAY = 2 psec

- Incident
- Transmitted
- 3rd Harmonic

PHASE

FIG 17
Figure 18: The envelope (maxima and minima of the fringes) of the third harmonic signal versus delay, in a phase matched mixture of Lithium and Magnesium (10 Torr of Lithium & 15 Torr of Magnesium). Note the enhancement at 1.8 psec.
PHASE MATCHED
THIRD HARMONIC VERSUS DELAY

Third Harmonic without Coherent Enhancement: $W_{30}/32$
Figure 19: The incident & transmitted pulse autocorrelation signals corresponding to the third harmonic enhancement data of figure 18.
TRANSMISSION VERSUS DELAY

SECOND HARMONIC SIGNAL

DELAY (psec)

FIG 19
APPENDIX A

TEST

DIMENSION HINH(250), BTH(250), BTM2(250), IN(55), CHIN(55)

DIMENSION HINH1(250), BTH1(250), BTM21(250), IN(55), CHIN(55)

DIMENSION HINH2(250), BTH2(250), BTM22(250), IN(55), CHIN(55)

DIMENSION HINH3(250), BTH3(250), BTM23(250), IN(55), CHIN(55)

DIMENSION FAS1(55), FAS2(55), FAS3(55), FAS4(55)

DATA LEECH, GREN, REU, NRE, YH, REU

READ 1090, FILE(6)

END
\[ x = \frac{\text{FLOAT}(IB) / \text{FLOAT}(NB)}{10.43}. \]

**$**

**INITIALIZE $**

\[ BS = NB*\text{ICOUNT} \]
\[ BS1 = NB + 1 \]

\[ j = 1: NB \]
\[ j = j + 1 \]
\[ j = j + 1 \]

\[ l = 1: NB \]
\[ l = l + 1 \]
\[ l = l + 1 \]

\[ j = 1: NB \]
\[ j = j + 1 \]
\[ j = j + 1 \]

\[ \text{IF}(\text{ICOUNT} > 2) \text{GO TO 12} \]
\[ \text{ICOUNT} = 0 \]

\[ \text{CALL SSDATA} \]
\[ \text{TYPE} = \text{IDATA}(4) \]
\[ A3 = \text{IDATA}(2) \]
\[ B3 = \text{IDATA}(5) \]
\[ C3 = \text{IDATA}(3) \]
\[ T3 = \text{IDATA}(6) \]
\[ R3P3 = A3 / B3 \]
\[ R3P3 = C3 / B3 \]
\[ R3P3 = T3 / B3 \]

\[ \text{CALL SSDATA} \]
\[ \text{TYPE} = \text{IDATA}(4) \]
\[ DP = \text{TYPE} - \text{TYPE} \]
\[ A2 = \text{IDATA}(2) \]
\[ B2 = \text{IDATA}(5) \]
\[ C2 = \text{IDATA}(3) \]
\[ T2 = \text{IDATA}(6) \]
4001 IMAX=IMAX+1
  IYMAX=IYP2
  TIGN=1
  C0N=0.
  ZF=0.
  OFS=0.
  IYPX=0.
  IF(IMAX.EQ.2) GO TO 20
4000 IYP=ID
  IYP3=IYP2
  IYP2=IYP1
  R25F3=R25P2
  R35F3=R35P2
  R35P2=R35P1
  R35P1=R35P2
  R65P2=R65P1
  R3=82
  R2=31
  GO TO 4003

CALL TSTBK0(IKEY)
IF(IKEY.NE.0) GO TO 103
  IYP1=IDATA(4)
  B1=IDATA(2)
  C1=IDATA(5)
  T1=IDATA(6)
  R25P1=A1/B1
  R35P1=C1/B1
  R65P1=T1/B1
  ID=-(IYP1-IYP2)*SIGN
  ID1=-(IYP2-IYP3)*SIGN
  ID2=-(IYP3-IYP4)*SIGN
  IF(DFAS.LT..25) GO TO 4050
4050 TYPF=2*IYPF
SUM=IYMAX*IYMIN
AMF=IYMAX-IYMIN
ARG=(IYPF-SUM)/AMP
IF (ARG.GT.1.) ARG=1.
IF (ARG.LT.-1.) ARG=-1.
DFAS=CONSIGN*(ACOS(ARG))/6.28318
X=DFAS+CON+IMAX=2
IF (IDMAX.GE.2) GO TO 445
IF (X.LT.XP) GO TO 80
XP=X
IF (X.GT.NP) GO TO 60
IF (B4.GT.WU3) GO TO 801
IF (B4.LT.WL1) GO TO 301
51 NBIN=(RATID*X-FLOAT(N))*FLOAT(N)+1;
IF (NBIN.LT.NB1) GO TO 52
NB1=NB1+1
GO TO 51
52 IF (B4.LE.WU1) GO TO 301
IF (B4.LT.WL2) GO TO 302
IF (W.LT.WL3) GO TO 30
N1IN3(NB1)=N1IN3(NBIN)+1
BINW3(NBIN)=BINW3(NBIN)+R35P4
BINW3(NBIN)=BINW3(NBIN)+R35P4
GO TO 50
502 IF (W.LT.WL2) GO TO 50
N1IN2(NB1)=N1IN2(NBIN)+1
BINW2(NBIN)=BINW2(NBIN)+R35P4
BINW2(NBIN)=BINW2(NBIN)+R35P4
GO TO 50
501 N1IN1(NB1)=N1IN1(NBIN)+1
BINW1(NBIN)=BINW1(NBIN)+R35P4
BINW1(NBIN)=BINW1(NBIN)+R35P4
GO TO 50
301 NB1=NB1+1
30 IF (A1.EQ.1023.0) IC2=IC2+1
IF (C1.EQ.1023.0) IC3=IC3+1
GO TO 20
C ***** END LOOP *****
C ***** PRINT OUT *****
60 IC02=ICOUNT.
ICOUNT=0
NIS(IG)=M1
N2=NB/N0
FASI(I)=FAS/FLOAT(NINT)
BINW1(I)=INT/FLOAT(NINT)
BIN3W1(I)=BIN2T/FLOAT(NINT)
BIN3W1(I)=BIN3T/FLOAT(NINT)

603 IN1(I)=NINT
IF(IN(I).LE.2) GO TO 10
III=I-1
INP=IN1(III)
IF(INP.NE.0) GO TO 10
IA=I-2
BINW1(III)=(BINW1(IA)+BINW1(I))/2,
BIN3W1(III)=(BIN3W1(IA)+BIN3W1(I))/2,
BIN3W1(III)=(BIN3W1(IA)+BIN3W1(I))/2.
BINT=0.
BIN2T=0.
BIN3T=0.

