**MODIFICATION OF ATOMIC COLLISION DYNAMICS
BY INTENSE ULTRASHORT LASER PULSES**

by

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CURRICULUM VITAE

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ABSTRACT

During the past decade there has been a great deal of effort put toward demonstrating that the dynamics of atomic collisions can be modified by the presence of intense laser fields. The term "modified collision dynamics" means here that the potential energy surfaces which govern the collision dynamics are actually distorted by the ac-Stark effect induced by the intense laser field. This results in altered probabilities for the scatterers to end up in certain outgoing channels. The attractiveness of the idea of modified collisions, of course, lies in the possibility of selectively controlling physical or chemical processes by judicious choice of laser frequency and intensity. If one uses laser pulses whose duration is less than an individual collision then the experimenter can actively change the shape of the potentials during the collision. In principle, if one can open and close reactive channels at appropriate times during the collision, one can strongly influence its outcome. In this thesis the first experimental observation of the modification of atomic collision dynamics by ultrashort laser pulses is reported.

In order to more fully understand the interaction of the ultrashort laser field with the colliding atomic system, a theoretical model was developed using a solution to Schroedinger's equation in Bloch equation form. The numerical solution was then averaged over various uncontrollable parameters present in the

experiment when using a thermally random distribution of atoms. Averaging over these parameters as well as using a realistic temporal pulse shape and spatial beam profile has proven to be extremely important in modeling the experimental outcome.

In order to perform the experiments a new laser system was developed to provide an intense, ultrashort, tunable, laser pulse at the high repetition rates necessary for efficient signal averaging. This laser system demonstrated for the first time both the use of a CW Nd:YAG laser to produce sub-picosecond laser pulses and the technique of synchronous amplification to efficiently amplify the weak dye oscillator pulses.

The output of the dye oscillator-amplifier combination was used to study the collision process $\text{Na}(3s) + \text{Ar} + h\nu \rightarrow \text{Na}(3P_{\frac{1}{2}}) + \text{Ar}$. It has been found that at fixed laser intensity the efficiency of exciting the $\text{Na}(3P_{\frac{1}{2}})$ state is higher for pulses shorter than a collision duration than for pulses with duration longer than the collision duration. This effect is caused by a time-dependant modification of the collision potentials by the intense laser field.

Details of the Bloch-equation theory and the new Nd:YAG based laser system will be reported in this thesis. Experimental studies of the intensity dependance, detuning dependance and Ar pressure dependance on the output fluorescence intensity are also presented.

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INTRODUCTION

Atomic collisions have long been studied both theoretically and experimentally, for they play a dominant role in many of the fundamental problems of astrophysical and laboratory interest. Long before the advent of the laser, the basic processes involving colliding atoms had been predicted and observed. As lasers were used in the studies, however, it became apparent that the laser could be used not only as a tool to investigate the colliding species before and after impact, but also to manipulate or control the process as well. Thus the laser field could be used as a free parameter to modify the collision dynamics and influence the collision outcome. A large number of theoretical predictions have been reported¹⁻⁶ describing the influence of an intense field on collision dynamics, but only four experiments published to date have clearly shown such effects.⁷⁻¹⁰ The term "modified collision dynamics" means here that the potential energy surfaces which govern the collision dynamics are actually distorted by the ac-Stark effect induced by the intense laser field. This results in altered probabilities for the scatterers to end up in certain outgoing channels. The

attractiveness of the idea of modified collisions, of course, lies in the possibility of selectively controlling physical or chemical processes by judicious choice of laser frequency and intensity.

In this thesis we have taken a novel approach to this problem, as first proposed by Lee and George,⁵ by carefully choosing the duration of the laser pulse to be shorter than the duration of an individual collision. This allows the experimenter to actively change the shape of the potentials during the collision. In principle, if one can open and close reactive channels at appropriate times during the collision, one can strongly influence its outcome. The intent of this work is to study the modification of atomic collision dynamics due to an intense laser pulse whose duration is shorter than the collision itself.

I.A Overview of Experiment

When two atoms collide (either identical or dissimilar atoms) their proximity causes a time-dependent shifting of energy levels in each of the two atoms. The potential energy diagram with respect to the interatomic separation R is shown in figure 1-1. Many previous researchers have attempted to carefully map out the shapes of these potentials versus R both theoretically and experimentally.¹¹⁻¹⁴ If one applies a laser pulse to the colliding system then one creates a dressed level which is a copy of the ground state curve raised in energy by the energy of the applied laser photon $\hbar\omega$. Through

Figure 1-1 Potential energy curves for two-level atom (A) colliding with a perturbing atom (B). A dressed level (dashed line) is created by the presence of a photon of energy $\hbar\omega$. The dressed level and real excited level cross at an interatomic separation R_c .

judicious choice of ω the dressed level, shown as the dashed line in figure 1-1, and the excited state level can intersect at a particular interatomic separation R_c . Given that the distance of closest approach (impact parameter) for the collision is less than R_c , then there are two points in space and time where curve crossings occur. This is graphically illustrated in figure 1-2 assuming one of the two atoms is stationary and there is no trajectory perturbation. When the atoms exist at a separation R_c the laser is "in resonance" with the excited state due to the shifting of the levels and population can, at this point, be moved from the ground to the excited state. If one of the atoms ends up in its excited state, it is said that an optical collision,^{1,7} or collisional redistribution,^{4,6,8,17} has taken place. Thus even with a laser severely detuned from a resonance line, population can be excited to that resonance line through collisional redistribution where the atom later fluoresces as illustrated in figure 1-3.

As illustrated in figure 1-2, there are two points in space and time where the atoms have an interatomic separation R_c and where the laser field is "in resonance" with the excited state. These points are called curve crossings. The time between curve crossings, T_c , can be calculated knowing the radius R_c , the impact parameter b , and the relative velocity of the colliding atoms. This time can be used to define "long" laser pulses as those whose duration is greater than T_c , and "short" pulses as those whose temporal duration is shorter than T_c . A "long" laser pulse therefore can interact with both curve crossings whereas a "short"

Figure 1-2 Geometry of the atomic collision. Straight line trajectories are assumed with one atom stationary and the other moving by at a velocity v in this reference frame. Given that the impact parameter (b) is less than R_c there are two points in space where the two atoms are at an interatomic separation R_c . The time it takes to travel between these two points is T_c .

Figure 1-3 Excitation mechanism in a picosecond optical collision. Excitation occurs at an interatomic separation R_c when the real and dressed levels are at their closest point. If excitation does occur then the atom later fluoresces at a frequency ω_0 .

pulse can interact with only one.

When two levels cross, either two real or a real and a virtual, they avoid one another due to the interaction between the two levels (given that there is an allowed transition between the two levels and that a laser field is present). In the case of a virtual level interacting with a real level this interaction is similar to the ac-Stark effect, with the smallest separation between the two levels being given by the Rabi frequency $\Omega = d \cdot E / \hbar$, where d is the dipole matrix element between the ground and excited states and E is the laser electric field amplitude. This is shown in figure 1-4 for the case of Na + Ar collisions. The simple Landau-Zener^{18,19} formula can be used to illustrate the principles involved (the Landau-Zener formula will be more completely described in Chapter II). This formula gives the probability of jumping across the avoided crossing as:

$$p = e^{-W} \quad , \quad (1-1)$$

where W is proportional to the laser intensity. The probability of moving from the virtual state to the real excited state in the course of completed collision is different for the cases of "long" and "short" pulses. In the "long" pulse case, where the avoided crossing is encountered twice, this probability is given by the probability of not jumping across the avoided crossing on the first pass but jumping on the second, plus the probability of jumping on the first pass but not on the second, or:

Figure 1-4 Detail of the avoided crossing for the case of Na-Ar collisions. The separation of the real and dressed level at the interatomic separation R_c is Ω .

$$p = 2e^{-W}(1-e^{-W}) \quad . \quad (1-2)$$

The "short" pulse case is illustrated in figure 1-5. At times before the laser pulse is present the target and perturber atoms are colliding with the target atom residing in the ground state. When the laser pulse is present the target atom is placed in the dressed level and traverses through the avoided crossing region. When passing through this region, however, if the target atom does not "jump" across the avoided crossing then it adiabatically moves onto the excited state potential. The dressed level then disappears, due to the laser pulse turning off, before the atoms can pass through a curve crossing point again. When this happens the target atom is trapped on the excited state potential curve. Thus the probability of ending up in the excited state in this case is:

$$p = 1-e^{-W} \quad , \quad (1-3)$$

or the probability of not jumping across the avoided crossing. One can see that at high laser intensities (large values of W) there is potentially great advantage in using "short" pulses ($p \rightarrow 1$) rather than "long" pulses ($p \rightarrow 0$). This is shown in figure 1-6. This results because at high intensity the separation between the avoiding potentials is great and the collision system adiabatically follows the avoiding potentials. In the "long" pulse case the system finds itself back in the initial state following the collision. In the

Figure 1-5 Excitation mechanism in a picosecond optical collision. Atom A is in the ground state as the atoms are coming together in the top diagram. When the atoms are at a separation equal to R_c the dressed level is created by the optical field causing the atom to move to the excited state. The atoms then move towards the impact parameter (b) and the optical field is turned off. Thus by purely adiabatic motions the atoms end up on the excited state. At times after the field is turned off the atoms move apart with the atom remaining in the excited state as shown in the bottom diagram.

Figure 1-6 Landau-Zener excitation probability curves for "long" and "short" pulses. The x-axis is in normalized units of W .

"short" pulse case the system adiabatically moves onto the excited state and is trapped there.

This thesis work is meant to experimentally verify these predictions, which were first described by Lee and George.⁵ In order to more completely treat the theoretical aspects of this interaction, a fairly rigorous theory using optical Bloch equations was developed to describe the collisional interaction when a realistically shaped pulse whose duration is shorter than a collision duration is applied. Experimentally an intense, stable, high-repetition-rate source of tunable ultrashort pulses had to be developed to investigate this intensity dependence for different duration pulses.

I.B Literature Review

Although Lisitsa and Yakovlenko¹ first described nonlinear effects in optical collisions more than ten years ago surprisingly little experimental work has been done on this problem and none has been done using ultrashort laser pulses. Part of the reason for this is that only recently have stable ultrashort sources become available to view the nonlinear variations in optical cross section predicted. In addition, at the same electric field strength a 7 nsec pulse has 7000 times as many photons as that in a 1 psec pulse making the fluorescence signal generated by the 1 psec pulse correspondingly

smaller.

Some work has been done, however, using pulses longer than the collision duration. Carlsten, Szoke, and Raymer¹⁷ made the first studies of the dependence of the collisionally redistributed signal on the laser detuning, laser intensity, and buffer gas pressure. The pulses used for these experiments were much longer than both the collision time and the time between collisions, so the results were due to multiple collisions rather than to individual collisions. For this reason, together with the experimental effects of spatial averaging and radiation trapping, the decrease in optical cross section at high laser intensities was not clearly observed.²⁰

In 1980 Bonch-Bruevich et. al.⁷ made the first experimental observations of the decrease in optical cross section at high laser intensities as predicted in Eq. (1-2). For this they used a frequency-doubled Nd:Glass laser with pulse durations of 10 psec at a wavelength of 530 nm, which happens to be 50 Å detuned to the red side of the Tl resonance line. This value of t_p is greater than T_c so it is considered a "long" pulse, however it is shorter than the time between collisions. Although the predicted non-linearity did occur, it was at a much higher laser intensity (10^9 W/cm²) than theoretically predicted (about 10^6 W/cm²). The results are presented in figure 1-7. Further interpretation of these results beyond what is shown in the figure is impossible because of the sketchy nature of the description of the experimental arrangement. Several questions are raised in reading this paper. The first is a lack of a value for the number density that could be used to rule out

Figure 1-7 Bonch-Bruевич results for decrease in optical cross section at high input laser intensities. The top photographs are oscilloscope traces of the fluorescence intensity for increasing (a to c) input laser intensities. The photodiode-oscilloscope combination could not resolve the individual pulses in the Q-switched train. Photograph c indicates that at the peak of the Q-switched train where the input laser intensity is highest, the fluorescence intensity is actually less than earlier and later in the laser pulse train where the input laser intensity is less. The graph at the bottom is a tabulation of the signal out versus the input laser intensity illustrating the fluorescence decrease at higher input intensities.

the possibility of self-focussing producing the observed effect. Another is the use of a train of pulses (as opposed to a single pulse) to perform the experiments with a time between pulses approximately equal to the e^{-1} decay time of the excited Tl atoms. This could lead to optical pumping effects at high laser intensities.

Light and Szoke⁴ proposed that the ideal 2-state model for the target atom, used by previous researchers because of its simplicity, did not adequately describe the optical collision dynamics correctly since the excited level is very rarely a single state but rather is composed of several states that can all interact with the laser field (these results will be more completely described in Chapter II). Kleiber et. al.⁸ observed this effect in the Sr-Ar collision system by using an intense 7 nsec pulse to excite the $m_j=0,+1,-1$ resonance levels at $\lambda=460$ nm in the Zeeman-degenerate Sr atom. Qualitative agreement was found between the theoretical predictions⁶ and the experimental results of Kleiber et. al.⁸ when observing the depolarization of the collision-induced resonance fluorescence as a function of the ratio of the Rabi frequency to the laser detuning. These results do indicate that the influence of multiple excited states should be taken into account when interpreting experimental results.

At much lower intensities Havey et. al.²¹ have investigated the branching ratio (the ratio of the atomic number that end up in the D₁ and D₂ lines) for the Na+Ar collision system as a function of the detuning from the two lines. They find a value of the branching ratio independent of the detuning when the argon pressure

is increased to the point that the time between collisions is less than the decay time of the Na 3P lines. At lower values of the argon pressure information on the shape of the potential curves can be determined from the ratios. Indications are made in this work that the Π state of the D₂ line significantly alters the shape of the potential curves from the D₁ line due to additional avoided crossings generated by the interaction of the two excited states.

Nakatsuka and Grischkowsky²² have looked at pulse propagation of picosecond laser pulses traveling in a near-resonant atomic vapor. In their experiments they used the large group velocity dispersion present near an atomic resonance to broaden or to compress the picosecond optical pulse with its broad spectral bandwidth. The Na atomic densities used were very high ($3 \times 10^{15} \text{ cm}^{-3}$) giving the large group velocity dispersion they desired. For the experiment described in this thesis, the density was kept much less than this so that the effects of dispersion would not alter the pulse shape as it propagated through the atomic vapor cell.

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CHAPTER II

THEORY

Before the invention of the laser, the effects of collisions were studied by observing the broadening of the resonance line (both in absorption and emission) due to dephasing collisions. In the last decade, however, considerable work has been carried out towards studying collision dynamics more directly. Intense lasers, which have recently been developed, can be used to access the extremely weak collisional signals. From this the processes of optical and radiative collisions have been experimentally observed. In this chapter a description of collisions, optical collisions, and curve crossings will be presented as a preface to the theory developed to describe picosecond optical collisions.

II.A General Atomic Collision Theory

When any two atoms collide their energy levels warp or distort due to the perturbation of one atom on the other. The magnitude of

the perturbation is given by the nature of the interaction between the two colliding atoms. Resonance collisions, or collisions between like atoms, have (as expected) a strong interaction and the levels warp severely (the energy levels are displaced as R^{-3} where R is the interatomic separation). Van der Waals collisions, or collisions between dissimilar atoms, do not have the same strong interaction and do not warp as much (as R^{-6}). Spectroscopically this warping translates into not only a broadening of the resonance line but also into a shift in the peak and an asymmetry in the lineshape.¹

Given that there is a one-to-one correspondence between the interatomic separation between the colliding atoms and the detuning they generate by their interaction, there are two regions in both the absorption spectrum and in space which require different theoretical methods to describe the scattered spectral output. The two regions are called the impact region and the quasi-static region. In the impact region the required interaction is less since the detuning is less, the interatomic separation for collisions in this region is greater, and therefore the time between collisions is short. Therefore, the scattered spectral output in the impact regime has resulted from many separate collisions which perform an averaging over the details of an individual collision. In the quasi-static region, however, the required interaction is much stronger, the interatomic separation is much smaller, and the time between collisions is much longer. In this case, the output is determined not by multiple collisions, but rather by a single collision. The point where one moves from one region to the other is rather

nebulous, but has been defined roughly as:²

$$\Delta_g = 1.65 v^{\frac{1}{2}} N_p^{\frac{1}{2}} \gamma_E^{-\frac{1}{2}} \quad (2-1)$$

where Δ_g is the spectral detuning from line center where the two regions meet, v is the mean relative velocity of the two atoms, N_p is the density of the perturbers, and γ_E is the half width at half maximum of the collision-broadened line. For foreign gas broadening the broadening rate is given as:

$$\gamma_E = 4.04 N_p v \left(\frac{C_6}{v} \right)^{\frac{2}{5}} \quad (2-2)$$

where C_6 is the Van der Waals interaction constant. In our particular case of Na+Ar collisions detuned to the red side of the D_1 line, Δ_g at the temperatures and pressures used in the experiment is:

$$\Delta_g = 6.8 \text{ \AA} \quad (2-3)$$

Laser detunings greater than this number are interacting with atoms in the quasi-static regime, and detunings less than this number interact with atoms in the impact regime. The detunings used in the experiments are much greater than this indicating that they were done in the quasi-static regime.