10 CONTINUE
DO 500 I=1,MB
BINT=0.
BIN2T=0.
BIN3T=0.
FA8=0.
INP=I
IF(INP.NE.0) GO TO 500
I=INP
LB=1-100
L1+=MINT+100
FA8=FA8+FLOAT(L1)
MINT=MINT+BIN2W2(L)
BINT=BINT+BIN3W2(L)

501 MINT=MINT+NINBI2(L)
LB=(I-1)*NINT+NINT/2
IF(MINT.LE.0) GO TO 111
IF(INP.NE.0) GO TO 604
C FAS2(I)=FAS/FLOAT(NINT)
FAS2(I)=FLOAT(I)
IF(FAS2(I).GT.0.0) GO TO 111
PRINT 555,FAS2(I)
555 FORMAT(F6.2)

111 BINW2(I)=INT/FLOAT(NINT)
BIN2W2(I)=BIN2T/FLOAT(NINT)
BIN3W2(I)=BIN3T/FLOAT(NINT)

604 IN2(I)=NINT
IF(I.LE.2) GO TO 500
III=I-1
INP=IN2(III)
IF(INP.NE.0) GO TO 500
IA=I-2
BINW2(III)=(BINW2(I)+BINW2(IA))/2.
BIN2W2(III)=(BIN2W2(I)+BIN2W2(IA))/2.
BIN3W2(III)=(BIN3W2(IA)+BIN3W2(I))/2.
114

1A S 3(1) = 1

1B N 2 W 3 (:i: ) = B I: N 2 / F L O A T (N I N T )

1B I N 3 (I ) = H I N T

IF (I . LE . 2) GO TO 502
I I T = -1
INP = B I N 3 (I I)
IF (I N P . NE . 0) GO TO 502
I A = 1
2

B I N W 3 (I ) = ( B I N W 3 (I ) + B I N W 3 (I A ) ) / 2.
B I N 2 W 3 (I I ) = ( B I N 2 W 3 (I ) + B I N 2 W 3 (I A ) ) / 2.
B I N 3 W 3 (I I ) = ( B I N 3 W 3 (I A ) + B I N 3 W 3 (I ) ) / 2.

502 CONTINUE
B I N 1 = 0 0 0 0 0 0
B I N 3 = 0
I D I = 1, N B
B I N = B I N 2 W 3 (I )
I F (R E A L (B I N ) . GT . B M A X ) B M A X = B I N
I F (R E A L (B I N ) . LT . B M I N ) B M I N = B I N
B M A X = 1 0 . 0 * B M A X / Y P L M A X + 3 ,
B M I N = 1 0 . 0 * B M I N / Y P L M A X + 3 ,
C A L L S Y M B O L ( X P L , B M A X - 2 )
C A L L S Y M B O L ( X P L , B M I N + 1 )
B I N = 0 . 0
S Y M = 0 . 0
R T P = 0 . 0
S Y M 2 = 0 . 0
S Y M 3 = 0 . 0
W R I T E ( 1 , 8 1 ) ( R C 1 , R C 2 , R C 3 , R I C P , R I C P 2 , ( B I N W 1 (I ) , I = 1 , N B )
W R I T E ( 1 , 8 1 ) ( B I N W 2 (I ) , I = 1 , N B ) , ( B I N W 2 (I ) , I = 1 , N B )
W R I T E ( 1 , 8 1 ) ( B I N W 3 (I ) , I = 1 , N B ) , ( B I N W 3 (I ) , I = 1 , N B )
W R I T E ( 1 , 8 1 ) ( F A S 1 (I ) , I = 1 , N B ) , ( F A S 1 (I ) , I = 1 , N B )
W R I T E ( 1 , 8 1 ) ( F A S 3 (I ) , I = 1 , N B ) , ( B I N 3 W 3 (I ) , I = 1 , N B )
W R I T E ( 1 , 8 1 ) ( B I N 3 W 3 (I ) , I = 1 , N B )
W R I T E ( 1 , 8 1 ) ( I N 1 (I ) , I = 1 , N B ) , ( I N 2 (I ) , I = 1 , N B )
101 C O N T I N U E
S L 1 F O R M A T ( I 6 )
S L 1 F O R M A T ( E 1 5 , 6 )
10 3 C A L L C L O S E F ( 1 )
C A L L O P E N F ( I , I F I L E , 1 )
C A L L A L P H A
R E A D 5 1 3 1
5 1 3 1 F O R M A T ( )
10 0 P R I N T 3 0 5
3 0 5 F O R M A T ( ' W I N D O W : H I G H 3 , M I D D L E 2 , L O W 1 ' )
R E A D 2 1 , 1 W
DO 306 I=1,NB
306 READ(1,81) BINW2(I)
DO 307 I=1,NB
307 READ(1,81) BINW2(I)
DO 308 I=1,NB
308 READ(1,81) BINW3(I)
DO 309 I=1, NB
309 READ(1,81) BINW3(I)
DO 305 I=1, NB
305 READ(1,81) FAS1(I)
DO 306 I=1, NB
306 READ(1,81) FAS2(I)
DO 307 I=1, NB
307 READ(1,81) FAS3(I)
DO 310 I=1, NB
310 READ(1,81) BINW1(I)
DO 311 I=1, NB
311 READ(1,81) BINW2(I)
DO 312 I=1, NB
312 READ(1,81) BINW3(I)
DO 314 I=1, NB
314 READ(1,81) FAS1(I)
DO 315 I=1, NB
315 READ(1,81) FAS2(I)
DO 316 I=1, NB
316 READ(1,81) FAS3(I)
CONTINUE
IF(IW.EQ.1) GO TO 310
IF(IW.EQ.2) GO TO 311
IF(IHP.EQ.0) GO TO 72
IF(IHP.EQ.2) GO TO 813
DO 73 I=1, NB
CHAN(I)=FAS3(I)
73 Y(I)=BINW3(I)
GO TO 74
813 DO 814 I=1, NB
CHAN(I)=FAS3(I)
814 Y(I)=BINW3(I)
GO TO 74
72 DO 75 I=1, NB
CHAN(I)=FAS3(I)
75 Y(I)=BINW3(I)
GO TO 74
311 IF(IHP.EQ.0) GO TO 312
IF(IHP.EQ.2) GO TO 815
DO 314 I=1, NB
CHAN(I)=FAS2(I)
314 Y(I)=BINW2(I)
GO TO 74
314 CC 318 I=1,NB
   CHAN(I)=FAS1(I)
318 Y(I)=INIW1(I)
74  CALL ERASE
   CALL SCALE(CHAN+5,NB)
   CALL SCALE(Y+5,NB)
   CALL YAXIS(3.3,CHAN+NB,IXTIT,9)
   CALL YAXIS(3.3,Y+NB,ITYTIT,9)
   CALL PLOT(3.3,CHAN,Y+NB,3)
   CALL PEN(0,2,0)
   CALL ALPHA
   CALL SCALE(591,RC1,RC2,RC3)
   CALL SCALE('WISH TO SEE NIN?INST1 FOR YES&2 FOR NO')
617 FORMAT()  CALL ERASE
   PRINT 519
   CALL SCALE('WISH TO SEE NIN?INST1 FOR YES&2 FOR NO')
   FORMT()  CALL ERASE
   PRINT 519
   517 FORMAT('WISH TO SEE NIN?INST1 FOR YES&2 FOR NO')
617 FORMAT()  CALL ERASE
   PRINT 519
   CALL SCALE('WISH TO SEE NIN?INST1 FOR YES&2 FOR NO')
   FORMT()  CALL ERASE
   PRINT 519
   GOTO 100
   PRINT 520
520 FORMAT('NBIN1',20X,'NINBI1')
521 CONTINUE
   READ 517
   CALL ERASE
   PRINT 520
522 FORMAT('NBIN2',20X,'NINBI2')
523 CONTINUE
   READ 517
   CALL ERASE
   PRINT 520
524 FORMAT('NBIN3',20X,'NINBI3')
523 CONTINUE
   READ 517
   CALL ERASE
   PRINT 519
   PRINT 530
530 FORMAT('WISH TO SEE NIN?INST1 FOR YES&2 FOR NO')
530 FORMAT('WISH TO SEE NIN?INST1 FOR YES&2 FOR NO')
   READ 517
   CALL ERASE
   PRINT 530
531 FORMAT('WISH TO SEE NIN?INST1 FOR YES&2 FOR NO')
532 CONTINUE
   READ 517
   CALL ERASE
   PRINT 530
533 FORMAT('WISH TO SEE NIN?INST1 FOR YES&2 FOR NO')
533 CONTINUE
   READ 517
   CALL ERASE
   END
APPENDIX B

MAIN

COMMON/JC/DT,SDW,EMAX,H1,H2,H5,H6,H7,FR,STK,W0
1,T2INV,TL11N,ABSU,ALFAL,ALFAU,GAM2,DOMO,IFAS,M,IMAX
COMMON/ARTS/TR(1024),TI(1024),TL(1024),TU(1024),TN(1024)
1,ERR(1024),ERI(1024),AI(40)
COMMON/OTHER/DINT,FINT,WIDTH,PHASE,SCA,FI,DO,DOM1,EN3M,AIM,
1AMP,CENTER,WID2,DLT,TINT,AUMA1,AUMA2,FAV1,FAUM,PEAK,IFAE3,IFAA3.
1DLY,IFREQ,MELY,DOM,EN3M,IFAE3,IFAA3,START,DLT1
COMMON/ARYS/ENG1(40),ENG2(40),ER3(1024),END(1024),FSG(40),EN3(40),
1AUTMAX(100),AUTMIN(100),AVE(100),BUTMAX(100),BUTMIN(100),AVE1(100)
2,AVDUM(100),AAIM(100)

CALL DEFINE
DO 1 IFREQ = 1,MOM
SCA = 0.
PEAK = 0.
DLY = DLT1
DO 2 IDLY = 1,MDLY
FAV = 0.
FAUM = 0.
FAUS = 1000000.
FAV WILL BE FRINGE AVERAGED SECOND HARMONIC
FAUM WILL BE AUTOCORRELATION MAXIMA
FAUS WILL BE AUTOCORRELATION MINIMA.
EN3M = 0.
AIM = 0.
EN3M = MAX OF 3RD HARMONIC ENERGY
AIM = MAXIMUM IONIZATION.
IFAE3 = PHASE CORRESPONDING TO EN3M
IFAA3 = PHASE CORRESPONDING TO AIM
AUMA1 = 0.
AUMA2 = 10000000.
FAV1 = 0.
AUMA1 = AUTOCORRELATION MAX BHP
AUMA2 = AUTOCORRELATION MIN BHP
FAV1 = FRINGE AVERAGED 2ND HARMONIC BHP.

PHASE = START
DO 3 IFAS = 1,NFAS
CALL pulse
CALL BLOCH
CALL CRLA
CALL AVERAG(FSG(IFAS),FAUM,MIFAS,FAUS,FAV,NFAS,IFAS)
CALL AVERAG(EN3(IFAS),EN3M,IFAE3,DUMY,AVDUM,NFAS,IFAS)
CALL AVERAG(AI(IFAS),AIM,IFAA3,DUMY,AVDUM,NFAS,IFAS)
CALL AVERAG(ENG2(IFAS), AUMA1, IIII, AUMA2, FAV1, NFAS, IFAS)

PHASE = PHASE + FINT
AUTMAX(IDLY) = FAUM
AUTMIN(IDLY) = FAMU
AVE(IDLY) = FAV
AEN3(IDLY) = EN3M
AAIM(IDLY) = AIM

AEN3, AAIM STORES 3RD HARMONIC ENERGY
AND IONIZATION AS A FUNCTION OF DELAY.

BUTMAX(IDLY) = AUMA1
BUTMIN(IDLY) = AUMA2
AVE1(IDLY) = FAV1

KIF = IDLY/4
IF(ABS(FLOAT(IDLY)/4.-FLOAT(KIF)).LT.0.1) CALL OUTPUT
IF(IDLY.EQ.4) CALL PLOT(EN3, EN3, EN3M, EN3M, 1, 1, NFAS, 1, FINT)
IF(IDLY.EQ.12) CALL PLOT(EN3, EN3, EN3M, EN3M, 1, 1, NFAS, 1, FINT)

CALL OUTPUT
CALL AVERAG(BUTMAX(IDLY), SCA, IJII, SWAT, WATS0, MDLY, IDLY)
CALL AVERAG(AUTMAX(IDLY), FEAK, IJII, SOWE, NAWE, MDLY, IDLY)

IDLY = IDLY + DINT
CALL ENVLOP

L0CM0 = L0CM0 + TINT
STOP
END
SUBROUTINE PLOT(AA, BB, SCA, SCB, ISEP, IAS, MEXT, IT, DX)

C THIS SUBROUTINE PLOTS TWO ARRAYS AA AND BB, OF DIMENSION MEXT
C EVERY IT (STEP SIZE DX). SCA AND SCB ARE THE MAXIMUM VALUES
C OF AA AND BB RESPECTIVELY. ISEP IS THE SEPARATION LINE
C BETWEEN BOTH ARRAYS. IF ISEP=1, BOTH ARRAYS ARE PLOTTED
C ON THE SAME GRAPH. IAS DESIGNATE THE ORDINATE OF A LINE
C OF DOTS (TO BE USED EITHER AS AN ASYMPTOTE, OR AN AXIS).