The details of atomic interactions in the impact regime have been thoroughly studied and are well understood.³⁻⁶ Collisions in

the non-impact region have not been as extensively studied due to the complicated trajectory and interaction dynamics of close individual collisions, and the extremely weak signal strengths one obtains in the far wings of the resonance line. In order to observe the atoms in this region, the laser beam used to probe the collisions must be made very intense (or have a high average power) to provide an observable signal. The intense applied optical electric field therefore not only observes the atoms, but necessarily also perturbs them. The effects of this perturbation surround the subject of optical collisions and much of the theoretical work⁷⁻⁹ done so far.

II.B Laser-induced Curve-Crossings

Common to all of the work done in the non-impact regime is the need to understand and describe curve-crossings. When two atoms collide, as shown in figure 2-1, their energy levels warp due to the interaction between the two atoms. At some particular interatomic separation, R_c , two levels could intersect giving rise to some possibility of transferring population from one curve to the other. If there is an allowed transition between these two levels, then the two levels "avoid" each other in a manner similar to the ac-stark effect. Two pairs of potentials can be defined: diabatic potentials ϕ_1 and ϕ_2 , and adiabatic potentials V_1 and V_2 . Diabatic potentials are defined as those that cross at the particular interatomic separation R_c whereas the adiabatic potentials are the

Figure 2-1 Potential energy curves illustrating the avoided crossing of two real atomic levels.

non-intersecting potentials. The dotted lines in figure 2-1 illustrate the diabatic potentials in the curve crossing region. Atoms starting on one of the two potential curves at $R \gg R_c$ move through the curve crossing region and can either jump to the other adiabatic curve or can remain on the one that they came in on. As the two atoms move apart there is another pass through the curve-crossing region and atoms can again undergo a change in the potential curve they are on at that point. In each collision where $b < R_c$, therefore, there are two curve crossings.

Generally, curves do not intersect except at extremely small interatomic separations where non-elastic and sticking collisions occur. One can create a dressed state, however, which can, with judicious choice of the laser frequency used, intersect an excited state potential curve at a chosen interatomic separation. This is illustrated in figure 2-2. When the laser field is applied to the atoms a dressed level is created which is an exact copy of the lower state raised up in energy by the value $\hbar\omega$. At a given value of there is a particular R_c where the excited level and the dressed level intersect. At this point we have a curve crossing similar to the ones described earlier. The strength of the interaction between the dressed and excited levels is given by Ω which is defined as:

$$\Omega = \frac{d_{12} \cdot E}{\hbar} \quad (2-4)$$

where d_{12} is the dipole matrix element between the ground state (1) and the excited state (2) of the target atom and E is the magnitude

Figure 2-2 Potential energy curves for the collision system Na-Ar. The dotted line indicates the dressed level created by the presence of the laser field of frequency ω_L . The real excited level and dressed level intersect at an interatomic separation R_c and have an energy separation at that point equal to Ω , the Rabi frequency.

of the applied electric field strength. In a manner similar to the ac-stark effect, the separation between the two adiabatic curves at the point R_c is given by the interaction strength Ω .

If the atom enters the collision on the dressed level and by moving from one adiabatic curve to the other ends up on the excited state, then the probability for it to end up in the excited state can be monitored by measuring the magnitude of the fluorescence which is emitted as the atom relaxes to the ground state. The variation in fluorescence intensity when one changes the input laser intensity or the laser detuning can tell one much about the interaction between the two atoms and the applied electric field.

II.C Landau-Zener Effect

The main points of interest in the study of curve crossings are the factors which determine whether the atom will make the transition across the avoided crossing. Landau,¹⁰ Zener,¹¹ and Stueckelberg¹² each independently derived the "Landau-Zener formula" describing the probability of "jumping" across the avoided crossing. Each used adiabatic potentials to describe the avoided crossing and made the following assumptions. First, the energy difference across the avoided crossing is much less than the kinetic energy of the colliding pair so that the loss in kinetic energy is negligible during a "jump" across the avoided crossing. And second, the transition region is so small that in it the detuning between the non-adiabatic potentials is a linear function of interatomic

separation and time, given the velocity of the atoms. Given these assumptions and by using the Weber's function they obtained the probability P of jumping across the avoided crossing as:

$$P = e^{-W} \quad (2-5)$$

where

$$W = \frac{2\pi h_{12}^2}{\hbar v \left| \frac{dV_2}{dR} - \frac{dV_1}{dR} \right|_{R_c}} \quad (2-6)$$

h_{12} is the non-diagonal Hamiltonian matrix element (a measure of the interaction strength), v is the internuclear velocity, and $V_1(R)$ and $V_2(R)$ are the diabatic potential curves of the two states. For the case of optical collisions the interaction matrix element is given by the Rabi frequency and the formula becomes:

$$W = \frac{2\pi \hbar \Omega^2}{v \left| \frac{dV_2}{dR} - \frac{dV_1}{dR} \right|_{R_c}} \quad (2-7)$$

When two atoms move past each other in unperturbed motions there are in general two points in their travel where $R=R_c$, as shown in figure 2-3. On the potential curves this corresponds to moving from $R=\text{infinity}$ to $R=R_{\min}$, then turning around and moving towards $R=\text{infinity}$ again. If $R_{\min} < R_c$ then there are two points in both time and space where $R=R_c$. The probability of jumping to ϕ_2 is the sum of the probabilities of jumping to ϕ_2 on the first pass but not on the second and not jumping on the first but

Figure 2-3 Geometry of the colliding atoms. The separation at closest approach is defined as b . If b is less than R_c then there are two points in both space and time when $R=R_c$.

jumping on the second, or:

$$P_{\text{long}} = 2e^{-W}(1 - e^{-W}) \quad (2-8)$$

This is plotted in figure 2-4 versus the applied optical electric field strength. One notices that the peak probability of transfer is one-half and goes to zero at both high and low values of the exponent W .

The intensity at which the maximum excitation probability occurs (as seen in figure 2-4) can be determined by taking the derivative of Eq. 2-8 and setting it to zero. The value of the electric field strength at this peak point in the curve can be related to the applied optical intensity at this point by the equation (in cgs units):

$$I = \frac{c}{8\pi} E^2 \quad (2-9)$$

where I is in units of ergs/sec-cm², E in units of SV/cm and c in units of cm/sec. Solving the derivative of Eq. 2-8, and using Eqs. 2-7 and 2-9 we obtain the following equation for I_{crit} :

$$I_{\text{crit}} = \frac{.69 \, c \, v \left| \frac{dV_2}{dR} - \frac{dV_1}{dR} \right| R_c \, \hbar}{16\pi^2 d_{12}^2} \quad (2-10)$$

The value for I_{crit} (and by use of Eq. 2-9 also the value for E_{crit}) will be used throughout this dissertation to indicate the point at which the non-linearity in the absorption cross section with

Figure 2-4 Landau-Zener probability curve for "long" laser pulses. The x-axis is in normalized units of W .

respect to applied laser intensity should occur. Calculated in this way, the value of I_{crit} is typically of the order of 10^6 W/cm^2 .

II.D Deficiencies in the Landau-Zener model

The Landau-Zener model for the interaction at curve crossings has been used extensively in the past work because of its inherent simplicity. This was particularly true in the early days before computers were developed, when large numerical simulations were impossible. There are, however, inaccuracies in the theory which make its use in making experimental predictions hazardous. Two assumptions made in the development of the theory have been correctly challenged.

The first is the assumption that the transitions can only occur in a small region very near the curve crossing point.¹³ This is in fact not the case. The transition region is a vaguely defined zone around the curve-crossing which is defined by not only the interaction strength but also the slopes of the two intersecting potential curves. Although it is most probable that transitions will be made at the point at which the two adiabatic curves are the closest, there is a finite probability that transitions can be made nearby. At high field strengths where the levels are well separated, the transition region can get quite large and the assumption that the two levels could be approximated by straight lines over the region becomes invalid. This assumption has been taken into account by

Bates¹³ and a qualitative curve illuminating the differences this assumption can make is displayed in figure 2-5. In this graph the solid line is the Landau-Zener model and the dotted line is the corrected curve. The abscissa is somewhat different than in figure 2-3; the axis is in units of energy of the colliding atoms or proportional to the square of the interatomic velocity (when this paper was written most collision studies were made with accelerated atoms and ions rather than those at thermal velocities as we have studied).

The second error comes in naively assuming that we have a true two-level atom. In nearly all atomic systems, this is not the case. Light and Szoke¹⁴ have taken into account the fact that there is not a single avoided crossing but rather many different ones which can interact with each other. In their paper, they consider the case of a four state atomic system such as that of the case of $\text{Sr}+\text{Ar}$ collisions. The cross sections of each of the three excited states ($m_j=0,+1,-1$) as a function of the impact parameter of the collisions were determined analytically and then numerically averaged. The total excitation cross section they calculated is shown in figure 2-6. In a later paper by Kleiber et al.¹⁵ a more rigorous calculation was made and similar results were found but without the "glitch" that occurs in the Light and Szoke results after the peak in the cross section. The dramatic difference in the behavior of the fall-off at high values of E illustrates clearly the effect of multiple states modifying the outcome of the optical collision. In the case of a resonance collision this problem is

Figure 2-5 Corrected version of Landau-Zener curve when one takes into account the finite transition region where excitation can take place. Curve taken from Reference 13.

Figure 2-6 Effect of the induction of a four level model (instead of the unrealistic two) on the excitation cross section for the case of Sr-Ar collisions. The dashed line which decays first is from the Landau-Zener model, the next dashed line is a more accurate two-state model, and the solid line is the effect of a four-state model. Curve taken from Reference 14.

exacerbated by the fact that there are a plethora of levels due to the strong interaction between the two colliding atoms. In the case of non-resonant collisions (such as Sr-Ar or Na-Ar) the problem is somewhat better however still remains far from an ideal two-level system.

II.E Curve crossings in the presence of ultrashort pulses

In 1979 Lee and George¹⁶ described the dramatic difference that one could obtain by using ultrashort laser pulses to address the colliding atoms rather than longer laser pulses. By using pulses shorter than the collision time itself, an increase in the excitation efficiency could be obtained as described in the Introduction. The formalism used and the results are particularly illustrative to the results of this dissertation and as such a review of this work is warranted.

Several assumptions and approximations were made to simplify the problem in order to achieve an analytically solvable result. Unrealistic square laser pulses were used without any attention paid to the broad spectral bandwidth this would generate. Straight line, constant velocity trajectories were assumed. The temporally square laser pulse was always assumed to be centered over the time that the atoms pass through the curve-crossing point. Given these assumptions, the major results of this paper are the analytic expressions for the number excited by an optical collision. These are:

$$N_{\text{exc}} = 4T_p N_p \nu \left[\pi R_c^2 + \frac{2\pi}{\Theta} [(\Theta R_c + 1)e^{-\Theta R_c} - 1] \right] \quad (2-11)$$

for the case of a pulse that is "short" compared to the collision time and:

$$N_{\text{exc}} = 2T_p N_p \nu \left[\frac{3\pi}{\Theta} - \frac{4\pi}{\Theta^2} (\Theta R_c + 1)e^{-\Theta R_c} + \frac{\pi}{\Theta^2} (2\Theta R_c + 1)e^{-2\Theta R_c} \right] \quad (2-12)$$

for the case of a pulse that is "long" compared to the collision time. Θ is defined as:

$$\Theta = \frac{2\pi}{\hbar \nu} \frac{d_{12}^2 E^2}{\alpha R_c} \quad (2-13)$$

where d_{12} is the electronic transition moment between the two levels, E is the electric field strength, and α is the absolute value of the difference in slopes of the two levels at the curve-crossing point. Here, for illustration, these equations have been evaluated for the particular case of Na-Na collisions. A laser detuning of 20 Å from the $3P_{1/2}$ resonance line in Na implies a curve-crossing at an interatomic separation of 14.9 Å.¹⁷ An interatomic velocity of 5×10^4 cm/sec used with the value $R_c = 14.9$ Å gives a time between curve crossings of about 5 psec. Therefore, the pulsewidth used for the "short" pulse case was 2 psec and for the "long" pulse case was 100 psec. The electronic transition moment is given as 1.074×10^{-17} esu-cm¹⁸ and the density of perturbers (and

Na atoms in this resonance collision case) is 10^{16} cm^{-3} .

Equations 2-11 and 2-12 are plotted using these parameters in figures 2-7 and 2-8. They are plotted together for comparison in figure 2-9. Apparent in figure 2-9 is the result that at moderate E fields (200 SV/cm) the number excited is larger for the long pulse case than the short pulse case. This is because a long pulse at the same electric field strength has more photons in the pulse and, even with a smaller optical cross section, the number excited could be greater. In order to accurately compare these two curves, equations 2-7 and 2-8 were divided by the number of incident photons and the number of Na atoms, giving a pulsewidth-dependant optical cross section. This is plotted in figure 2-10. One can clearly see in this plot the advantage that one has in using short pulses over long pulses in exciting the population at high field strengths.

It is interesting to note that the cross section curves in both pulsewidth cases asymptotically come to the same value at small values of the electric field strength. This is as one might expect since we are used to finding a return to the perturbative approximation for the number excited when viewing a saturation curve at low laser intensities. In this case, however, the collision physics is inherently different for the case of "long" versus "short" laser pulses. The dipole moment of the atom is the same in both cases but the atoms must still go through the curve crossing point even at low laser electric field strengths. As they pass through the curve crossing point the method of exciting the atom is different for the two cases.

Figure 2-7 Number of atoms excited versus applied electric field strength in Na-Na collision system for "short" laser pulses. Equation taken from Reference 16.

Figure 2-8 Number of atoms excited versus applied electric field strength in Na-Na collision system for "long" laser pulses. Equation taken from Reference 16.

Figure 2-9 Number of atoms excited versus applied electric field strength in Na-Na collision system for both types of pulses illustrating the excitation population difference. Equations taken from Reference 16.

Figure 2-10 Optical crosssection in Na-Na collision system for laser detuned 20Å to the red of the D₁ line. This plot was made by dividing the results of Figure 2-9 by the number of photons per pulse and the number of Na target atoms.

Lee and George go further and discuss the ratio of the excitation probability for the case of the short pulse to the long pulse. This ratio can be thought of as a measure of the advantage one has in using short pulses over long pulses. The ratio is plotted in figure 2-11. This illustrates that at high laser electric field strengths there is a tremendous advantage in using short pulses over long pulses. This was the stimulus for the work in this dissertation.

II.F Bloch Equation Theory of Picosecond Optical Collisions

In order to more accurately describe the collision dynamics in the presence of picosecond optical pulses, a new theory has been developed. This theory describes the solution to the two-level Schroedinger equation in Bloch equation form over the duration of the collision event. Several of the difficulties with the results of both Landau, Zener and Stuckelberg and Lee and George have been avoided in taking this approach.