C PLOTS 2 ARRAYS (FOR INSTANCE AMPLITUDE AND PHASE)
C WITH A SCALE FACTOR SCA AND SCB
C DX IS THE STEP SIZE OF THE PARAMETER (TIME OR FREQUENCY).
C THE NUMBER OF POINTS PLOTTED IS (MEXT/IT).

DIMENSION AA(1024), BB(1024), PINE(120)
DATA BLANK/' '/, DOT/'./, CROS/'+'/, STAR/'*'/
WRITE(6,205) SCA, SCB
205 FORMAT(' SCA= ',E15.7, ' SCB= ',E15.7,/
SCA = FLOAT(ISEP-1)/SCA
IF (ABS(SCB).GT.1E-8) SCB = FLOAT(81-ISEP)/SCB
IF (ISEP.EQ.1) SCA=SCB
DO 200 J=1,120
200 FINE(J) = DOT
WRITE(6, 201) PINE
201 FORMAT(2H T,9X,120A1)
DO 202 J=1,120
202 PINE(J) = BLANK
FINE(IAS) = DOT
FINE(1) = DOT
FINE(IAS) = DOT
DO 203 I=IT,MEXT,IT
T = (I)*DX
IL = INT(SCA*AA(I)) + 1
JL = INT(SCB*BB(I)) + ISEP
FINE(IL) = CROS
FINE(JL) = STAR
WRITE (6, 204) T, FINE
204 FORMAT(F9.5,2X,120A1)
FINE(IL) = BLANK
FINE(JL) = BLANK
FINE(IAS) = DOT
FINE(IAS) = DOT
RETURN
**SUBROUTINE PULSE**

COMMON/JC/DT, SDW, EMAX, H1, H2, H3, H4, H5, H6, H7, FR, STK, W0
1, T2INV, T1INV, ABSU, ALFAL, ALFAU, GAM2, DOMO, IFAS, M, IMAX
COMMON/ARYS/TR(1024), TI(1024), TL(1024), TU(1024), T0N(1024)
1, ERR(1024), ERI(1024), A(40)
COMMON/OTHER/DINT, FINT, WIDTH, PHASE, SCA, PI, DL, DOM1, LAMP, CENTER, WD2, DLY, TINT, AUMA1, AUMA2, FAV1, FAVUM, PEAK, 1I1LY, IFREQ, MDLY, NOM, IFAS, IFAAM, START, DLY1, COEF
COMMON/ARAY/EMG1(40), ENG2(40), ER3(1024), E913(1024), FSG(40), EN3(40), 1AUTMAX(100), AUTMIN(100), AVE(100), BUTMAX(100), BUTMIN(100), AVE(100), 2, AEN3(100), AAIM(100)

ESG2 = 0.
ESG1 = 0.
AMP1 = AMP*COS(PHASE);
AMP2 = AMP*SIN(PHASE);
DO 1 I=1,M
   EF1 = EXP(-((I*DT-CENTER)**2)/WD2)
   EF2 = EXP(-((I*DT-CENTER-DLY)**2)/WD2)
   ERR(I) = AMP*EF1 + AMP1*EF2
   ER1(I) = AMP2*EF2
   ERR2 = ERR(I)**2
   ERI2 = ERI(I)**2
ESG1 = ERR2 + ERI2 + ESG1
ESG2 = ESG2 + (ERR2-ERI2)**2 + 4.*ERR2*ERI2
ESG1 = TOTAL ENERGY OF SUPERPOSITION PULSE GOING TO THE
HEAT PIPE;
ESG2 IS THE TOTAL ENERGY IN SECOND HARMONIC OF THE SUPERPOSITION
PULSE.
ENG1(IFAS) = ESG1*DT
ENG2(IFAS) = ESG2*DT/((AMP**4)*WIDTH);
ENG1(IFAS) IS THE TOTAL ENERGY AS A FUNCTION OF PHASE;
ENG2 IS THE TOTAL SECOND HARMONIC ENERGY
AS A FUNCTION OF PHASE.
RETURN
END
* * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * *

SUBROUTINE ENVLOF

* * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * * *

COMMON/JC/DT,SDW,EMAX,H1,H2,H3,H4,H5,H6,H7,FR,STK,W0
COMMON/ARYS/TR(1024),TI(1024),TL(1024),TU(1024),TW(1024)
COMMON/OTHER/DINT,FINT,WIDTH,PHASE,SCAL,PI,EL,DOM1,
LAMF,CENTER,WD2,DLY,TINT,AUMA1,AUMA2,FV1,FAUM,PEAK,

NORMALIZATION OF THE AUTOCORRELATIONS:
WE CHOOSE TO SET THEM =1 AT THE MAXIMUM DELAY.
FOR THE INTERFEROMETRIC AUTOCORRELATION AFTER THE HEAT PIPE:
WE SET THE AVERAGE OF THE LOWER AND UPPER ENVELOPE = 1
THIS IS EQUIVALENT TO CHOOSING THE SCALE OF THE INTENSITY
AUTOCORRELATION TO BE SUCH THAT

\[
\text{AVE}(\text{MDLY}) = 1.
\]

DO 1 I=1,MDLY

\[
\text{BUTMAX}(I) = \text{BUTMAX}(I) / \text{AVE}(\text{MDLY})
\]

\[
\text{BUTMIN}(I) = \text{BUTMIN}(I) / \text{AVE}(\text{MDLY})
\]

\[
\text{CALL AVERAGE}(\text{AVE}(I),\text{XMK},\text{IMK},\text{XMIN},\text{PAV1},\text{MDLY},I)
\]

\[
\text{CALL AVERAGE}(\text{AVE}(I),\text{ZMA2},\text{IMZ},\text{ZMIN},\text{FAV2},\text{MDLY},I)
\]

\[
\text{CALL AVERAGE}(\text{AEN3}(I),\text{SMX3},\text{IPK3},\text{ZMIN},\text{FAV3},\text{MDLY},I)
\]

\[
\text{AVE}(I) = \text{AVE}(I) / \text{AVE}(\text{MDLY})
\]

\[
\text{AUTMAX}(I) = \text{AUTMAX}(I) / \text{AVE}(\text{MDLY})
\]

\[
\text{AUTMIN}(I) = \text{AUTMIN}(I) / \text{AVE}(\text{MDLY})
\]

\[
\text{WRITE}(6,201) \text{DOMO}
\]

FORMAT(27H1 DETUNING (AT ZERO FIELD): ,F8.2,///)

WRITE(6,203)

FORMAT(6H DELAY,5X,12H AUTOMAX BHP,5X,12H AUTOMIN BHP,
15X,12H AUTOMAX AHP,5X,12H AUTOMIN AHP,///)

\[
\text{DLY} = 0.0
\]

DO 204 I=1,MDLY

\[
\text{DLY} = \text{DLY} + \text{DINT}
\]

\[
\text{WRITE}(6,205) \text{DLY},\text{BUTMAX}(I),\text{BUTMIN}(I),\text{AUTMAX}(I),\text{AUTMIN}(I)
\]