Several assumptions and inclusions were made in developing this theory which ought to be detailed. They are:

- i.) Straight-line classical trajectories are assumed with no change in the atomic velocities over the course of the collision.
- ii.) A linearly polarized input laser electric field is

Figure 2-11 Ratio between the efficiency of exciting with "short" laser pulses versus "long" laser pulses. The ratio is greatest at the highest values of applied electric field strength. This plot was made by dividing the results for "short" pulse by the results for "long" pulse illustrated in figure 2-10

assumed.

iii.) The input laser pulse shape is Gaussian with different pulse durations.

iv.) Pure R^{-3} potential shape for the difference potential ($V_2 - V_1$) in the case of resonance collisions (Na+Na).

v.) Pure R^{-6} potential shape for the difference potential in the case of van der Waals collisions.

vi.) Ground and excited state potentials are spherically symmetric between the two colliding atoms.

vii.) The transition dipole moment is unaffected by the collision itself. The magnitude remains constant and independent of R .

viii.) The transition between the ground state and excited state is an allowed transition.

ix.) Time between collisions is much longer than both the collision time and the laser pulse width so as to neglect the effects of multiple collisions.

x.) The impact parameter between the two colliding atoms is

determined solely by thermally random motions of the atoms.

xi.) The time of closest approach can occur at any point during the laser pulse shape.

xii.) The detuning of the laser from the resonance line is small compared to the kinetic energy of the colliding atoms so that there is little loss in kinetic energy when an atom is excited. This assumption is related to i.) above which assumed constant velocity motion over the course of the collision event.

xiii.) No extra coupling terms are included to account for the effects of nuclear motions of the two atoms or the translational motion of the electron when it is excited from an S to P state.¹²

xiv.) The laser frequency is near a resonance of one of the colliding atoms and the rotating-wave-approximation is valid over all of the collision event.

xv.) There is only one excited level and one ground state level providing a pure two-level system.

xvi.) The theory can be adequately described by the

evaluation at one value of the velocity and generalized for the entire Maxwell-Boltzmann velocity distribution.

Nearly all of these assumptions are straight-forward and provide little opportunity for disagreement. Assumptions xv. and xvi. are, however, suspect. The variation in the output of this theory has been studied versus different atomic velocities and has proven somewhat valid the assumption xvi.. Section II.L will illustrate the velocity dependance of the transition probability. As mentioned in the previous section, Light and Szoke¹⁴ have illustrated the need to treat the system as more than a two-level system. This was not done in the present theory for the following reasons. First, accurate potentials for the excited state levels are not available. The exact form of these potentials is particularly important for the case of multiple excited states to estimate the strength and form of their interaction. Secondly, multiple detunings from the multiple excited potential surfaces are more difficult to incorporate into a standard Bloch equation form. And third, in order to perform the numerical solution on the two-level model took nearly ten days of computer time. Further complications in the numerical solution could only make the code run for a longer period of time.

The two-level Schroedinger equation in standard Bloch equation form is given as:¹⁹

$$\dot{u} = -(\omega_0 - \omega)v \quad (2-14)$$

$$\dot{v} = (\omega_0 - \omega)u + \kappa Ew \quad (2-15)$$

$$\dot{w} = -\kappa E v \quad (2-16)$$

where u and v are the in-phase and in-quadrature components of the atomic dipole moment, w is the single-atom population difference or the inversion, ω_0 is the resonance frequency, ω is the laser frequency, E is the magnitude of the applied electric field strength and κ is a measure of the strength of the atomic line, defined by:

$$\kappa \equiv \frac{2d_{12}}{\hbar} \quad (2-17)$$

where d_{12} is the magnitude of the atomic dipole matrix element.

Modifications were made in equations 2-14, 2-15, and 2-16 to adapt them for use describing the optical collision. First E was replaced by $E(t)$, a time-dependent description of the envelope of a gaussian temporal laser pulse shape as given by assumption iii. Second, the value of ω_0 was replaced by one which is time dependent. This time-dependent resonance frequency comes about due to the shifting of the energy levels as the atoms collide. This modification follows assumptions i., iv., and v. Equations 2-14,

2-15 and 2-16 are therefore rewritten as:

$$\dot{u} = -(\omega_o(t) - \omega)v \quad (2-18)$$

$$\dot{v} = (\omega_o(t) - \omega)u + \kappa E(t)w \quad (2-19)$$

$$\dot{w} = -\kappa E(t)v \quad (2-20)$$

The form of $\omega_o(t)$ can be determined knowing the type of collision (resonance or van der Waals) together with the assumption of straight-line atomic trajectories. If one assumes rectilinear motion, then one can calculate the interatomic separation as a function of time as:

$$R(t) = (b^2 + v^2(t - t_o)^2)^{\frac{1}{2}} \quad (2-21)$$

where b is the impact parameter (separation between the two atoms at the point of closest approach), v is the velocity of the perturbing atom, and t_o is the time of closest approach between the two colliding atoms (the frame of reference has been chosen so that the target atom is at rest and the perturbing atom moves by it). In the case of resonance collisions the resonance level as a function of interatomic separation R is given as:

$$\Delta\omega(R) = \pm \frac{C_3}{R^3} \quad (2-22)$$

where C_3 is the interaction constant. Combining equations 2-21 and 2-22 we obtain:

$$\omega_0(t) = \omega_{00} - \frac{C_3}{(b^2 + v^2(t-t_0)^2)^{\frac{3}{2}}} \quad (2-23)$$

for the time-dependent change in resonance frequency.

When using laser pulses of the order of 1 picosecond in duration there is a finite and sizeable spectral bandwidth associated with it due to the uncertainty principle. This large bandwidth does not have to be explicitly taken into account; its effect is seen by solving for the temporal dynamics over the entire short laser pulse. An example of the effect of the large laser bandwidth will be shown later in this chapter.

Now that the functional form of all of the parameters is known, the differential equation can be solved numerically. A variety of different numerical techniques were used to solve the modified Bloch equations.

II.G Numerical Solutions of the Modified Bloch Equations

The numerical solution of the three coupled differential equations 2-18 - 2-20 was found using an altered form of the Euler-Cauchy polygonal approximation.²⁰ This particular method was

chosen after trying several other versions of the Euler method, and after the Runge-Kutta type proved to take more computing time to maintain the same accuracy as the method chosen. The parts of the solution that were particularly expensive in terms of the time they took were the calculations of the electric field strength and the time-dependent detuning and therefore they are calculated only once in each "predictor-corrector" cycle. The form of the numerical solution is:

$$y_{k+1} = y_k + \frac{1}{2} [f_k + f(x_{k+1}, y_k + f_k \Delta x)] \Delta x \quad (2-24)$$

In words, the parameters are evaluated at the initial point and are used with the differential equation to find the predicted final point. The slopes at the initial and final points are then averaged together and the average value is then used to determine the corrected final point. This is graphically displayed in figure 2-12.

The Bloch equations have another relationship which can be used in this situation. The values for u , v , and w , are related¹⁹ (since we can ignore relaxation when using picosecond pulses) through the equation:

$$u^2 + v^2 + w^2 = 1 \quad (2-25)$$

which can be used to eliminate one of the three differential equations providing two equations with two unknowns. Instead of

Figure 2-12 Differential equation method of solution. The top diagram illustrates using the slope at the initial point to find the predicted value at the next x value. The slope at the predicted point is averaged with the slope at the initial point to determine the corrected value as illustrated in the bottom diagram.

doing this, the relationship provided an easy method to check on the accuracy of the numerical solution by noting the value of one minus the sum as shown in equation 2-25.

An example of the numerical solution to the modified Bloch equations is displayed graphically in figure 2-13. In this case a resonance collision was addressed with a 10 psec pulse. The detuning, plotted in figure 2-13b illustrates that a 10 psec pulse is a "long" pulse since the two points in time where the detuning goes through zero (system passes through a curve crossing) are separated by a time less than 10 psec. The end result of w plotted in figure 2-13f is, of course, the number of interest since it represents the number of atoms excited during the optical collision. One notices that there is change in this value only during a zero crossing in the detuning as expected, since this is where there is a curve-crossing and the two potential curves interact. The instantaneous frequency of oscillation of u , v , and w is proportional to the magnitude of the effective field vector defined as:

$$|\Omega| = (\kappa^2 E^2 + (\omega_o(t) - \omega)^2)^{\frac{1}{2}} \quad (2-25A)$$

This is indeed the case since the oscillation frequency is the greatest at the peak of both the detuning and the laser pulse electric field strength. The error can be seen to be increasing during the period of rapid oscillation of u and v (Figures 2-13d and 2-13e). The numerical solution obviously cannot fully maintain the accuracy during this period as seen by the monotonically increasing

Figure 2-13 Results of numerical solution. The applied electric field is plotted in a.), the detuning is plotted in b.), and the error defined as $u^2 + v^2 + w^2 - 1$ is plotted in c.). The three components of the pseudospin vector are plotted in d.), e.), and f.). u and v are the in-phase and in-quadrature components while w is related to the excitation population or probability by $(w+1)/2$.

error in figure 2-13c.

Using this particular form of the Euler method, the error should decrease as N^{-2} where N is the number of iterations. This was tested and the results are plotted in figure 2-14. The slope of the errors indicates that the decrease is actually greater than the N^{-2} dependence expected.

For reasons that will become apparent later, the speed in solving this differential equation over the temporal region of interest had to be optimized fully. Several methods were used to undertake this. First, a variable step size in the numerical solution was instituted. Since the rapid oscillation is the main culprit in providing the error, the variable step size was inversely related to the magnitude of the effective field vector providing an error less than a 10^{-3} to 10^{-4} in each differential equation solution (the need for this small error will be illustrated later). Secondly, since there is virtually no change in the value of w before the first curve crossing, even in the presence of the electric field, the numerical solution is started just before the first crossing. The pseudospin vector (the vector described by (u,v,w)) is initialized by assuming (this was checked with the full numerical solution) that the vector adiabatically follows the movement of the effective field vector prior to the curve-crossing. The initial values for u , v , and w at $t=t_i$ are:

Figure 2-14 Error generated as defined by $u^2+v^2+w^2-1$ for different numbers of iterations taken over the same time range.

$$u(t_i) = \frac{\kappa E(t_i)}{(\kappa^2 E^2(t_i) + (\omega_o(t_i) - \omega)^2)^{\frac{1}{2}}} \quad (2-26)$$

$$v(t_i) = 0 \quad (2-27)$$

$$w(t_i) = \frac{-(\omega_o(t_i) - \omega)}{(\kappa^2 E^2(t_i) + (\omega_o(t_i) - \omega)^2)^{\frac{1}{2}}} \quad (2-28)$$

And third, after the second curve crossing the pseudospin vector is precessing around the effective field vector and will continue to until the dipole (excitation) decays. While the laser is still present, however, there are Rabi oscillations present on the w curve (inversion) which impede obtaining the final value for w. The angle between the effective field and pseudospin vector will remain constant over the remainder of the laser pulse in an adiabatic following form similar to that prior to the first curve crossing. Therefore, the final value can be calculated knowing the instantaneous values of u, v, and w at t_f , just after the second curve crossing. The final value for w as a function of the instantaneous values of u, v, w, and E at a time $t=t_f$ is calculated to be:

$$w_F = \frac{-\kappa E(t_f)u + (\omega_o(t_f) - \omega)w}{(\kappa^2 E^2(t_f) + (\omega_o(t_f) - \omega)^2)^{\frac{1}{2}}} \quad (2-29)$$

These three methods combined to reduce the computation time by a

factor of thirty.

II.H Single-Trajectory Results of Numerical Solution

Solutions were made to the modified Bloch equations for both resonance and van der Waals collisions. Collisions between two Na atoms were considered for the resonance collisions, and collisions between Na and Ar atoms were considered for the van der Waals collisions. The parameters associated with these collisions when making optical collision transitions between the $3S_{1/2}$ and the $3P_{1/2}$ levels are tabulated in table I.

The results for the two different cases are qualitatively similar. The value of I_{crit} found using the numerical solution is somewhat greater and the excited population is somewhat less for the case of Na-Ar collisions than for Na-Na collisions; however the dynamics remain similar. For this reason, discussions of the qualitative shapes seen in the numerical solutions will not be shown for both cases, but rather just for the Na-Na case. The final fully integrated solution will, however, be shown for both cases.

Many plots were made illustrating the dynamics of the picosecond optical collisions. Three representative ones are shown in figures 2-15, 2-16, and 2-17. In all three runs the pulsewidth is 10 psec ("long") with the peak of the pulse centered at the time of closest approach of the collision. A detuning of $20 \text{ \AA}$ gives a value of R_c equal to $14.9 \text{ \AA}$; the impact parameter, for this particular

TABLE 1

$$\begin{aligned}
 D_1 \quad & \lambda = 5895.9 \text{ \AA} \\
 & g_1=2 \quad g_2=2 \\
 & A_{21} = .618 \times 10^8 \text{ sec}^{-1} \\
 & f = .322
 \end{aligned}$$

$$\begin{aligned}
 D_2 \quad & \lambda = 5890.0 \text{ \AA} \\
 & g_1=2 \quad g_2=4 \\
 & A_{21} = .622 \times 10^8 \text{ sec}^{-1} \\
 & f = .647(18)
 \end{aligned}$$

Resonance Collisions Na + Na D₁ line

$$C_3 = 3.6 \times 10^{-8} \text{ rad cm}^3/\text{sec}(17)$$

$$k_{br} = 3.07 \times 10^{-7} \text{ cm}^3/\text{sec}(21)$$

Van der Waals Collisions Na + Ar D₁ line

$$C_6 = 3.8 \times 10^{-31} \text{ rad cm}^6/\text{sec}(22)$$

$$k_{br} = 8.82 \times 10^{-10} \text{ cm}^3/\text{sec}(23)$$

Figure 2-15 Results of numerical solution for a peak applied electric field of 50 SV/cm. The applied electric field is plotted in a.), the detuning is plotted in b.), and the error defined as $u^2 + v^2 + w^2 - 1$ is plotted in c.). The three components of the pseudospin vector are plotted in d.), e.), and f.). u and v are the in-phase and in-quadrature components while w is related to the excitation population or probability by $(w+1)/2$.

Figure 2-16 Results of numerical solution for a peak applied electric field of 100 SV/cm.

Figure 2-17 Results of numerical solution for a peak applied electric field of 150 SV/cm.

trajectory, $10 \text{ \AA}$, is less than this, separating the two curve crossing points by about 4.5 psec. The results of the numerical solution indicate that E_{crit} , the laser field strength above which the population begins to decrease for "long" pulses, is equal to 88 SV/cm. (Note: these plots were made before the inclusion of the computing time-saving measures mentioned in the previous section) Figure 2-15 was made at a value of $E=50 \text{ SV/cm}$ (less than E_{crit}), figure 2-16 at $E=100 \text{ SV/cm}$ (about E_{crit}) and figure 2-17 at $E=150 \text{ SV/cm}$ (greater than E_{crit}). The curves of w in each of the cases illustrates the change in population (or inversion) of the colliding atoms. In figure 2-15 there is an increase in the population at both of the two curve crossings. The probability of jumping across the curve crossing at this particular field strength can be seen to be about 15% on the first pass and about 25% on the second pass. This difference is due to the first crossing setting the dipole oscillating and the timing of the second curve crossing with respect to the first can either act in phase or out of phase with this oscillating dipole. This will be illustrated graphically when integrating over different impact parameters. In figure 2-16, the probability of jumping across the first avoided crossing is about 50%, and therefore there is as much population in the excited state as is in the ground state when making the second curve crossing. Since the probability of jumping from the ground state to the excited state is the same as jumping from the excited state back to the ground state, the population changes very little on the second pass. In figure 2-17, the probability is nearly 75% on the first pass

sending nearly all the population to the excited state; on the second pass nearly all the population then returns to the ground state. This illustrates very clearly the advantage that one has in addressing only "half" the collision (or only one curve crossing) at high electric field intensities. As shown in figure 2-17, if the second curve crossing was not addressed, then the population would remain in the excited state giving a much higher excitation efficiency.

A similar set of runs has been done for the case of a "short" laser pulse. The final population values obtained from both the "short" and "long" cases are plotted in figure 2-18. The "long" case is shown in figure 2-18a. The curve rises and falls in a similar fashion to that observed in both the Landau-Zener model and the Lee and George model. The value $E_{\text{crit}} = 88 \text{ SV/cm}$ corresponds to a laser intensity of $I_{\text{crit}} = .9 \text{ MW/cm}^2$, as compared to 1.4 MW/cm^2 predicted by the simple Landau-Zener formula (see Eq. (2-10)). The small "bump" after the main peak is due to a residual coherence between the two curve crossings. This has been predicted previously¹², albeit with respect to an increasing interatomic velocity rather than increasing electric field strength, and is called a Stuckelberg oscillation. The "short" pulse case is shown in figure 2-18b and exhibits a ringing itself at higher field strengths. The main result is that in the "short" pulse case the population does not decrease at high field strength, in qualitative agreement with Figure 2-9.

Figure 2-18 Final values of w for different peak applied electric fields. Graph a.) illustrates the dependence for "long" laser pulses while graph b.) illustrates the dependence for "short" laser pulses.

II.J Off-Resonant Excitation of non-Colliding Atoms

Whenever one applies a laser field detuned somewhat from a resonance line there is a finite probability that direct excitation of the line (atoms that are not colliding and do not have their levels warped) can occur. This mechanism could provide a method of obtaining excited-state population that is different from the optical collision, thereby complicating interpretation of the results.

The effect of this second, and competing, process was modeled using the same numerical solution as before however eliminating the time-dependent detuning and replacing it with a static detuning. The results are plotted in figure 2-19 for a 1 psec pulse, figure 2-20 for a .5 psec pulse, and figure 2-21 for a .1 psec pulse. A 10 psec case was also run, however there was virtually zero contribution to the population over the entire range of electric field strengths. The excited-state population can be seen to increase with increasing electric field strength in the applied laser pulse and also with decreasing laser pulse duration. There are several ways to view these results. In the Bloch sphere one can assume that the pseudospin vector does not adiabatically follow the Bloch vector exactly, and gets worse as one increases the change in electric field strength as a function of time either by increasing the peak electric field or by decreasing the pulse duration. In the spectral domain one notices that as pulse durations get shorter the Fourier transform bandwidth of the pulse must get larger, and more and more spectral

Figure 2-19 Effect of off-resonant excitation of non-colliding atoms when a 1 psec pulse is applied. Note the different x-scale from previous graphs.

Figure 2-20 Effect of off-resonant excitation of non-colliding atoms when a .5 psec pulse is applied.

Figure 2-21 Effect of off-resonant excitation of non-colliding atoms when a .1 psec pulse is applied.

components can overlap the resonance line giving rise to more excited-state population. In each of the three cases there is an oscillation in the population with respect to the different peak electric field strengths of the applied laser pulse. When one applies a pulse off resonance one induces Rabi oscillations in the excited-state population whose frequency is a function of the intensity of the applied laser field. At a given pulse duration there is a certain pulse energy at which the Rabi oscillation will be either at a peak in its oscillation or a trough. This gives rise to the variation in excited-state population as a function of electric field strength. In some sense, this is similar to the case of self-induced transparency where one can define at a given pulse duration a laser electric field strength which produces an off-resonant 2π pulse.

As is seen in figure 2-19, even at 30 times the value of E_{crit} for the case of a 1 psec pulse the contribution to the excited state population due to off-resonant excitation is less than 1%. This proves that this will not be a problem in interpretation of our results.

II.K Variation in Probability with respect to b and T

Since the experiments are to be done in a vapor cell where there is no control over either the time of closest approach between colliding atoms (T) or the impact parameter of the collisions (b),

one must average the results over all the possible values of these two variables. In order to find the range that must be integrated over, numerical solutions were made with different values of both b , the impact parameter, and T , the difference in time between the point of closest approach between the two atoms and the peak of the applied laser pulse.

The variation in excitation with changing impact parameter is graphed in figure 2-22. Figure 2-22a is the dependence for 100 psec pump pulses whereas figure 2-22b is the dependence for 1 psec pulses. For both examples the peak of the laser pulse was timed to be at the point of closest approach of the colliding atoms. In the "long" pulse case the intensity is below the value of E_{crit} and there is a great deal of oscillation due to the coherence between the two curve crossings. Obviously there is still a large excitation probability even at the small impact parameters which must be taken into account. The "short" pulse case does not show this oscillation since only one of the two curve crossings can be addressed (Note that there is a change in the horizontal scale between the two curves). With the laser pulse temporally centered over the point of closest approach of the atoms, the only curve crossings that can occur are those when the atomic trajectory is approximately tangent to the circle of radius R_c . In this case the value of R_c is $14.9 \text{ \AA}$. The excitation probability extends all the way to $18 \text{ \AA}$ before it decays away, indicating that we can indeed obtain excited population without the benefit of a curve crossing as predicted by Bates.¹³ These curves indicate that one must take into account impact

Figure 2-22 Variation of excitation probability with different impact parameters. Graph a.) illustrates the dependence for "long" laser pulses while graph b.) illustrates the dependence for "short" laser pulses. In both cases the time difference between the point of closest approach and the peak of the laser pulse was set to zero.

parameters out to $18 \text{ \AA}$ when averaging over impact parameter.

The dependence on T , the difference in time between the peak of the laser pulse and the point of closest approach between the two atoms, is plotted in figure 2-23. In this case R_c was again $14.9 \text{ \AA}$, the impact parameter was $10 \text{ \AA}$, and the applied E field was set at the value for E_{crit} . In figure 2-23a a 10 psec pulse was used and the two curve crossings are indistinguishable since the laser pulse overlaps both of them. Figure 2-23b a 1 psec pulse was used and the two curve crossing points are well separated from each other. These two curves indicate that one must average over a time which is the greater of four times the pulse duration (FWHM) of the laser pulse or the time that it takes the atoms to move apart sufficiently that the detuning is large enough that there is no chance of excitation. Inspection of these two curves indicates that this occurs at an interatomic separation of $40 \text{ \AA}$ for the case of Na-Na collisions.

The excitation probability as a function of these two parameters has been plotted in the form of a contour plot. This is shown in figure 2-24. The electric field was held at the value for E_{crit} and the pulse duration is 1 psec. One can clearly see the circle of radius R_c mapped out due to the increased time delay necessary for the laser pulse to overlap the curve crossing when there is an impact parameter less than R_c . It is interesting to note that there is a greater probability, as seen by the increasing contours, near the point where the perturbing atom's trajectory makes it near tangent to the circle of radius R_c . The perturbing atom spends a longer time in the curve crossing region when it is nearly

Figure 2-23 Variation of excitation probability with different times between the peak of the laser pulse and the point of closest approach. Graph a.) illustrates the dependance for "long" laser pulses while graph b.) illustrates the dependance for "short" laser pulses. In both cases the impact parameter was set to $10\text{\AA}$ and R_c was set to $15\text{\AA}$.

Figure 2-24 Contour plot of the excitation probability for "short" laser pulses versus different values of both the time of closest approach and impact parameter. The contour lines are at 10% differences from the peak excitation probability.

tangent. This indicates that by spending a longer time in the curve crossing region one can excite with a higher probability.

II.L Variation in Probability with Respect to Velocity

The velocity dependence of the picosecond optical collision dynamics was tested by varying the velocity used in the numerical solutions. The result is plotted in figure 2-25 for a 1 psec input pulse at an electric field strength of E_{crit} . Indications made by equation 2-6 in the Landau-Zener model were that lower velocities would lead to a higher excitation probability than higher velocities. This is indeed the case. At higher velocities the time spent in the curve crossing region is less contributing to a lower probability, as was indicated in the previous section. At extremely low velocities, however, the atoms stay in the curve crossing region so long that transitions can be made between the potentials twice during the same curve crossing. This is the reason for the drop at small velocities. When the Maxwell-Boltzmann velocity distribution is taken into account the probability is peaked at near the mean velocity even with this velocity dependent excitation probability.

Although there is obviously a dependence on the velocity in the numerical solution, it was not averaged over to perform the final results. The reasons for this are twofold: first, one of the three experimental variables (impact parameter, time between the point of

Figure 2-25 Variation in the excitation probability with velocity. The line to the x-axis indicates the mean value of the velocity and the velocity used in the "fully" averaged numerical solutions.

closest approach and peak of the laser pulse, and velocity) had to be eliminated due to computer limitations. Velocity dependence was obviously not as strong as the other two. Second, the mean velocity used is indeed, even with the observed velocity dependence, a reasonable average result over the possible velocities. Little is lost in understanding the dynamics or in estimating the signal by making the assumption that the results could be predicted using a single value for the velocity.

II.M Averaging Method for b and T

In order to average over the different values of b and T, probabilities must be calculated not only for the excitation probability given these input parameters as illustrated in a previous section, but also the probability that these values for b and T will indeed occur. The geometrical arrangement is shown in figure 2-26. Two atoms are colliding and we again assume that one is stationary and the other is moving along the z axis. Since everything is spherically symmetric from any orientation we can transform into this cylindrical geometry. The two atoms are separated, at one particular instant in time, by an interatomic separation R with the velocity an angle θ from the interatomic bisector. The number of atoms at a distance R is given by:

$$N_A(R) = N_p 4\pi R^2 dR \quad (2-30)$$

Figure 2-26 Geometry of the collision indicating the impact parameter b and the time between the peak of the laser pulse and the point of closest approach T .

where N_p is the number of perturbing atoms. The geometry of the collision is illustrated in figure 2-26. All possible collision orientations can be transformed to this coordinate system. Given this formalism, the number going in a direction θ can be shown to be:

$$N(\theta) \propto \sin(\theta) \quad (2-31)$$

where θ ranges from 0 to $\pi/2$. So, for one stationary target atom, the number at a distance R with a velocity at an angle θ , with orientation ϕ is:

$$N(R, \theta, \phi) = N_p 4\pi R^2 \sin\theta dR d\theta d\phi \quad (2-32)$$

The density of collisions with these specific parameters is:

$$N_F(R, \theta, \phi) = N_p N_A 4\pi R^2 \sin\theta dR d\theta d\phi \quad (2-33)$$

We can now make use of an equivalence formula of infinitesimal volume elements in three coordinate systems: cartesian, spherical, and cylindrical. This formula is given as:

$$dx dy dz = R^2 \sin\theta dR d\theta d\phi = r dr dz d\phi \quad (2-34)$$

and is graphically illustrated in figure 2-27. It is important to note that R does not equal r . A simple substitution can be made

Figure 2-27 Spherical and cylindrical coordinate systems used in the fully averaged numerical solution.

using the infinitesimal formulae transforming equation 2-33 from spherical to cylindrical coordinates. The result is:

$$N(r,z,\phi) = N_p N_A r dr dz d\phi \quad (2-35)$$

By observation of the cylindrical coordinate diagram in figure 2-27 we can determine that r is what we normally call the impact parameter and designate it b . Z is the distance from closest approach which we call s . If the perturber atom is at the origin when the peak of the laser pulse occurs then we can see that s is simply given as:

$$s = vT \quad (2-36)$$

where T is the time between the point of closest approach and the peak of the applied electric field. Since the atomic collision is cylindrically symmetric, one can integrate over ϕ giving:

$$\int_0^{2\pi} d\phi = 2\pi \quad (2-37)$$

Equation 2-35 can therefore be rewritten as:

$$N(b,s) = N_p N_A \delta\pi^2 b db ds \quad (2-38)$$

We then define $P_{ex}(E, t_p, \Delta, b, s)$ as the probability of being excited if a laser pulse detuned by Δ , with a peak electric field strength of E and a pulse width t_p , arrives at a time T (where T is

related to s by equation 2-36) from the point of closest approach to an atomic collision with impact parameter b . Using this definition we obtain the integrated result:

$$N_{ex}(E, \tau_p, \Delta) = 8\pi^2 N_p N_A \int_0^{b_{max}} b \int_{-s_{max}}^{s_{max}} P_{ex}(E, \tau_p, \Delta, b, s) db ds \quad (2-38)$$

where $N_{ex}(E, \tau_p, \Delta)$ is the number excited per unit volume at a given detuning, electric field strength and pulsewidth. The value of P_{ex} is the result of a numerical solution to the modified Bloch equations for a particular impact parameter and time of closest approach. We then integrate over these results in order to obtain the final result for the number excited in a random distribution of atoms.

Preliminary runs were made to decide how many different values of b and T were needed to adequately integrate over the different ranges. In order to evaluate how many integration steps were necessary to integrate over b the value of T was set to 0. The result for 100 different values of b over a range from $b=2\text{\AA}$ to $b=18\text{\AA}$ is displayed in figure 2-28 for the "long" pulse case of Na-Na collisions. By averaging over the values of b the general shape is preserved however the Stuckelberg oscillation evident in figure 2-18 is washed out due to the varying time between curve crossings for the different impact parameters. Later runs indicated that 50 different values of b were sufficient to produce a result accurate to 5%. Although even 50 values of b do not follow exactly the fast oscillation evident in figure 2-22, the large number of values used

Figure 2-28 Numerical solution result for "long" laser pulses when averaging over different impact parameters.

and the varying oscillation frequency ensure that there is no consistent bias of the position of the chosen values of b with respect to the phase of the oscillations. Therefore this is an accurate integration over the different possible values of b .

To test the number of values needed to integrate over T , the value of b was set to be $10\text{\AA}$. The result for an integration of 10 different values of T from 0 to 2 times the pulse FWHM for the "long" pulse case in Na-Na collisions is displayed in figure 2-29. Further tests at different values of b and E indicated that 20 values of T were in fact needed to adequately integrate over T . In this case the general shape is considerably different than that of figure 2-18 for the single trajectory case. At high laser E fields the excitation probability does not decay to zero as is the case for infinite pulse durations, but rather decays to only half of the peak magnitude. The reason for this is displayed graphically in figure 2-30. Even at the highest E fields where the excitation probability is reduced in the "long" pulse case, there are two times during the pulse duration where the E field is indeed equal to E_{crit} . Therefore the population is generated not at the peak of the "long" pulse but rather by collisions which occur on the temporal wings of the pulse where the E field happens to be at E_{crit} . This can be easily seen in the contour plot in figure 2-31 where excitation is plotted versus both time delay from the peak of the laser pulse to the point of closest approach and the peak applied E field. The excitation does indeed occur on the wings of the pulse when the peak E field is made greater than E_{crit} . This particular feature is present when

Figure 2-29 Numerical solution results for "long" laser pulses when averaging over different values of T the time to closest approach.

Figure 2-30 Graphical demonstration of the effect of realistic pulseon probability at high laser intensities when one averages over a realistic pulse shape.

Figure 2-31 Contour plot of the excitation probability versus the time of closest approach and the peak electric field strength for "long" laser pulses. This plot indicates clearly that at the high laser intensities the excitation probability comes not from the peak of the pulse but rather in the wings where E is approximately equal to E_{crit} . The contour lines are at intervals equal to 10% of the peak value.

averaging over the real laser temporal pulse shape and reduces the sizeable difference between "short" and "long" pulses discussed by Lee and George¹⁶

II.N Averaged Results of Numerical Solution

In order to calculate the final averaged results of the numerical solution to the modified Bloch equations, 1000 individual solutions to the differential equations had to be made for each value of the E field tested. In most runs only ten values of E field were calculated, however this required up to 40000 seconds of CPU time on the Cyber 170 which corresponds to nearly a week of time waiting for the result. This is the reason that so much care was taken to optimize the speed of the solution to the differential equation.

The result average over b and T for the case of Na-Na collisions is presented in figure 2-32. The number density of Na atoms was set at 10^{16} cm^{-3} and runs were done for both 2 psec pulses ("short") and 100 psec pulses ("long"). The number excited is calculated to be much greater for the 100 psec pulse than the 2 psec pulse. This is because at the same electric field strength the 100 psec pulse has 50 times more photons than the 2 psec pulse. In order to properly compare the excitation probability for these two curves the results were divided by the pulsewidth to take out this bias. The result is shown in figure 2-33. As predicted the "short" pulse does have a higher excitation cross section than a "long" pulse at

Figure 2-32 "Fully" averaged result for Na-Na collisions. The dependence of the excited state population on the peak applied electric field strength is plotted for 100 psec pulses and 2 psec pulses.

Figure 2-33 "Fully" averaged result for Na-Na collision. The excited state population is divided by the applied electric field pulse width to eliminate the bias caused by the greater number of photons in the 100 psec pulse at the same peak electric field strength as the 2 psec pulse. This plot indicates that even after averaging over the impact parameter and the time to closest approach the difference in excitation efficiency for "short" versus "long" laser pulses is evident.

high intensities, although the ratio is not as great as that predicted by Lee and George¹⁶ and illustrated in figure 2-11. The reason for this was clearly shown in the previous section as being primarily due to the realistic pulse shape used.

Similar runs were made for Na-Ar collisions. The differences in the code were simply a change in the potential shape as a function of R (R^{-6} instead of R^{-3}), changes in the range of impact parameters required to integrate over, and specifying the number of Na (target) atoms to be 10^{14} cm^{-3} and the number of Ar (perturber) atoms to be $2 \times 10^{18} \text{ cm}^{-3}$. The result divided by the pulsewidth is shown in figure 2-34 for 1.6 psec pulses and 10 psec pulses. As in the case of Na+Na collisions, the "short" pulse case has a larger cross section at high intensities than the "long" pulse case.