FORMAT(F9.5,2X,F12.6,5X,F12.6,5X,F12.6,5X,F12.6)

\[
\text{WRITE}(6,206)
\]

FORMAT(6H DELAY,5X,12H AUTOAVE BHP,5X,12H AUTOAVE AHP,,
15X, 12H 3RD HAR MAX, 5X, 15H IONISATION MAX.
DLY = 0.0
DO 207 I = 1, MDLY
WRITE(6, 218) DLY, AVE1(I), AVE(I), AEN3(I), AAIM(I)
218 FORMAT(F8.5, 2X, F12.6, 5X, F12.5, 5X, F12.6, 5X)
207 DLY = DLY + DINT
WRITE(6, 208)
208 FORMAT(1H1, 10X, ' AUTOCORRELATION PLOT BHP', //)
WRITE(6, 2) SCA
2 FORMAT( ' PEAK VALUE', E15.7)
CALL PLOT(BUTMAX, BUTMIN, 10.0, 10.0, 1, MDLY, 1, DINT)
WRITE(6, 209)
209 FORMAT(10X, ' AUTOCORRELATION PLOT AHP', //)
WRITE(6, 2) PEAK
CALL PLOT(AUTMAX, AUTMIN, 10.0, 10.0, 1, MDLY, 1, DINT)
WRITE(6, 3) PAV1, PAV2
3 FORMAT(' PEAKS OF THE INTENSITY AUTOCORRELATIONS: ', 19X
1 ' BEFORE TRANSMISSION ', F9.5, ',', 20X, ' AFTER TRANSMISSION
2 F9.5)
CALL PLOT(AVE1, AVE, 4.0, 4.0, 1, MDLY, 1, DINT)
RETURN
END
SUBROUTINE AVERAG(Y,YMAX,MAX,YMIN,YAV,NDIM,I)

THIS SUBROUTINE EXITS IN YMAX THE MAXIMUM OF THE FUNCTION Y.
YMIN MINIMUM Y.
YAV AVERAGE Y.
MAX INDEX CORRESPONDING TO YMAX.

THIS SUBROUTINE HAS TO BE INSERTED IN A LOOP THAT SCANS THROUGH THE FUNCTION Y; NDIM IS THE NUMBER OF POINTS (DIMENSION OF THE ARRAY THAT MAY CORRESPOND TO Y).
I IS THE INDEX OF THE INDEX VARIABLE IN Y.
PRIOR TO THE LOOP, THE FOLLOWING INITIALIZATIONS HAVE TO BE PERFORMED:
YAV TO ZERO
YMIN TO A NUMBER LARGER THAN THE TRUE YMIN
YMAX TO A NUMBER SMALLER THAN THE TRUE YMAX.

LOGICAL L

L = (Y.GT.YMAX)
IF (L) YMAX = Y
IF (L) MAX=I
IF (Y.LT.YMIN) YMIN = Y
   YAV = YAV + Y/FLOAT(NDIM)
RETURN
END
SUBROUTINE OUTPUT

COMMON/JC/DT,SDW,EMAX,H1,H2,H3,H4,H5,H6,H7,FR,STK,W0
1.T2INV,T1INV,ABSU,ALFAL,ALFAU,GAM2,DOM0,IFAS,M,IMAX
COMMON/ARYS/TR(1024),TI(1024),TL(1024),TU(1024),TW(1024)
1,ERR(1024),ERR(1024),AI(1024)
COMMON/OTHER/DINT,FINT,WIDTH,PHASE,SCA,PI,DE,DOM1,EN3M,AIM,
LAMP,CENTER,WID2,DLY,TINT,AUML1,AUML2,FAV1,FAUM,FEAK,IFAS3,IFAAN,
2IDLY,IFREQ,HOLD,DOM,NFAS,IFAS3,IFAMM,START,DL1,C0EF
COMMON/ARRAY/ENG1(40),ENG2(40),ER3(1024),EI3(1024),FSG(40),EHI3(40),
1AUTMAX(100),AUTMIN(100),AVE(100),BUTMAX(100),BUTMIN(100),AVE(100)
2,AEN3(100),AAM(100)
WRITE(6,101) DOMO,DLY
FORMAT(17H DETUNING (AT ZERO FIELD): ,F3.2,
13H DELAY = ,F8.5)
WRITE(6,102) FAV1,AUML1,AUML2
FORMAT(17H FRINGE AV BHP = ,E8.2,
129H AUTMAX BHP AT THIS DELAY = ,E8.2,
229H AUTMIN BHP AT THIS DELAY = ,E8.2)
WRITE(6,104) EN3M,IFAS3,AIM,IFAM
FORMAT( "MAXIMUM THIRD HARMONIC AT THIS DELAY : ",
1F10.6,\' CORRESPONDING PHASE: \',18,\'/
2\' MAXIMUM NUMBER OF IONS AT THIS DELAY: \',F10.6,\'/
3\' CORRESPONDING PHASE: \',18,\'/
WRITE(1,105)
FORMAT(6H PHASE, 6X,13H 2ND HARM BHP,2X,
113H 2ND HARM AMP,2X,15H 3RD HARM ENERGY,2X,
213H INPUT ENERGY,2X,11H IONIZATION,/)
SUBROUTINE CRLA

**COMMON**/JC/DT, ED, EMAX, H1, H2, H3, H4, H5, H6, H7, FR, STK, W0
1, T2INV, T1INV, ABSU, ALFAL, ALFAU, G, DMO, IFAS, M, IMAX

**COMMON**/ARYS/ TR(1024), TI(1024), TL(1024), TU(1024), TW(1024)
1, ERR(1024), ERI(1024), AI(40)

**COMMON**/OTHER/DINT, FINT, WIDTH, PHASE, SCA, PI, DZ, DOM1,
LAMP, CENTER, ND2, DLL, TINT, AUMA1, AUMA2, FAV1, FAV2, PEAK,
2IDLY, IFREQ, MDLY, NOM, NFAS, IFAS3, IFAS4, START, DLL1, CCEF

**COMMON**/RAY/ENG1(40), ENG2(40), ER3(1024), ET3(1024), FS(40), EN3(40),
1AUTMAX(100), AUTMIN(100), AVE(100), BUTMAX(100), BUTMIN(100), AVE1(100)
2, AEN3(100), AAAI(100)

SFSE = 0.
SENS = 0.

UNIT DISTANCE 13.5 CM (AT 1 TORR), DZ = (0.2)*13.5 CM.

DO 11 I = 1, M

ALPHA = ALFAL*(TL(I)-1.)+ALFAU*TU(I)

ALPHA=0.