The difference between the two at high electric fields is not as great in the case of Na+Ar collisions as it is in the case of Na+Na collisions. This is because the value of R_c for a detuning of $20 \text{ \AA}$ is $6 \text{ \AA}$ for the Ar perturber and $15 \text{ \AA}$ for the Na perturber (due to the stronger interaction of resonance collisions). This translates into a longer time between curve crossings in the Na case. A 2 psec pulse is much shorter than the time between curve crossings for the resonance collision case (Na), however a 1.6 psec pulse is close to the time between curve crossings for the van der Waals collision case (Ar). A 1.6 psec pulse is not, strictly speaking, a perfect "short" pulse. Shorter pulses were not used in the theory, however, because the input to the theory was driven by

Figure 2-34 "Fully" averaged result for Na-Ar collision. The excited state population is divided by the applied electric field pulse width to eliminate the bias caused by the greater number of photons in the "long" laser pulse. A 1.6 psec pulse was used to model the "short" laser pulse while a 10 psec pulse was used to model the "long" laser pulse. The choice of pulse widths is meant to model more closely the actual experimental conditions.

the experimental developments. 1.6 psec pulses were experimentally easy to generate at the proper wavelength and did not overlap spectrally with the resonance line for either 15 or 20 Å detunings. Shorter pulse durations made the results somewhat questionable as to whether the signal was due to direct line excitation (due to the broad laser bandwidth as shown in section II.J) or due to the optical collision dynamics.

II.0 Spatial Averaging

Experimentally it is extremely difficult to generate a laser pulse whose spatial profile is square or uniform across the diameter of the beam. Gaussian shaped output beams are usually generated by lasers since they provide the most stable diffraction limited operation. Altering the shape after it has been generated is very lossy. Therefore, in practice one must use a beam with a spatially varying intensity. In experiments such as those described in this work this does present problems since the desired result is strongly intensity dependent.

In order to model the result the realistic spatial beam profile will have on the experimental results, a further computer program to average over a gaussian beam profile was developed. Solutions of the modified Bloch equation resulted in a variation in excitation number for different values of peak applied E field. If one has a gaussian beam profile then there are annuluses, each of

constant E field radially outward from the center of the beam. Since one can determine, from the numerical solutions previously obtained, the excitation number for each of these E field values, the total number can be calculated. This calculation was made for the case of Na-Ar collisions and the result is plotted in figure 2-35. In addition, since experimentalists usually measure intensity rather than E field, the horizontal axis is in units of W/cm^2 . As one can see the large difference between the two cases of "long" and "short" pulses is further decreased by the effects of spatial averaging. The dotted curve on this graph is the result if one uses a 10 psec adiabatic square pulse and does not include the effects of spatial averaging. This dramatically shows the consequences of using realistic temporal and spatial beam profiles. Three dimensions of gaussians (time, and x and y in space) all contribute to diminish the difference between "short" and "long" pulses.

II.P Theoretical Conclusions

Calculations based on the new theory using modified Bloch equations obtain results similar to those of both Landau and Zener and Lee and George. Averaging over realistic values of both the temporal and spatial beam parameters, however, has a dramatic effect on the results. This is to be expected since a varying intensity beam is being used to probe an extremely non-linear effect. The results do show, however, that the effect can be seen if the

Figure 2-35 Spatially averaged result for Na-Ar collision. The x-axis is plotted in intensity rather than electric field strength in order to make it more comparable with the experimental results. After fully averaging the numerical solution the difference between the "long" and "short" laser pulses is not quite as dramatic. For reference the result without averaging over the realistic temporal or spatial profile for the "long" pulse case is plotted as a dotted line.

experiment is done with sufficient signal-to-noise ratio.

Assumptions made in developing this theory are reasonable barring one (assumption xvi) made about the assumption of a two-level atom. As discussed in section II.D, Light and Szoke indicated the need to include all excited-state levels when observing the variation in excitation efficiency with E field. The dramatic difference at high E field was shown to be diminished when four states are used instead of the unrealistic two (see figure 2-6). Coupled with the effects of both temporal and spatial averaging which also tend to reduce the difference (see figure 2-35) this could make the distinction between "short" and "long" pulses only observable at extremely high laser E fields.

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CHAPTER III

EXPERIMENTAL DEVELOPMENT

In any experimental thesis the great majority of time spent is involved in building, developing, optimizing, and understanding the operation of the apparatus used to perform the experiment. This is particularly true when the system developed and used is considered "state of the art" and developments made today always point to the next change which could give better stability, energy, or temporal performance. After this is fully completed, however, the experiment can be performed quickly and accurately with high statistical significance. The experiments reported in this dissertation could not have been done, as will be demonstrated, without the superb flexibility and operation of the "state of the art" laser system and data acquisition system used. These results seem to corroborate the above experimental philosophy of fully understanding and optimizing the experimental apparatus before performing the actual experiments.

During the course of this work, several major developments have been made in laser science which have not only contributed to the operation of the laser system used, but have also made significant contributions to the advancement of laser technology and

in particular ultra-short-laser pulse technology. The first result and cornerstone for the rest of the work was the demonstration of the pumping of an ultra-short pulse dye laser with the output of a frequency-doubled CW mode-locked Nd:YAG laser.¹ The Nd:YAG pumped dye oscillator has undergone a great deal of development resulting in a source of sub-100 femtosecond (10^{-15} sec) pulses with the good stability and temporal pulse shapes desirable for amplification. Using a Nd:YAG laser as the pump laser not only allowed the use of a more stable and rugged source, as compared to the normal mode-locked argon laser, but also provided an easier means of short pulse amplification. Synchronous amplification, as we have named this process, uses a relatively short amplification pulse highly synchronized to the weak ultrashort pulse to be amplified.² The advantages of this system are its efficiency, lack of ASE (amplified spontaneous emission), stability, and superb beam quality, all resulting in an intense, clean, highly focussable beam for experiments. A weak link in the amplification process was the use of krytron drivers used in Pockels cell switchouts and as a result a new type of switchout was developed based on a microwave triode.³ This new triode design offers an extremely long lifetime with very little electrical jitter or drift. Not only was this Pockels cell used as a switchout but it has also been placed inside a laser cavity to act as a Q-switcher and cavity-dumper providing a much more stable and intense pulse at high repetition rates for short pulse amplification.⁴

In this chapter each of these advancements will be described

as well as other experimental apparatus which was used in conjunction with the dissertation experiments. These not only include the laser system but also the construction and characterization of two sodium cells and a high-speed computerized data acquisition system.

III A. WHY A Nd:YAG BASED SYSTEM?

The standard method used for generating sub-picosecond dye laser pulses at the time that this work began was to use an argon laser to pump a dye oscillator. This oscillator was mode-locked either passively using a CW argon⁵ (passive modelocking) or actively using a mode-locked argon⁶ (synchronous pumping), both of which provided picosecond and sub-picosecond dye laser pulses. Each of the two methods had their advantages and disadvantages. Passive modelocking produced relatively stable short pulses but was extremely dependent on the quality of the modelocking dye used and could be operated at only one wavelength. Synchronous pumping did not use a modelocking dye and therefore provided tunability but did not give as short a pulse width and was critically reliant on the dye oscillator cavity length. Both of these lasers suffered from the same problem: they both were pumped using an argon laser. Argon lasers do provide an intense beam at 5145 Å which matches somewhat the absorption band of Rhodamine 6G, the dye used in all of the sub-picosecond dye oscillators at the time. Where these lasers suffer is in their stability, ruggedness, mode-locked pulse duration, mode-locked pulse

energy, and day to day pointing stability. In addition argon tubes do not last forever and to replace the tube (approximately every 24 months) not only takes a great deal of valuable laser time but also costs on the order of \$10,000, resulting in an extreme liability on even the most adequately funded research programs.

In order to amplify the ultra-short dye pulses a Q-switched Nd:YAG laser was usually used by others as the amplification pump source which provided a 20 nanosecond pulse at a lasing wavelength of 532 nm with between 100 and 500 millijoules of energy.⁷ Q-switched Nd:YAG lasers can be made to oscillate in a single longitudinal mode, however this is experimentally quite difficult and in practice was rarely done. As a result the multimode behavior gave rise to severe amplitude stability problems over the length of the Q-switched pulse. This pulse was then used to transversely pump a series of dye amplifier stages. The amplifier stages were filled with various types of organic dyes and absorbed the green energy resulting in a population inversion in the stages. The short, weak, ultrashort dye pulse then came through each amplifier stage and by stimulated emission transferred some of this stored energy into the ultrashort pulse. As we will show, the storage time in the dye amplifier is dependent on the gain in the amplifier: the higher the gain, the shorter the storage time due to effects of ASE. Since this system was used in high gain, the storage time was approximately 150 psec. Therefore, the population in the amplifier stage had no memory beyond 150 psec and was cycled more than 100 times over the course of the Q-switched pump pulse. Because of the multimode behavior of the

Q-switched laser, the lack of synchronization between the pump and ultrashort pulses, and the short storage time in the amplification stage, this resulted in an extremely unstable amplifier when run in the linear regime (operation in the saturated regime of the amplifier is discouraged due to temporal pulse broadening effects on the ultrashort laser pulse). In addition, the cycling of the population merely increased the amount of ASE generated along the same beam line as the amplified ultrashort laser pulse, indicating the need for many saturable absorbers to eliminate this ASE.

In order to overcome many of these shortfalls in both the oscillator and amplifiers we developed a new technique of short pulse generation and amplification based solely on Nd:YAG technology. The advantages of Nd:YAG lasers are that they are rugged and stable with a shorter mode-locked pulse duration. In addition the maintenance costs for Nd:YAG lasers are far less than argon lasers, with only water, filters, and lamps to change with a laser down time for each taking only about 30 minutes. By synchronizing the output of a pulsed Nd:YAG laser with that of a CW mode-locked Nd:YAG laser with an accuracy of less than 40 psec and using the output of the pulsed Nd:YAG to pump a dye amplifier chain, one can synchronously amplify an ultrashort dye laser pulse efficiently and with a low background of amplified spontaneous emission.

III.B CW MODE-LOCKED Nd:YAG LASER

Before the advent of this work the CW pumped Nd:YAG laser had been used primarily for cutting, welding, and scribing metal and semiconductor surfaces when used in the Q-switched mode. Very little work had been done in optimizing this laser for use in a scientific setting. Many modifications had to be made in the laser to make it the valuable stable source it is today.

A diagram of the current configuration of the CW pumped Nd:YAG laser⁸ is given in figure 3-1. The laser rod is pumped by a CW krypton arc lamp. The stability of the current used to pump this lamp is therefore critical to the stable operation of the laser. The method used to provide this current is to rectify and integrate the three phases of a 208 three phase service and filter out the 60 Hz and 360 Hz components using a high voltage high current low pass filter. As this was not sufficient, we added a second LC low pass filter⁹ to further filter out undesired noise. The output stability of the lamps when the SCR's used to rectify the phases are balanced and the extra filtering is installed has been measured to be $< .040 \%$. The lamp and laser rod are water cooled. In order to reduce the water pressure fluctuations due to the pump vibrations, soft walled tubing was used to replace the hard tubing between the laser power supply and the laser head. This is meant to absorb the pump vibrations, preventing them from reaching the laser head and producing an unstable water flow. There is currently some question as to whether this is really necessary since there is turbulent flow

Figure 3-1 Layout and adaptations made to the CW pumped actively modelocked Nd:YAG laser.

used across the lamp and the rod in order to increase the cooling efficiency.¹⁰

In order to modelock the Nd:YAG laser further modifications had to be made to preserve a good modelocking condition. A requisite for good modelocking is that there be a perfect match between the frequency used to drive the modelocking crystal and the frequency of the laser cavity given by $c/2L$ where c is the speed of light and L is the optical path length of the laser cavity. In the past a variable frequency oscillator was used to drive the modelocking crystal and was tuned to match the frequency given by the length of the cavity. Since the laser cavity was set on an aluminum rail, and aluminum has a rather large thermal expansion coefficient ($23 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$), the frequency needed adjusting any time the ambient temperature changed. In practice this meant that a great deal of time was spent "chasing" the length of the cavity by adjusting the modelocking frequency. Modifications were therefore made to ensure that the cavity length changed as little as possible. These included remounting all of the components (and in particular the end mirrors) on a rail of Super-Invar which has a thermal expansion coefficient 1000 times less than that of aluminum ($.03 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$). Cooling for the quartz modelocking crystal was, in the past, provided using the same water flow that was used to cool the lamp and rod. This had three severe disadvantages. The first was that the RF electronics could not be left on at all times in order to stabilize the oscillator and amplifier networks since the laser cooling, and therefore the modelocker cooling, was turned off at night. The second is that when

the RF was turned on it took time for the crystal to come to thermal equilibrium. This played havoc with the modelocking efficiency since the resonance frequency of the quartz modelocking crystal is temperature dependent. The third disadvantage was that the cavity length of the laser changed over a one to two hour period since as the modelocking crystal warmed up its temperature changed and the effective optical length of the cavity changed as well due to the temperature dependent index and expansion of the quartz modelocking crystal. As a result, it took nearly two hours for the laser to fully warm up in the morning. For all of these reasons the cooling supplied was disabled and replaced with a separate cooling water unit which could be run at all times. This allowed the RF to be run all the time, and this, in turn, solved all three of the disadvantages. The warm-up time was reduced to about 20 minutes after this change was made due to the stabilizing of the temperature of the laser rod and lamp output. There is some indication that the intracavity laser beam heats up the internal components of the cavity due to absorption of the $1.06\text{ }\mu\text{m}$ radiation in the mirrors and mirror coatings, modelocking crystal, and brewster plate. Due to the index change and thermal expansion in these regions, the cavity length is modified further. The warm up procedure is therefore to turn on the lamps and let the laser warm up while lasing.

One of the end mirrors of the laser cavity was placed on a precision linear translation stage. This allowed us the ability to match the cavity length to the modelocking frequency rather than the modelocking frequency to the cavity length as before. It is much

easier to make an extremely stable single frequency source than a stable variable frequency source.¹³ The stability required when pumping an ultrashort dye laser with these pulses is that the modelocking frequency be stable to $1:10^7$ or on the order of 1 Hz out of the 49.9 MHz used to drive the crystal. For this reason we replaced the variable frequency RF oscillator with a single frequency one and matched the cavity by adjusting the position of one end mirror.

The other end mirror in the cavity was placed physically as close as possible to the modelocking crystal. Because the modelocker is driven with a sinusoidal RF there are two points in time when the RF passes through a zero point. This results in two points in time when the modelocker presents no diffraction loss to the laser cavity (this is the reason why the output repetition frequency is twice that used to drive the modelocker). Since the pulse which is reflected back and forth in the laser cavity must travel through the modelocking crystal, off the end mirror and then through the modelocker again, the time between these two passes through the crystal should be minimized in order to reduce the loss applied to the intracavity pulse. This loss is illustrated in figure 3-2. In addition, when there is time-dependent loss such as this in the cavity, the laser searches for the point of least loss, and this searching leads to a phase jitter of the output pulses with respect in the input RF. Minimizing this parameter t_1 we believe should reduce the jitter since a change in phase with respect to the RF will add loss to both passes through the modelocker as illustrated by the

Figure 3-2 Cavity loss due to finite distance between the laser end mirror and the modelocker. The solid curve represents the time-dependent loss in the cavity applied by the acousto-optic modulator. The time t_1 is the time it takes the pulse in the cavity to travel from the modelocker to the mirror and back to the modelocker. The larger the value of t_1 (i.e. the greater the distance from the modelocker to the mirror) the larger loss that is inserted into the cavity.

pulses in figure 3-2.

The output from the Nd:YAG laser⁸ is 10 Watts in a TEM₀₀ beam with 90 picosecond pulses at a repetition rate of 100 MHz. The lasing wavelength is 1.064 μm . The amplitude stability of the laser has been improved by the modifications of the laser cavity and is currently stable to <1% pulse to pulse. Six watts of RF power are absorbed by the modelocking crystal providing the short, 90 psec pulses. Increasing this power to over 10 watts decreases the pulse width to as short as 50 psec due to the increased modulation depth in the crystal, however, at a decrease in amplitude stability. This decrease in pulse width with increasing RF power absorbed is illustrated in figure 3-3. We believe that the tradeoff between these two should be towards a more stable output pulse since this instability will propagate non-linearly throughout the dye oscillator and amplifier.

In order to pump a visible dye laser, the infrared beam must be frequency doubled to the green in order to match the absorption profiles of the dyes used. The requirements that are placed on the non-linear crystal are severe. It must have a very low absorption at both 1.06 μm and .532 μm wavelengths since there is a large average power passing through the crystal. It must have a reasonably high damage threshold since these pulses are short and when focussed have a power density of >10 MW/cm². Finally, it must have a high non-linear coefficient for efficient doubling of the 1.06 μm radiation. At the time this work was started, there was only one crystal that satisfied all of these criteria, barium sodium niobate

5W

6W

7W

8W

9W

10W

Figure 3-3 CW autocorrelations of the Nd:YAG laser output for different input RF powers to the modelocker. The time scale per division is 35 psec. The pulsewidth, if one assumes a Gaussian pulse shape, varies from 90 psec for the 5W case to 70 psec for the 10W case.

($\text{Ba}_2\text{NaNb}_5\text{O}_{15}$). This crystal has one of the highest known non-linear coefficients ($1.46 \times 10^{-11} \text{ m/V}$)¹⁴ and has very low absorption ($.002 \text{ cm}^{-1}$ for $1.06 \mu\text{m}$ and $.04 \text{ cm}^{-1}$ for $.532 \mu\text{m}$).¹⁵ Also, this crystal can be cut for 90° phase matching which, even in the focussing condition, allows generation of an undistorted second harmonic beam profile essential for optimum dye laser pumping. A 10 cm focal length lens is used to focus the infrared beam in the temperature stabilized barium sodium niobate frequency-doubling crystal. With an 8 watt input, the output power in the green was approximately 500 mW, indicating a conversion efficiency of $>6\%$. The crystal has a length of 5 mm and must be maintained at 105°C ($\pm 0.5^\circ\text{C}$). Due to self-absorption the crystal experiences some self-heating and a lower phase matching temperature must be set. This absorption gives rise to a peculiar shaped doubling curve which is shown in figure 3-4. The $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ has a low damage threshold reported to be 25 MW/cm^2 .¹⁶ The peak power is 10 MW/cm^2 when focussed on the crystal with an f-number of 50 and as such, great care must be taken to maintain the amplitude stability of the Nd:YAG laser to avoid damaging the doubling crystal.

This focussing condition corresponds to a confocal beam parameter of 50 mm, far greater than the 1.7 mm required for optimum conversion efficiency. Damage considerations make this choice inevitable.

Nd:YAG lasers have been frequency doubled in the past, using an intra-cavity doubling crystal.¹⁷ The choice to frequency double external to the Nd:YAG cavity was made for two reasons: first, to

Figure 3-4 Doubled output power versus crystal temperature. The sharp fall-off at low values of the temperature indicates the presence of self-heating (due to absorption of the frequency-doubled .53 μm light). At low input laser powers the shape of the output power versus temperature curve does not have this sharp feature, is somewhat symmetric, and is centered at 105.0 °C.

limit the number of elements inside the Nd:YAG cavity for ease in stabilization and secondly, for pulse width considerations. The crystal, when placed inside a laser cavity, not only acts like an etalon to reduce the intracavity bandwidth, but also doubles the short, intense pulses better than the longer pulses. As a result, the point of least loss in the laser cavity is not at the shortest pulses as before, but at longer duration pulses. At these conversion efficiencies, the pulse width of the doubled output is shortened in an external configuration. The phase matching bandwidth of $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ has been calculated to be $6 \text{ \AA}$, whereas the laser bandwidth is $0.3 \text{ \AA}$, so a green output reduced by $\sqrt{2}$ is expected.

In late 1982 a new crystal was developed called KTP (KTiOPO_4) which offered several advantages over $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$. The non-linear coefficient was as high as $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ however, the absorption at $1.06 \mu\text{m}$ and $.532 \mu\text{m}$ was less, the damage threshold was ten times higher, and it was an angle tuned crystal rather than temperature tuned.¹⁸ All of these advantages gave far superior output characteristics as well as a tremendous ease in the operation of the frequency-doubler. With 10 Watts at $1.06 \mu\text{m}$ input into the KTP frequency doubler one obtains up to 2.2 Watts of light at $.532 \mu\text{m}$. The higher efficiency ($>20\%$) is obtained because the higher damage threshold, low thermal absorption, and large angular acceptance angle allow higher input intensities to be applied to the crystal. In a quadratic doubling regime such as this, the higher the input intensity, the higher the efficiency. Other advantages to this crystal are that it is non-hydroscopic and is chemically and thermally stable allowing for little worry about crystal degradation

due to the presence of water vapor in the air as is a great worry with KDP and CDA crystals.

Because of the obvious advantages in using KTP, the use of $\text{Ba}_2\text{NaNb}_5\text{O}_{15}$ was discontinued by us in 1983. Barium sodium niobate does, however, continue to be a good candidate for frequency doubling Nd:YAG lasers albeit with deficiencies as compared with KTP.

III.C SUBPICOSECOND CW MODE-LOCKED DYE OSCILLATOR

As mentioned at the beginning of this chapter, during the course of this thesis research the first subpicosecond dye laser oscillator was developed using a frequency-doubled Nd:YAG laser as the pump source. The advantages of this pumping source have been outlined earlier. A tremendous amount of work has been done in redesigning, optimizing, and understanding the operation of the CW mode-locked dye laser over the past 5 years -- enough to fill an entire thesis! For the benefit of the reader, not all of the changes and experiments done on the dye oscillator will be recorded. Only the first, an intermediate, and the most recent dye laser configurations will be described.

III.C.1 THREE-MIRROR DYE OSCILLATOR

The first dye laser configuration is illustrated in figure 3-5.¹ The dye laser cavity is of the folded astigmatically compensated type¹⁹ with a cavity length equal to half of the cavity length of the Nd:YAG laser. The dye laser cavity was set at half the cavity length for a more stable cavity configuration and a lower threshold pump power. However, as every other pulse passes through the dye jet, it sees a loss rather than gain. This gives rise to an alternating peak pulse height, however the dye tends to also act as a saturable absorber to further reduce the pulse width on every other pass. The cavity was mounted on a low thermal expansion super invar slab in minimize cavity length fluctuations. A piezo-ceramic crystal is mounted on one end mirror for fine tuning the dye laser cavity. The dye laser is free of any bandwidth limiting elements to assure the shortest possible pulses. An output coupler transmission of 5% was used, giving an average output power of 30 mW.

Measurements of the pulse widths of both the Nd:YAG laser and the dye laser were made using the zero-background autocorrelation technique.^{20,21} Two non-collinear beams are intersected in non-linear crystals (KDP or LiO₃) to give a zero-background signal as one of the arms of the Michelson interferometer is scanned. A linear potentiometer is attached to the changing arm and the output is sent to an oscilloscope in order to have a real-time knowledge of the pulsewidth. This feature is particularly useful when optimizing the laser for minimum pulsewidth.

Short-pulse operation has been achieved using this system. Pulse widths are consistently shorter than 2 psec. A typical autocorrelation scan of the dye laser pulse is shown in figure 3-6. The output amplitude and pulse width jitter of the dye laser is, as expected, strongly related to the stability of the Nd:YAG laser. A slight amplitude instability of the Nd:YAG laser is amplified due to the cumulative effect of the frequency doubling and the dye laser running at threshold. Stability is also a strong function of the modelocking efficiency when the laser pulse width is in the subpicosecond regime. Dye laser cavity length detunings of as little as $5\text{ }\mu\text{m}$ have resulted in an unstable dye laser output as well as longer pulse widths. This is in agreement with references 22,23.

Cavity length fluctuations of the dye laser were thought to be the major cause of instability limiting the pulse width of the dye laser. Cavity length fluctuations are primarily due to a 200 Hz variation of $.2\text{ }\mu\text{m}$ in the dye jet. A single longitudinal mode helium neon laser was injected into the dye laser cavity and Fabry-Perot fringes were formed as a result of reflections off the two end mirrors. By locking onto one of the fringes and feeding back an error signal to the piezo-ceramic crystal, this cavity length fluctuation was significantly reduced. This feedback did not, however, produce any shorter pulse widths from the dye laser. As there were no bandwidth limiting elements in the cavity this negligible improvement in pulse width with cavity length stabilization implies either that other instabilities in our system prevent us from obtaining shorter laser pulses or that the feedback

Figure 3-6 Autocorrelation trace of the dye oscillator output at a laser wavelength of 5810 Å. The pulsewidth of 453 fsec assumes a sech^2 pulse shape.

circuit increased the cavity length fluctuations at a different frequency.

Amplitude instabilities of the pump laser can give rise to pulse width broadening of the dye oscillator, indicating a need to use the most stable pump source possible. The frequency-doubled Nd:YAG laser can be made to run with a more stable output and a shorter pulse than that of the argon laser. As illustrated in figure 3-7, a slight variation in the amplitude of the pump pulse causes a time or phase shift of the dye laser pulse with respect to the pump pulse. When one is synchronously pumping a dye oscillator, the dye pulse duration is affected by the result of thousands of passes through the dye jet. A phase jitter such as that proposed here obviously will broaden the pulse since it must reform itself in time as the pump pulse amplitude varies up and down. One way to reduce this jitter is to run very high above threshold so that the jitter is less for the same amount of amplitude variation. In pure actively mode-locked lasers, however, one cannot run very high above threshold because of the possibility of satellite pulses occurring. A plot (figure 3-8) has been made illustrating the size of this phase jitter as a function of the amplitude instability of the pump source. In this model a 70 psec pump pulse was used 10% above threshold. One can see that there is a significant temporal variation even for a 2% variation which is what was estimated for the stability of the 532 nm pump.

Cresyl-violet was added to the rhodamine 6G to act as a slight saturable absorber as well as extending the tuning range to 640 nm.

Figure 3-7 Temporal variation of the loss line crossing due to variation of the pump pulse energy. The two curves are integrals of Gaussians with a 10% difference in pulse energy. Since the dye oscillator pulse occurs where the pump pulse energy crosses the loss line, the difference in time where the curves cross the loss line illustrates the amount of time that the dye pulse must reshape itself over if a 10% energy fluctuation is present.

Figure 3-8 Variation of the time between the frequency doubled Nd:YAG laser pulse and dye laser pulse due to the fluctuation of the energy of the Nd:YAG laser pulse. The dye oscillator is assumed to be run 10% over threshold. A $\pm 1\%$ fluctuation implies a temporal variation of 3 psec.

The pass through the jet without the pump pulse present provides an opportunity to trim the pulse width on a previously useless pass. This saturable absorber effect produced pulses shorter than 400 fsec and was very instrumental in reducing satellite pulse formation. These pulses represented the shortest pulses obtained at the time using the synchronous pumping technique.

The significance of this oscillator is not in its superior pulse width or power output, as neither was significantly better than that available at the time this work was completed. Rather, the fact that a dye laser could be pumped by the output of a CW mode-locked Nd:YAG laser was the significant point since it provided a rugged and stable method for producing subpicosecond pulses with the added benefit of an easier and more efficient method of amplification.

III.C.2 RHODAMINE 6G - DQOCI DYE OSCILLATOR

The technique of adding a saturable absorber to the gain jet in order to reduce the pulse width was extended using the dye DQOCI.²⁴ This absorber has a very fast recovery time, measured using time-delay fluorimetry on a jitter-free streak camera²⁵ to be less than or equal to 3 psec. A fast recovery can be used to trim the back edge of the dye pulse while the saturable absorption is used to trim the front edge. DQOCI in ethylene glycol was chosen as the absorber, not only for its fast recovery, but also because its absorption band matches the rhodamine 6G emission band, leading to a

large intensity discrimination and maximum wavelength tuning range.

The new oscillator configuration is shown in figure 3-9. A Z cavity configuration was used to eliminate the production of a second pulse in the cavity which is unseen by us on both an oscilloscope and the autocorrelation but which has been observed by other experimenters.²⁶ This cavity did provide two outputs (which allowed one output to be used for experiments and the other to be used to monitor the stability and pulse width of the dye oscillator) and was not essential for the generation of short pulses. It should be noted that the jet did not occupy any strategic position in the cavity. The dye was a mixture of 5×10^{-4} M rhodamine 6G and 3×10^{-5} M DQCI. A 2 μ m uncoated pellicle tuned the laser wavelength without restricting the laser bandwidth.

Seventy-femtosecond pulses were obtained at 605 nm using this new dye mixture. Figure 3-10 shows a typical autocorrelation trace. The dye laser exhibits an optimum stability and maximum output power at minimum pulse widths, reflecting the dramatic effect of the saturable absorber on the operation of the laser. The integrated spectrum width is over 120 Å and suggests a frequency chirp on the pulse. In a test through dispersive media such as BK7 glass, we found that pulses starting from a different initial pulse width can be stretched or compressed according to the central frequency of the laser, suggesting a negative or positive frequency chirp across the frequency tuning range. The frequency chirp variation could stem from the interplay between the normal and anomalous dispersions in the solvent, rhodamine 6G, and DQCI dye mixture. At frequencies

Figure 3-9 Experimental layout of the hybridly modelocked CW dye oscillator.

Figure 3-10 Pulse autocorrelation of the output of the hybridly modelocked CW dye oscillator. If one assumes a sech^2 pulse shape then the pulsewidth is 70 fsec.

corresponding to the minimum pulse width the use of dispersive glass stretches the optical pulse and seems to indicate a positive frequency chirp. This indicates the need for a dispersion-free oscillator cavity to allow the pulse to form itself without the broadening effects of dispersion.

This oscillator configuration remains the one which provides the shortest pulses obtained directly out of a synchronously pumped laser. This configuration was abandoned, however, for three reasons. The first was that the supplier of the DQOCI failed to deliver a product with the same short dye lifetime. Time-delay fluorimetry measured dye lifetimes of 25 to 100 psec indicating a lack of adequate quality control in producing the dye. Extensive negotiations with our supplier failed to correct this problem. The second reason was the cost of the dye. At \$200 a gram extensive testing of different batches of DQOCI was impossible. In addition, there was some evidence that the dye aged over a period of 2-4 weeks (this is very similar to the more documented aging characteristics of the dye DODCI) resulting in need to change both the DQOCI and the rhodamine 6G since they are mixed together. Reoptimizing the concentrations for optimum pulse duration every two weeks limited our ability to have a stable, reliable source of ultrashort pulses as desired. The third reason was that the stability of the dye laser was extremely critical with respect to the cavity length of the dye oscillator and also with respect to the length of the Nd:YAG oscillator. We have found that the stability that one needs in a normal synchronously pumped cavity is 1/100th of the length of the

output pulse in space. For example, a 70 fsec pulse has an extent in space of $21\text{ }\mu\text{m}$ indicating a need for both cavities to be stable to $2100\text{ }\text{\AA}$. A new oscillator was needed to relieve this restriction on the cavity length stability.

III.C.3 SYNCHRONOUSLY PUMPED CPM LASER

In early 1981 Fork et al. reported²⁷ a new method of generating sub-picosecond pulses using a pure cw argon ion laser as the pump source. Two jets were used in their cavity, one for the gain and the other as the saturable absorber. By adjusting both the concentration of the dyes used and the thickness of the absorber jet, one could obtain 90 fsec pulses which were very stable in amplitude. There were two pulses in the cavity which collided in the saturable absorber jet, giving rise to the name of the laser, the colliding pulse mode-locked (CPM) laser. The colliding of the two pulses in the absorber medium tended to balance the amplitudes of the two counter-propagating pulses as well as cooperate in bleaching the saturable absorber completely to decrease the loss in the cavity. The most stable operation with the least loss in the cavity occurred when the two pulses collided in the saturable absorber jet. Recently, Valdmantis et al. have proposed²⁸ that the action of this laser is not due to this bleaching of the saturable absorber but to a balancing of the self-phase modulation, group velocity dispersion, saturable absorption, and saturable gain, which all cooperate to

provide a stable, ultrashort output. With either of the two explanations, the colliding pulse output is stable and short -- a feature that should be explored and exploited in all ultrashort laser configurations.

Fork et al.²⁹ in a different paper invented a prism sequence which, when placed inside a laser cavity, provides a method of adjusting the cavity through a point of zero group velocity dispersion irregardless of the other elements in the cavity. This solves the problem of dispersion broadening the dye laser pulse as it is being formed in the dye cavity, which we felt was in effect in the rhodamine 6G-DQOCI laser discussed previously.

In order to incorporate these two techniques into a synchronously pumped dye laser we had to redesign the laser cavity.³⁰ A necessary condition for a dye laser to be both synchronously pumped and CPM is that the critical position of the saturable absorber (in order to assure that the pulses do collide in the saturable absorber jet) be unaffected by adjustments of the cavity length. The use of an antiresonant ring³¹ as one end mirror of the linear cavity enables this condition to be satisfied.

An antiresonant ring consists of a 50% optical beam splitter and a ring formed by mirrors to return the beams to the splitter. An incoming pulse from the linear part of the cavity is split into two equal parts by the 50% splitter; when the two pulses recombine on the splitter, they constructively interfere so that the entire pulse is returned to the cavity. As suggested by Siegman³¹ and subsequently demonstrated with Q-switched Nd:YAG³² and Nd:Glass³³ lasers,

colliding-pulse mode locking may be obtained by situating a saturable absorber exactly opposite the 50% splitter in the antiresonant ring.