ABSOR = ABSU*TU(I)

DRE1 = ERR(I)*(TR(I)+ABSOR) + ERI(I)*(TI(I)-ALPHA)

DIE1 = ERI(I)*(TI(I)-ABSOR) + ERI(I)*(TI(I)+ALPHA)

DER3 = -COEF*(TI(I)*ERI(I) - TR(I)*ERR(I))

DEI3 = COEF*(TI(I)*ERR(I) + TR(I)*ERI(I))

ENB = ERR(I)**2 + ERI(I)**2

WRITE(6,454) ENB

454 FORMAT(//,15X,F10.5)

ERR(I) = ERR(I) - DRE1*DS

ERI(I) = ERI(I) - DIE1*DS

EN4 = ERR(I)**2 + ERI(I)**2

WRITE(6,545) EN4

545 FORMAT(//,15X,F10.5)

WE PUT IN THE OLD ARRAYS ERR, ERI, THE NEW (PROPAGATED)
FUNDAMENTAL FIELDS AT OMEGA - LINEARIZING MAXWELL'S EQUATIONS.

ER3(I) = DER3*DS

EI3(I) = DEI3*DS

F2ER = ERR(I)**2

F2EI = ERI(I)**2

SFSE = F2ER**2 + F2EI**2 +2.*F2ER*F2EI + SFSE

SENS = SENS + ER3(I)**2 + EI3(I)**2

SFSE = SFSE/10.

FSE(IFAS) = SFSE*DT/(AMP**4)*WIDTH

EN3(IFAS) = SENS*DT

FSG = 2ND HARMONIC ENERGY AFTER HEAT PIPE;

EN3 = 3RD HARMONIC FIELD ENERGY.

RETURN

END
THE INPUT TO THIS SUBROUTINE IS A COMPLEX ELECTRIC FIELD GIVEN BY THE ARRAYS ERR(1024), ER(1024) (DIMENSION SET TO M). THE PARAMETERS ARE THE FREQUENCY MISMATCH FROM RESONANCE DOMO = OMEGA0 - 2*OMEGALIGHT, AND ALL THE MEDIUM PARAMETERS SPECIFIED IN SUBROUTINE DEFINE.

LOGICAL LI

SEW = 0.

CCL = 0.

CCU = 0.

THESE THREE VARIABLES WILL BE USED TO MEASURE THE ENERGY ABSORPTION.

IMAX = 0.

EMAX = 0.

EMAX WILL BE THE PEAK FIELD (OF MOD(ERR,ERI)), AND IMAX THE CORRESPONDING INDEX.

QR = 0.

QI = 0.

WL = WO

WU = 0.

QR3 = 0.

QI3 = 0.

WU3 = 0.

WL3 = 0.

QR1 = 0.

QI1 = 0.

WU1 = 0.

WL1 = 0.

QR0 = 0.

QI0 = 0.

WU0 = 0.

WL0 = 0.
QR2 = 0.
QI2 = 0.
WU2 = 0.
WL2 = 0.

C START OF THE MAIN TIME LOOP
DO 6 I=1,M
   IJ = 0
   C IJ = NUMBER OF PREDICTOR-CORRECTOR LOOPING.
   GR = ERR(I)
   GI = ERI(I)
   IF (ABS(GR).LT.1E-20) GR = 0.
   IF (ABS(GI).LT.1E-20) GI = 0.
   E2 = GR**2
   E4 = GI**2
   FI2 = 2.*GI*GR
   FR2 = E2 - E4
   E4 = E2 + E4
   DOM = DOMO + STK*E4
   DOM = DOM + 2.0*FI
   C DOMO IS THE MISMATCH FROM RESONANCE OM0-2OMLIGHT;
   C STK IS THE STARK SHIFT.
   T12 = T2INV + GAM2*E4
   C IS THE LOSS OF COHERENCE DUE TO T2 AND IONIZATION
   L1 = E4*GT.EMAX
   IF (L1) IMAX = I
   IF (L1) EMAX = E4
   FR = QR + H1*QR0 - H2*QR1 + H3*QR2 - H4*QR3
   XI = QI + H1*QI0 - H2*QI1 + H3*QI2 - H4*QI3
   FU = WU + H1*WU0 - H2*WU1 + H3*WU2 - H4*WU3
   PL = WL + H1*WL0 - H2*WL1 + H3*WL2 - H4*WL3
   IF (ABS(FR).LT.1E-20) FR = 0.
   IF (ABS(XI).LT.1E-20) XI = 0.
   IF (ABS(FU).LT.1E-20) FU = 0.
   IF (ABS(PL).LT.1E-20) PL = 0.
   HR = QR + H5*QR0 - H6*QR1 + H7*QR2
   HI = QI + H5*QI0 - H6*QI1 + H7*QI2
   HU = WU + H5*WU0 - H6*WU1 + H7*WU2
   HL = WL + H5*WL0 - H6*WL1 + H7*WL2
   C THE CORRECTOR LOOP
   C THE DIFFERENTIAL EQUATIONS CAN BE FOUND HERE.
   DU = -(T1INV + ABSU+E4)*FU + .5*(FR*FR2 + XI*FI2)
CU = HU + H4 * DU
DL = -.5 * ( FR2*FR + FI2*XI )
CL = HL + H4 * DL
WC = CU - CL

DI = DOM*FR - T12*XI - WC*FI2
CI = HI + H4 * DI
DR = -DOM*CI - T12*FR - WC*FR2
CR = HR + H4 * DR
DI = DOM*CR - T12*XI - WC*FI2

CASE POPULATION INVERSION

CI = HI + H4 * DI
IF (ABS(CR).LT.1E-20) CR = 0.
IF (ABS(CI).LT.1E-20) CI = 0.
IF (ABS(CU).LT.1E-20) CU = 0.
IF (I.LT.9) GO TO 8
IF (ABS(CR*CI*CU - FR*XI*PU).GT.FR) GO TO 9
GO TO 8

9 IF (IJ.GT.3) GO TO 8
   FR = CR
   XI = CI
   PU = CU
   PL = CL
   IJ = IJ + 1
   GO TO 7
8
   QR = CR
   QI = CI
   WL = CL
   WU = CU
   QR0 = DR
   QI0 = DI
   WU0 = DU
   WL0 = DL
   QR1 = QR0
   QI1 = QI0
   WU1 = WU0
   WL1 = WL0
   QR2 = QR1
   QI2 = QI1
   WU2 = WU1
   WL2 = WL1
   QR3 = QR2
   QI3 = QI2
   WU3 = WU2
WL3 = WL2
TR(I) = CR
TI(I) = CI
TL(I) = CL
TU(I) = CU
TW(I) = WC
SDW = SDW + DU - DL
CCL = CCL + DL
CCU = CCU + DU