A diagram of the dye laser is shown in figure 3-11. The laser consists of a four-mirror linear cavity, for which one end mirror is a 5% output coupler mounted on a translation stage and the other is an antiresonant ring. The gain medium is a 200 μm jet of rhodamine 6G in ethylene glycol, and the saturable absorber is a 20 μm jet of DODCI in ethylene glycol. A sequence of four Brewster prisms is placed in one side of the cavity and is adjusted to compensate for any other dispersion in the cavity. This is accomplished by adjusting one of the prisms in and out of the beam to introduce a variable amount of quartz into the laser cavity. In this manner the cavity can be adjusted through the point of zero dispersion.

With a DODCI concentration of $2 \times 10^{-3} \text{M}$ and a pump power of 1.5W the output power is 80 mW at 615 nm. In the CPM regime, the laser generates pulses with an autocorrelation width of 130 fsec, which for sech^2 pulse shapes, corresponds to a real pulse width of 85 fsec. A typical autocorrelation trace is shown in figure 3-12. The laser spectrum is shown in figure 3-13, and has a width of 60 Å FWHM indicating a time-bandwidth product of 0.41. The transform limit²¹ for sech^2 pulses is 0.31. The prism sequence is optimized by adjusting one of the prisms -- the optimization is demonstrated in figure 3-14. When the sequence over-compensates (negative group velocity dispersion present in the laser) the cavity then one obtains pulse width and spectrum as shown in figure 3-15a. The pulse has the same shape as that at optimum, however it is

Figure 3-11 Experimental layout for the synchronously pumped CPM laser

Figure 3-12 Pulse autocorrelation of the output of the synchronously pumped CPM laser. If one assumes a sech^2 pulse shape then the pulsewidth is 85 fsec.

Figure 3-13 Laser spectrum to go with the autocorrelation trace in figure 3-12. The spectral width FWHM is 60 Å. If one assumes a sech^2 pulse shape then the time-bandwidth product is .41.

Figure 3-14 Variation of the pulse duration for different amounts of glass inserted into the cavity. The minimum pulse width is assumed to be at the point where there is zero group velocity dispersion in the cavity.

Figure 3-15 Pulse autocorrelations and laser spectrums for different signs of the group velocity dispersion. Case a.) is when the cavity is overcompensated and there is negative group velocity dispersion present. Case b.) is when the cavity is undercompensated and there is positive group velocity dispersion present. Case b.) was the normal condition in lasers run without the prism sequence and has the same pulse shape as was seen in previous synchronously pumped lasers.

slightly broader. When one under-compensates (positive group velocity dispersion present) one obtains that shown in figure 3-15b. The pulse is has large wings and is broad, and looks similar to the pulse shapes obtained in the previous laser configurations. This indicates that the previous lasers did indeed have positive group velocity dispersion in the cavity as was predicted.

The synchronously pumped CPM laser operates reliably with a pulse width that varies between 70 and 100 fsec depending on the age of the DODCI used in the absorber jet. The dye lasts for about two weeks and then is changed in order to ensure sub-100 femtosecond pulse durations.

A cavity dumper was added to the laser cavity as well (the prism sequence had to be eliminated in this case) in order to increase the output energy per pulse for some experiments. No change in the output pulse duration was observed by the insertion of this cavity dumper. A Spectra-Physics cavity dumper was modified³⁴ to run at 100 MHz and provided a 20 ns acoustic pulse to "pluck" one pulse out of the cavity. The contrast is shown in figure 3-16. Figure 3-16a shows the recovery of the dye oscillator cavity after the pulse is extracted, while figure 3-16b shows the pre- and post-pulses of about 10% of the main pulse. With losses, an output energy of 2 nJ was obtained at a repetition rate of 5 MHz.

a.)

cavity recovery

b.)

switched out pulse

Figure 3-16 Cavity-dumped output. The dye cavity recovery is illustrated in a.), while the output pulse contrast is illustrated in b.). The cavity dumper was run at 5 MHz in these photographs.

III.D SYNCHRONIZED AMPLIFYING PULSE

The main advantage to using a Nd:YAG laser to pump a dye laser is the ease in amplifying the ultrashort pulse after it is formed. In order to amplify the ultrashort pulse we employ a synchronized energetic pulse to pump the amplifiers in a method we have called synchronous amplification. There were three methods used to form this synchronized energetic pulse; using a pulsed regenerative amplifier, RF coupled Q-switched laser and a CW pumped regenerative amplifier. Each of these will be described.

III.D.1 PULSED REGENERATIVE AMPLIFIER

In any pulsed laser the lasing starts from a fluorescence noise burst which is then preferentially amplified by multiple passes through the lasing medium. In a modelocked and pulsed laser, a restriction is that the noise burst must occur at a particular point in time so that the pulse inside the cavity is synchronized to the modelocker frequency. Regenerative amplification offers a method to create a pulse inside a laser cavity without relying on noise to produce the pulse. By injecting a pulse from a second laser cavity which has more energy than the energy in a noise burst, the injected pulse is preferred and then dominates the laser output. Furthermore, the output pulse is perfectly synchronized to the injection pulse in a method we call optical synchronization.

By employing a pulsed Nd:YAG cavity to be the cavity for the regenerative amplifier and seeding it with the output from the CW mode-locked Nd:YAG laser used to pump the dye oscillator the need for an energetic pump pulse (from the pulsed regenerative amplifier) precisely synchronized to the ultrashort dye pulses can be satisfied. Previous researchers had used pulsed Nd:YAG regenerative amplifiers to provide a synchronized long and short pulse for laser fusion studies.³⁵ We built such a system and used it to pump and fully characterize the dye amplifier chain.

A diagram of the laser system used to produce the high energy pulse^{36,37} optically synchronized to the dye pulses is shown in figure 3-17. When the output from the CW mode-locked Nd:YAG oscillator is frequency doubled, not all of the $1.06\text{ }\mu\text{m}$ radiation is converted to $.532\text{ }\mu\text{m}$. The remainder is separated from the second harmonic with a dichroic beam splitter and sent to the regenerative amplifier. The beam is then passed through a Pockels cell switchout in order to "pluck" one pulse out of the train of 100 MHz at the repetition rate of the pulsed regenerative amplifier (10 Hz). A double Pockels cell switchout is used for three reasons. First, it provides isolation for the doubling crystal from a spurious back reflection from the amplifier. Even a slight back reflection can have a relatively high energy and when focussed onto the doubling crystal could exceed the damage threshold for the crystal. The double Pockels cell switchout had a contrast of $1:10^5$ which was deemed sufficient to protect the doubling crystal from backreflections. The second reason for the switchout was to prevent

Figure 3-17 Experimental layout for the optically synchronized pulse regenerative amplifier. Diagram taken from reference 36.

the train of 100 million pulses per second from bleeding away the gain in the pulsed Nd:YAG rod as it was being pumped but before the regenerative amplifier was injected. The third reason for the switchout was to provide a single pulse to be injected into the cavity. The regenerative amplifier cavity is a pulsed flashlamp pumped Nd:YAG rod placed between mirrors. The cavity was made as long as possible so that higher order spatial modes due to self-focussing would disperse in space before returning to the laser rod and be reamplified. By installing two polarizers and a Pockels cell one could frustrate the laser cavity (with zero volts applied to the Pockels cell) and allow the gain to build up in the rod. When the rod is fully pumped, then one pulse is switched out from the train of 100 MHz and the Pockels cell in the regenerative amplifier is fired to clear the cavity optically, allowing the injected pulse to rattle back and forth in the cavity. After 20-25 round trips through the cavity the energy has been completely extracted from the rod, and the pulse is cavity dumped out of the regenerative amplifier. The energy in the single $1.06\text{ }\mu\text{m}$ pulse is 1 mJ. A double pass amplifier was then used in order to boost the energy in the single $1.06\text{ }\mu\text{m}$ pulse further to 30 mJ. Because the dyes in the dye amplifier chain absorb $.532\text{ }\mu\text{m}$ radiation, a frequency-doubling crystal is used to convert the pulse to the green. A CDA crystal is used and we obtain a 70 psec 10 mJ pulse of $.532\text{ }\mu\text{m}$ wavelength at a repetition rate of 2 Hz.

This laser configuration satisfied the requirements that were needed for both temporal synchronism and green pulse energy.

However, there were several details which inhibited its continuous operation. Since the CW oscillator and the regenerative amplifier were coupled optically, any time the CW oscillator needed to be realigned or the doubling crystal adjusted, the regenerative amplifier had to be realigned completely. It took on the order of six hours to fully align the regenerative amplifier, so this was a serious problem. The other major problem was the use of krytrons in the electronics circuitry which drove the Pockels cell switchout and intercavity Q-switcher. Krytrons supply the high voltage needed to electro-optically switch the polarization of the optical beam. These devices do, however, have a temporal jitter on the order of 1 nsec, drift over the course of the day and need replacing every 10^6 shots which was about once a month. The jitter and drift in the electronics circuit was transformed directly into an amplitude instability in the regenerative amplifier and from there into the dye amplifier chain. The third limitation was the repetition rate limited to 2 Hz by both the pulsed Nd:YAG laser head and the krytron electronics. This limited repetition rate made signal averaging quite difficult and time consuming. For all of these reasons we pursued a different method of forming an energetic synchronized optical pulse.

III.D.2 RF SYNCHRONIZED MODE-LOCKED Q-SWITCHED LASER

In order to modelock the CW Nd:YAG laser an acousto-optic

crystal is driven with a high power RF source whose frequency corresponds exactly to one over the cavity round-trip time. The RF is therefore well synchronized to the train of pulses emitted from the laser. By driving two different lasers with the same RF source, the trains of pulses from the two lasers are synchronized to better than 20 psec.³⁸ By taking advantage of this synchronization we were able to again achieve a source of high power synchronized pulses for short pulse amplification.

The two lasers that were RF synchronized were the main CW Nd:YAG oscillator used to drive the dye oscillator and a second CW Nd:YAG laser which is not only mode-locked but also Q-switched.³⁹ The laser configuration is diagrammed in figure 3-18. All of the modifications made to the CW mode-locked Nd:YAG laser (as described earlier) were incorporated into this cavity as well. In this laser a second acousto-optic crystal (Q-switcher) is placed exactly in the middle of the laser cavity. The same RF is used to drive both the Q-switcher and the modelocker crystals. Each acousto-optic crystal can act as a modelocker and define where in the cavity the laser pulse occurs. This results in competition between the two acousto-optic crystals which in turn leads to an amplitude instability. In order to avoid this problem, cable lengths were carefully chosen between the two acousto-optic crystals to adjust the RF phase between them as well as placing the Q-switcher exactly in the optical center of the laser cavity. The Q-switcher induces a large loss in the cavity which nearly frustrates lasing. The weak lasing (prelase) that does occur establishes the nicely shaped and

Figure 3-18 Experimental layout for the RF synchronized modelocked and Q-switched Nd:YAG laser.

short optical pulse as well as synchronizes the pulse to the modelocking RF. By nearly frustrating the cavity, gain can be stored in the laser rod which can be released by shutting off the loss due to the second acousto-optic crystal and allowing the laser to Q-switch. A train of 20-25 pulses (FWHM) separated by 10 nsec is thereby produced with a total energy of 3 mJ.

The peak pulse in the train can be switched out by the use of an external Pockels cell switchout resulting in a 100 μ J pulse at 1.06 μ m wavelength at a repetition rate up to 500 Hz. The repetition rate of the laser is limited by the time required for the relaxation oscillations to decay and the prelude to stabilize completely. This is illustrated in figure 3-19.

To measure the timing accuracy (jitter) between the two pump lasers the intensity autocorrelation of each was taken. A Gaussian pulse shape was assumed to determine their respective pulse widths. The cross correlation was then measured between the two lasers. The jitter was calculated, assuming a Gaussian distribution using:

$$t_{cc}^2 = t_1^2 + t_2^2 + t_j^2 \quad (3-1)$$

where t_{cc} , t_1 , t_2 , and t_j are the full-width half-maximum widths of the cross correlation, first laser, second laser, and the timing fluctuation, respectively. In this manner the jitter is measured to be approximately 40 ps. There is a large uncertainty in this value due to the inherent difficulty in using the cross correlation for jitter measurement; however, this value is viewed as

Figure 3-19 Drawing of the laser output when the modelocked and Q-switched laser is run at 500 Hz. The spikes are the relaxation oscillations which die down into a prelude shown as the dark band. After the laser is Q-switched the laser shuts off and recovers with relaxation oscillations to complete the cycle.

an upper limit in light of previous work on mode-locked oscillator synchronization³⁸. The autocorrelations of each pump laser and the cross correlation between the two are as shown in figure 3-20. This figure is included to illustrate the relative clarity of the data, not the relative widths since the horizontal scales are not equal.

One major advantage to RF synchronization rather than optical synchronization is the ease in timing between the oscillator and the amplifier. The delay can be varied by changing the RF phase to the amplifier pump laser mode locker eliminating the need for an optical delay line. In this manner, the timing could be optimized without any change in the optical alignment.

The output of the mode-locked and Q-switched laser can be amplified by using an auxiliary Nd:YAG amplifier and reducing the repetition rate to 7 Hz. The amplifier setup is illustrated in figure 3-21. After the optical-pulse switchout a single pulse has an energy of 100 μ J. The amplifier chain consists of three in-line amplifiers. The single pass gain of each can be as high as 30. Rod diameters in the first two heads are 7 mm and are separated by a spatial filter. The spatial filter not only filters out the "hot spots" on the beam but also acts as a beam expander to optimize the size of the beam for passage through the next Nd:YAG amplifier rod. Following the second amplifier is a focusing beam expander and a vacuum spatial filter. The output of the third amplifier (9-mm diameter) is a 100 psec pulse of up to 100 mJ energy.

The IR pulse from the amplifier chain is frequency-doubled in an angle-tuned KD*P crystal. An overall energy conversion

Crosscorrelation
Maximum Jitter: 43 psec

Autocorrelation
Nd: YAG number 1
Pulsewidth: 110 psec

Autocorrelation
Nd: YAG number 2
Pulsewidth: 90 psec

Figure 3-20 Autocorrelations of the CW modelocked laser and CW modelocked and Q-switched laser together with the crosscorrelation of the two lasers indicating the temporal synchronization of the two lasers. The x-scales are not the same. The photographs are shown merely to illustrate the quality of the data. The maximum jitter measured using this method is 43 psec.

Figure 3-21 10 Hz amplifier experimental setup. Spatial filters are used throughout the amplifier chain both to clean the spatial beam profile of any "hot" spots as well as reduce the energy loss due to ASE in the amplifier chain. The output energy at 1.06 μm was 80 mJ while the output energy at .53 μm was 50 mJ. The output pulsewidth was measured to be 100 psec.

efficiency of 60% is routinely obtained. Both the green and the IR are used to drive experiments, detection equipment, and the dye amplifier chain.

The careful consideration of the beam quality and beam diameter throughout the amplifier chain, coupled with the excellent output characteristics of the RF coupled Nd:YAG Q-switched (QS) laser and a new Pockels cell design (this will be described in the next section) result in a superb output beam for experiments. The output beam quality is excellent as evidenced by the figure of 60% conversion to the second harmonic. The pulse-to-pulse energy fluctuation out of the QS laser and the new Pockels cell switchout is $\pm 0.8\%$ as illustrated in figure 3-22a. Adding the three in-line amplifiers increases this fluctuation slightly to $\pm 2\%$ as shown in figure 3-22b.

The excellent beam quality and pulse-to-pulse energy stability are particularly important when using the output to drive a dye amplifier. As will be described in section III.E, the dye amplification process is highly non-linear and extremely sensitive to changes in both the beam quality and the pump energy.