SDW = SDW*DT
CCL = CCL*DT
CCU = CCU*DT

USE ONE OF TWO ALTERNATIVE WAYS TO CALCULATE THE IONIZATION
IN AI(IFAS) (THEY SHOULD BE EQUIVALENT).
AI(IFAS) = - (CCL + CCU)

RETURN
END
**SUBROUTINE DEFINE**

**COMMON**/JC/DT, DSW, EM, H1, H2, H3, H4, H5, H6, H7, FR, STK, W0
1, T2INV, T1INV, ABSU, ALFAL, ALFU, GAM2, DOM0, IFA, M, IMAX
COMMON/ARYS/TR(1024), TI(1024), TL(1024), TU(1024), TN(1024)
1, ERR(1024), ERI(1024), AI(10)
COMMON/OTHER/DINT, FINT, WIDTH, PHASE, SCA, FI, EI, DOM1,
1, MF, CENTER, W12, DLY, TINT, AUM1, AUM2, FAV1, FAUM, PEAK,
1, MDLY, IFREQ, MDLY, NCM, NFAS, IFAE3, IFM, START, DLY1, COEF
COMMON/ARYS/ENG1(100), ENG2(100), ERI3(1024), EI3(1024), FSC(40), ENH(40),
1, AUTMAX(100), AUTMIN(100), AVE(100), BUTMAX(100), BUTMIN(100), AVE1(100)
2, AEN3(100), AAUIM(100)

NCM = 4
NCM = 1 IS THE NUMBER OF FREQUENCY INTERVALS

FI = 3.14159265359

DLY1 = 0.0
DLY = DLY1

TINT = 300.

IS A FREQUENCY INTERVAL IN 10EXP9 SEC-1

DOM0 = 300.

WIDTH = 0.0062

WIDTH IS THE FWHM OF THE PULSE IN NS.

MDLY = 12

DLY = 2.0/WIDTH/MDLY

IS THE DELAY INTERVAL IN NS.

THERE WILL BE MDLY-1 DELAY INTERVALS.

NFAS = 20

THERE WILL BE 20 PHASE POINTS.

FINT = 2.0/PF/(NFAS-1)

FINT IS THE PHASE INCREMENT

VALUE OF THE DETUNING EXCLUDING STARK SHIFT.

ALL TIMES ARE IN NS. AND ALL FREQUENCIES IN NS E-1

(IN NS E-1 AND NOT IN GHZ AS IN THE PHYSICAL REVIEW PAPER).

T2 = 5.

PHASE RELAXATION TIME IN NS.

T1 = 50.

ENERGY RELAXATION TIME IN NS.

T2INV = 1./T2
T1INV = 1./T1

T2INV = 5.
H3 = H7 * 37.
H4 = H7 * 3.
H5 = H7 * 19.
H6 = H7 * 5.
DLMAX = MDLY*DINT
TIMAX = M*DT

WRITE(6,1) MDLY,DINT,DLMAX,M,DT,TIMAX,NFAS,FINT

FORMAT:  *****************, // RANGES, *****************, //, 5H DELAYS,
18H,18H from 0 TO,13,2H *,F7.5,2H = ,F6.3,/,5H TIME,8X,
210H FROM o TO,14,2H *,F8.6,2H = ,F6.3,/,6H PHASE,9X.
310H FROM 0 TO,13,2H *,F7.5,6H = 2PI,/) 
FRMAX = NOM+TINT

WRITE(6,2) DOMO,NOM,FINT,FRMAX

FORMAT: 15H FREQUENCY FROM,F7.2,3H TO,13,2H *,F7.3,2H = ,
1F8.2,/, ******************************, //, PULSE PARAMETERS' ,
2/, *****************************, //
WRITE(6,3) AREA, WIDTH, AMP, CENTER

FORMAT:  ' PULSE AREA', F8.4,5X, ' PULSEWIDTH (FWHM/1.774) ',F8.6,
3/, ' PEAK FIELD AMPLITUDE', F10.3,7X, ' CENTER', F8.5,/'
4/, *****************************, //, MEDIUM PARAMETERS '/
WRITE(6,4) ALFAL, ALFAU, ABSU, STK, GAM2, COEF

FORMAT: ' ALFAL = ',F8.3,4X, ' ALFAU = ',F8.3,6X, ' ABSU = ',F8.3,
16X, ' STK = ',F8.3,/, ' GAM2 = ',F8.3,8X, ' COEF = ',F8.3)
WRITE(6,5) T2INV, T1INV

FORMAT: RELAXATIONS: 1/T2 = ',F8.3,/,15X, ' 1/T1 = ',F8.3

WRITE(6,6) DZ

FORMAT:  ***************, DZ = ',F5.3,/)  ***************,

WRITE(1,2) DOMO,NOM,TINT,FRMAX
WRITE(1,3) AREA, WIDTH, AMP, CENTER
WRITE(ALFAL, ALFAU, STK, GAM2, COEF
WRITE(1,5) T2INV, T1INV
WRITE(1,6) DZ

RETURN
END
APPENDIX C

The derivation for the equations of $\sigma_{12}, \sigma_{22}$ and $(\sigma_{11} + \sigma_{22})$ will be given in this appendix.

In general the matrix elements $\rho_{ij}$ can be expanded harmonically as,

$$\rho_{ij}(t) = \sum_{n=-\infty}^{\infty} \sigma_{ij}(n\omega, t) e^{i n \omega t}$$  \hspace{1cm} (C.1)

where the slowing varying amplitude approximation (will be used to solve the off resonant density matrix elements) is given by,

$$\sigma_{ij}(n\omega, t) \ll \omega \left| n \sigma_{ij}(n\omega, t) \right|$$  \hspace{1cm} (C.2)

Neglecting the reaction of the third harmonic field (which is rather small) we have the real electric field as,

$$E(z,t) = \vec{E}(z,t) e^{i(\omega t - k z)} + C.C.$$  \hspace{1cm} (C.3)

where $\vec{E}(z,t)$ is polarised along the $\hat{z}$-axis and writing $\mu$ for $\mu_\perp$, $E$ for $E(z,t)$, $\mathcal{E}$ for $\mathcal{E}(z,t)$ we have the equation of motion of the density matrix elements as

$$im \frac{\partial \rho}{\partial t} = [ H_0 - \mu E, \rho ]$$  \hspace{1cm} (C.4)
The density matrix we consider here are $\rho_{11}$, $\rho_{22}$, $\rho_{12}$ and $\rho_{e}$ and their complex conjugates. Taking these matrix elements of equation (C.4) and using $H |ij\rangle = h_i |i\rangle$ and $\omega_{ij} = \omega_i - \omega_j$ we have,

$$\left(\frac{\partial}{\partial t} - i \omega_2 + 1/T_2\right) \rho_{12} = -i/h \sum_{\ell} (\rho_{\ell e} \mu_{\ell 2} - \mu_{\ell e} \rho_{\ell 2})$$  \hspace{1cm} (C.5)

where the phase relaxation time $T_2$ is introduced phenomenologically. The equation for $\rho_{22}$ with the population relaxation time $T_1$ is,

$$\left(\frac{\partial}{\partial t} + 1/T_1\right) \rho_{22} = -(i/h) \sum_{\ell} (\rho_{\ell e} \mu_{\ell 2} - \mu_{\ell e} \rho_{\ell 2})$$  \hspace{1cm} (C.6)

The equations for $\rho_{e}$ and $\rho_{2e}$ are respectively

$$\left(\frac{\partial}{\partial t} + i \omega_2\right) \rho_{2e} = -(i/h) \sum_{k=1,2} (\rho_{2k} \mu_{2e} - \mu_{2k} \rho_{2e})$$  \hspace{1cm} (C.7)

$$\left(\frac{\partial}{\partial t} + i \omega_1\right) \rho_{1e} = -(i/h) \sum_{k=1,2} (\rho_{1k} \mu_{1e} - \mu_{1k} \rho_{1e})$$  \hspace{1cm} (C.8)

The only harmonic that is relevant for the oscillation of $\rho_{12}$ is $n = 2$ in equation (C.1), ie.

$$\rho_{12}(t) = \mathcal{S}_{12}(2\omega, t) e^{i 2\omega t} + \mathcal{S}_{12}^*(2\omega, t) e^{-i 2\omega t}$$  \hspace{1cm} (C.9)
we see from equation (C.5) that with $E=0$ the natural frequency of oscillation of $\rho_1$ is $2\omega$ and so the antiresonant amplitude $\sigma_1^{\star}$ can be neglected. The relevant harmonics for the expansion of $\rho_1$ and $\rho_2$ are the first and third harmonics like,

$\rho_1(t) = \sigma_1(\omega,t)e^{i\omega t} + \sigma_1^{\star}(\omega,t)e^{-i\omega t} + \sigma_1(3\omega,t)e^{i3\omega t} + \sigma_1^{\star}(3\omega,t)e^{-i3\omega t}$

substituting this in equations (C.7) & (C.8) and using slowly varying amplitude approximation (equation (C.2)) we have,

$\sigma_1(\omega,t) = \frac{\varepsilon \sigma_{11}(2\omega,t)M_1}{\pi (\omega + \omega_1)}$ \hspace{1cm} (C.10)

$\sigma_1(3\omega,t) = \frac{\varepsilon \sigma_{12}(2\omega,t)M_2}{\pi (3\omega + \omega_1)}$ \hspace{1cm} (C.11)

Similarly expressions for $\sigma_2(\omega,t)$ and $\sigma_2(3\omega,t)$ can be derived. Using all these in equation (C.5) we get,

$\left(\frac{\partial}{\partial t} + i(2\omega - \omega_2) + 1/T_2\right)$

$= -\left(\frac{1}{m}\right) \sum_{\ell} \left[ \varepsilon \sigma_{1\ell}(\omega,t)M_{\ell\alpha} + \varepsilon \sigma_{1\ell}(3\omega,t)M_{\ell\alpha}^\star \right]$
Substituting for $\sigma_{1e}(\omega,t)$, $\sigma_{1e}(3\omega,t)$, $\sigma_{e2}(\omega,t)$ and $\sigma_{e2}(3\omega,t)$ above we get,

\[
\left( \frac{\partial}{\partial t} + i (2\omega - \omega_2) + \frac{1}{\hbar^2} \right) \sigma_{12} = \sum_{l} \left( \frac{\mu_{1e} \mu_{e2}}{\omega + \omega_{e2}} \sigma_{22} + \frac{\mu_{1e} \mu_{e2}}{\omega + \omega_{e2}} \sigma_{11} \right) \mathcal{E}^2
\]

\[
+ \sum_{l} \left( \frac{|\mu_{2e}|^2}{\omega + \omega_{e2}} + \frac{|\mu_{2e}|^2}{3\omega + \omega_{e2}} \right) \mathcal{E}^2
\]

\[
= \frac{\omega_2}{\hbar^2} (\sigma_{22} - \sigma_{11}) \gamma_{12} \mathcal{E}^2
\]

Using TPR condition $\omega_2 = 2\omega$ and identity $\omega_{1e} = \omega_{e2} + \omega_{2e}$ the above equation can be written as,

\[
\left[ \frac{\partial}{\partial t} + i (\omega - \omega_2 - \delta \omega_{21}) + \left( \frac{1}{\hbar^2} + \frac{\omega}{2} \right) \right] \sigma_{12} = \frac{i}{\hbar^2} (\sigma_{22} - \sigma_{11}) \gamma_{12} \mathcal{E}^2
\]

where the Stark shift $\delta \omega_{21}$ is given by the real parts of the polarisabilities as,

\[
\delta \omega_{21} = \frac{1}{\hbar} (\alpha_1' - \alpha_2') |\mathcal{E}|^2
\]

and the polarisabilities are
\[ \alpha_1 = \alpha_1' = \frac{1}{\hbar} \sum E \frac{2\omega E_1 |\mu_{1e}|^2}{\omega E_1^2 - \omega^2} \quad (C.16) \]

\[ \alpha_2 = \alpha_2' - i \alpha_2'' = \frac{1}{\hbar} \sum E \left( \frac{|\mu_{2e}|^2}{\omega E_2 - \omega} + \frac{|\mu_{2e}|^2}{\omega E_2 + \omega} \right) \quad (C.17) \]

The polarisability \( \alpha_2 \) is complex because of the coupling to the continuum. The intensity dependent ionisation \( \gamma \) is given by,

\[ \gamma = \frac{2}{\hbar} \alpha_2'' |E|^2 \quad (C.18) \]

and the transition matrix element \( \gamma_{i2} \) is,

\[ \gamma_{i2} = \sum E \frac{\mu_{i e} \mu_{e2}}{\omega E_2 + \omega} \quad (C.19) \]

where the summation is over all virtual intermediate levels, bound or free. Similarly we can get the equation for \( \sigma_{22} \) as,

\[ \left( \frac{\partial}{\partial t} + \frac{1}{T_1} \right) \sigma_{22} = -\frac{2}{\hbar} Re \left[ \gamma_{i2} \sigma_{i2} E \right] - \gamma \sigma_{22} \quad (C.20) \]

finally the equation for the total population becomes

\[ \frac{\partial}{\partial t} (\sigma_{11} + \sigma_{22}) = -\gamma \sigma_{22} \quad (C.21) \]
REFERENCES


2. Poluektov et al., "Coherent propagation of high-power light pulses through a medium under conditions of two-quantum interaction", JETP lett., Vol 20, No.8, (October 20, 1974), 243-244.


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