III.D.3 TRIODE POCKELS CELL DRIVER

A new electrical driver for a Pockels cell was developed which gave far superior output characteristics than that which was previously available. Previous methods used to drive Pockels cells

Figure 3-22 Output energy stability histograms from the RF synchronized oscillator (a.) and the amplifier chain (b.). The oscillator has a stability of $\pm 0.8\%$ while the amplifier output is $\pm 2\%$.

used an avalanche-stack-driven krytron to switch the high voltage needed for the Pockels cell. A square pulse was generated of the appropriate voltage and was then propagated across the Pockels cell in a 50 ohm geometry. Simple calculations can show that the current required during this pulse is on the order of 100 amps and the stored charge required is $1\mu\text{C}$. This is a lot of current to dissipate, especially when running at high repetition rates. In addition, the output pulse had a pulse-to-pulse jitter of 1 nsec and a drift over the course of the day of 30 nsec. As the krytron aged, a further drift of 100's of nsec had to be taken into account. The circuit had a lifetime of 10^7 shots or about one month of operation at 7 Hz. Operation at higher repetition rates (such as 500 Hz or 1 kHz) was deemed possible but not practical since the lifetime and drift time of the circuit would be decreased by the same factor by which the repetition rate was being increased. In summary, the Pockels cell circuit did not offer the reliable, consistent, hands-off operation desired. A new circuit was therefore developed to solve several of these difficulties.

The electrical diagram of the Pockels cell driver³ is shown in figure 3-23. The main component in this circuit is a "hard tube" microwave triode which offers a method of switching high voltages with negligible jitter, drift, or lifetime problems. The lifetime of the tube is guaranteed by the manufacturer to 1 year which is already far superior to the krytron switchout. The circuit is built directly onto the Pockels cell itself, with no cable lengths from the circuit to the crystal. By treating the crystal as a capacitor rather than

Figure 3-23 Electrical diagram of the new high-repetition-rate pockels cell driver.

as part of a 50 ohm transmission line, much less charge is needed to apply the necessary voltage to the crystal. The capacitor C_1 is charged through resistor R_1 to the desired voltage. When the circuit is fired, an external pulse is used to trigger the two-transistor avalanche stack T_1 and T_2 . The grid of the microwave triode (which was biased in the off state to -150V) is injected with charge stored in C_2 sending it suddenly to a positive voltage. This fires the triode, which dumps the high voltage present on the capacitor C_1 causing the charge present on the other plate of C_1 to pulse charge the Pockels cell (C_3). The voltage across the Pockels cell rapidly increases to the same voltage as was across C_1 since the capacitance of the Pockels cell is much less than C_1 . This voltage then decays with a time constant given by the RC time of the Pockels cell capacitance and the shunt resistance (it can obviously be tailored to the needs of the experiment somewhat by changing the shunt resistance). Ordinarily, this gives a decay time of between 1 and 10 μ sec as shown in figure 3-24. If the voltage applied is the half-wave voltage of the Pockels cell, then the transmission of an optical pulse through two crossed polarizers with this Pockels cell between them is shown in figure 3-25a. If the voltage applied is the full-wave voltage of the crystal than the transmission is as shown in figure 3-25b. One can see that this configuration can be used to switch a single pulse out of a train of pulses. The only difficulty is that the full-wave voltage is as much as 11 kV, which is close to the limit for the triode. To solve this problem an avalanche stack (T_{1-17}) is placed across the Pockels

Figure 3-24 Electrical output waveform from the pockels cell driver. The driver produces a voltage pulse with a 5 nsec risetime and a 1-10 μ sec decay time depending on the value of the resistor across the pockels cell.

Figure 3-25 Temporal transmission profiles from the pockels cell between crossed polarizers for different voltage outputs from the electrical driver. Figure 2-25a provides a step transmission function since the voltage rises to the half-wave voltage of the pockels cell. Figure 2-25b produces a transmission pulse since the voltage passes through the half-wave voltage of the pockels cell as it rises to the full-wave voltage of the pockels cell. Figure 2-25c is also a transmission pulse which is produced by a voltage pulse with peak voltage at the half-wave voltage. The voltage decay time is shortened dramatically by the implimentation of the avalanche stack across the pockels cell.

cell and self-avalanches at a voltage given by the number of transistors placed in series. If one adjusts the number so that the peak voltage corresponds to the half-wave voltage of the crystal, than one obtains an electrical pulse applied to the Pockels cell with a 5 nsec rise-time and a 5 nsec fall-time. The optical transmission in this case is 100% at the peak with a 2 nsec window FWHM as shown in figure 3-25c.

The shot-to-shot jitter is unmeasurable at <100 psec. A slightly modified circuit from this one has achieved repetition rates of 10 kHz limited only by the current supplied by the power supplies. One Pockels cell used now has $>5 \times 10^9$ shots accumulated with no change in the operating characteristics.

Experimentally, the greatest improvement in the switchout is its reliability. The switchout has become an element which now runs unattended by itself reliably, rather than an element which one must battle and monitor continuously throughout the day. In addition, one can run the switchout at high repetition rates, allowing for an increase in signal averaging capabilities. Both of these improvements have allowed for the study of weak, non-linear effects such as that studied in this thesis.

III.D.4 CW PUMPED REGENERATIVE AMPLIFIER

The most recent source used to generate a high-power pulse^{4,39} synchronized to the dye oscillator pulses combines

components and techniques used in each of the three previously described laser amplifiers. In the past the repetition rate of Nd:YAG amplifiers has been limited by the flash lamps and power supplies to about 30 Hz. Alternatively a cw mode-locked and Q-switched laser could be used as an amplifier source; however, the selection of a single pulse from the Q-switched pulse train reduces the available energy to about 100 μ J. In addition, the repetition rate of such a system is limited to 500 Hz for good stability to be ensured. All of these limitations can be overcome by using a cw Nd:YAG laser operated as a regenerative amplifier.

The configuration of such an amplifier integrated into a system as the source for a dye amplifier is shown in figure 3-26. The input pulse is a portion of the 1.06 μ m radiation from the cw mode-locked Nd:YAG laser that is used to pump the dye laser. These pulses are then injected into a fiber for transport to the regenerative amplifier. The use of an optical fiber not only decouples the alignment of the two lasers but also allows the oscillator and amplifier to be in different locations (tables or even rooms) without the need for long beam paths. A 4% reflector follows the fiber and serves to protect the fiber end from being damaged by leakage from the amplifier. The injected pulses are of approximately 10 pJ energy and 100 psec temporal duration. A quarter-wave plate is placed in the cavity both to frustrate cw lasing and to reject the injected pulses prior to the Pockels cell being fired. The amplifier is triggered by applying a voltage sufficient for a quarter-wave rotation to the injection Pockels cell (PC 2) to compensate for the

Figure 3-26 Experimental layout for the CW pumped regenerative amplifier.

quarter-wave plate. This action not only Q-switches the laser but also traps one of the injected pulses in the cavity. Following approximately 45 round trips (500 nsec), the cavity-dumping Pockels cell (PC 1) is triggered and the pulse is rejected from the cavity off one of the polarizers.

The regenerative amplifier has been further simplified by using a single Pockels cell for both the Q-switching and the cavity-dumping operations. By eliminating one of the two Pockels cells, the intracavity loss is reduced and the output energy should increase as well.

The system can currently be triggered at any repetition rate up to 2 kHz limited by acoustic ringing present in the lithium niobate Pockels cells which distrupts the stability of the regenerative amplifier. A plot of output pulse energy versus repetition rate is shown in figure 3-27. It is expected that the pulse energy will remain constant until the repetition rate approaches the inverse storage time of the Nd:YAG (3 kHz). Figure 3-28 illustrates this behavior in a heuristic plot of the output energy and power versus repetition rate. This would indicate that the roll-off at 1 kHz present in figure 3-27 is due to either uncertainty in the measurement, acoustic ringing, or a thermal effect in the Pockels cells. The output pulse energy is 0.8 mJ with two Pockels cells used in the regenerative amplifier cavity and 1.1 mJ with a single Pockels cell.

At one kilohertz the average power is 1 W, far below the average power one can obtain from a laser which is simply cw

Figure 3-27 Average output power and output energy per pulse for different values of the repetition rate.

Figure 3-28 Heuristic plot of the regenerative amplifier output for repetition rates up to 5 kHz.

mode-locked (10W). The reason for this is two-fold. First, the repetition rate is not the optimum as one can see in figure 3-28. Second, a high contrast dielectric ("hard") polarizer is used in this cavity which strips off 99% of the incorrect polarization on each pass. Thermally induced birefringence in the Nd:YAG rod optically rotates the beam considerably, and this in turn causes a loss in the cavity. Normal cw mode-locked lasers do not have a "hard" polarizer in the cavity, but rather a brewster plate which only eliminates 15% of the incorrect polarization on each pass. The second point is the major factor in the lower output power from the regenerative amplifier and causes one to consider using a different laser rod -- one which has a lower thermally induced birefringence. One possible candidate is Nd:YLF which not only has a lower thermal birefringence, but also a higher stored energy.^{40,41}

Since the amplifier is to be used as a source of pump pulses for the dye amplifiers, the 1.06 μm pulses are frequency doubled using either a 3-mm potassium titanyl phosphate (KTP) crystal or a CDA crystal. Energies of 350 μJ at 532 nm are obtained which indicates a 40% efficiency. This is the optimum value obtainable given a gaussian pulse in both time and space as illustrated in figure 3-29. The output-energy stability of this system is shown by generating a histogram of the doubled energy of 32000 consecutive laser shots as illustrated in figure 3-30. The energy fluctuation at .532 μm is $\pm 1.2\%$ with 99% of the shots falling within $\pm 2.7\%$. The excellent stability of the amplifier output pulse makes this source ideal for pumping a dye amplifier chain.

Figure 3-29 Conversion efficiency versus input laser intensity into a KDP frequency doubling crystal for different laser pulse shapes. The maximum efficiency for a pulse which is both temporally and spatially gaussian is approximately 50%. Graph taken from reference 42.

Figure 3-30 Output energy histogram for 32,000 consecutive regenerative amplifier laser shots at .532 μm . The energy fluctuation is $\pm 1.2\%$ with 99% of the shots falling within $\pm 2.7\%$.

III.E SYNCHRONOUS DYE LASER PULSE AMPLIFICATION

As mentioned earlier in this chapter the main reason for developing the doubled Nd:YAG pumped dye oscillator and the synchronized pulsed Nd:YAG output was in order to more efficiently amplify the dye oscillator pulses using a process we call synchronous amplification. Over the past five years, considerable effort has been expended towards fully understanding the dye amplifier parameters necessary for optimum short pulse amplification^{2,4,36} given different input pulse powers and repetition rates. Two of the dye amplifier configurations will be described in detail: a 7 Hz, 1 mJ output dye amplifier, and the 1kHz, 10 μ J output dye amplifier used to perform most of the thesis experiments.

The idea of synchronous amplification is simply illustrated in figure 3-31. A pump pulse is focussed into a cell of organic dye and is absorbed, placing many of the dye molecules in the first excited singlet state of the dye. A weak pulse to be amplified enters the cell a precise time after the pump pulse and is amplified by stimulated emission to access the energy stored in the dye amplifier cell. By synchronizing the pumping pulse and the pulse to be amplified, a much more efficient amplifier can be built with little background amplified spontaneous emission (ASE).

There are three general mechanisms for the decay of the gain in the dye amplifier stage; spontaneous emission of the dye

Figure 3-31 Synchronous amplification geometry illustrating collinear pumping.

molecules, molecular reorientation which realigns the dye molecules in a different polarization which cannot be accessed by the weak input pulse, and ASE which steals the stored energy from the short pulse for itself. Each of these mechanisms has been studied thoroughly.

The fluorescence lifetimes of the dyes used in the amplifier are greater than 1 nsec in all cases. Kiton red was used for the majority of the dye amplifiers developed and has a lifetime of 2.8 nsec⁴⁴ which is much longer than either the pump or dye pulse temporal durations. As a result it does not have a noticeable effect on the decay of the gain for synchronous amplification with 100 psec pulses.

Molecular reorientation is the loss of gain due to the fact that the dye molecules which are excited in one polarization can, due to normal thermal motion, rotate and thus be inaccessible to the short pulse to be amplified in the same polarization as the pump pulse. The reorientation time is dependent on the choice of solvent used in the amplifier stage and was measured experimentally for solvents of both water and ethylene glycol.

When a gain medium is pumped, spontaneously emitted photons are generated and propagate out into all directions in space. If the photons propagate in the direction that the gain medium was pumped then the photons "see" gain and are amplified. This ASE can grow and thereby remove a large amount of the the stored energy in the amplifier cell. The ASE plays the dominant role in the decay of the gain in high-gain amplifiers and can, in extremely high gain

amplifiers, cause the gain to be depleted in a time equal to the time for light to propagate across the cell. In all of the dye amplifier cells used the gain region is pencil-like as illustrated in figure 3-32. The ASE generated therefore is very directional and travels on the same beam path as the amplified pulse. As a result, this noise source must be eliminated by other means in order to have a clean single pulse output from the dye amplifier.

Figure 3-33 is the experimental setup used to measure the decay time of the gain in a one-stage amplifier.³⁶ The input pulse was delayed by a multipass delay line, made collinear with the pump beam by the use of a dichroic mirror, and then focussed into the dye amplifier cell. By noting the gain as a function of the delay between the pump and the probe, the decay of the gain could be mapped out. The transient gain in a single dye amplifier is plotted in figure 3-34. One can clearly see the integral of the pump pulse on the leading edge of the gain, and then the gain depletion on the trailing edge due to the mechanisms previously mentioned.

Although there is no physical reason that the decay should be exponential on the trailing edge, it was found to be very close to that. The $1/e$ time was then used as the gain storage time in the amplifier for studies of the effects of ASE and molecular reorientation. Experimentally, the logarithm of the data was taken and a best-fit straight line was calculated from just beyond the peak of the gain profile to the end of the run. An example of this is shown in figure 3-35. From this best-fit straight line, we calculated the slope and from that the $1/e$ time of the data.

Figure 3-32 Propagation direction of the ASE given the geometry of the gain region.

Figure 3-33 Experimental layout used to make measurements of the gain storage time in the dye amplifier chain.

Figure 3-34 Temporal profile of the gain in the dye amplifier stage.

Figure 3-35 Logarithm of the temporal gain profile illustrating the superb exponential fit. The decay time of the exponential can then be used as a measure of the storage time in the dye amplifier stage.

Figure 3-36 is a plot of the gain storage time in the amplifier as a function of the pump pulse intensity for two different solvents, water and ethylene glycol. Molecular reorientation effects should be unaffected by pump intensity. The graph therefore implies (at the lowest values of the pump intensity where ASE is minimized) that the molecular reorientation time in water is 290 psec and that in ethylene glycol is 600 psec. Reorientation times for water found in the literature⁴⁵ correspond quite well with our data, however the decay time of ethylene glycol do not⁴⁶. This discrepancy could be due to a loss of viscosity when water is present in the ethylene glycol. A very small amount of water (perhaps absorbed from water vapor in the air) reduces the viscosity of the solvent tremendously and thereby the reorientation time.

At higher pump powers and gains ASE tends to dominate the decay time of the gain due to its ability to access more of the available excited state population. This is clearly illustrated in figure 3-36 by the decline of the gain storage time at higher pump powers. At extremely high values of the pump power (higher than that plotted here) the decay time was measured to be less than 30 psec which approximates the transit time across the dye amplifier cell (1 cm).

In order to amplify the input pulses fully the pump power, and therefore the gain, was adjusted carefully to provide the highest gain in the amplifier. As the curve in figure 3-34 illustrates, the decay time at these high pump powers is about 150 psec implying that the time jitter between the dye and pump pulses must be less than 40

Figure 3-36 Variation of the gain storage time versus input pump intensity for two different dye solvents. Graph taken from reference 36.

psec in order to maintain a stable amplifier gain for all input pulses. The fact that we could even measure features which have components as short as 25 psec means that the synchronism between the pump and dye pulses is good to better than this number.

Because the storage time of the amplifier is on the order of 150 psec, if one uses a pump pulse which is much longer than this, the population is cycled up and down by the ASE with a cycle time of 150 psec. This energy is wasted and in fact goes primarily into ASE, a noise background on the amplified dye pulse. By reducing the number of cycles to one, in our case, the ASE generated is minimized. This is a major advantage of synchronous amplification as compared with other unsynchronized amplifiers with >5 nsec pulse durations.⁷

Two dye amplifier designs will be described: one for high pump powers at a 7 Hz repetition rate, and the other for lower pump powers at a 1 kHz rate. In each design several dye amplifier stages were incorporated together, each operating in a similar manner to the single stage studied previously. Adjustments in the pump power and focussing allowed for the optimum gain while reducing saturation in the amplifier. Saturation, although it does access the greatest number of excited molecules, was to be avoided because the input pulse was temporally reshaped as it passed through the amplifier. If the leading edge of the pulse saw more gain than the trailing edge (as is true in a saturated amplifier), then the pulse is irreversibly broadened temporally.

Using the 10 mJ green pump pulse provided by the RF-coupled

amplifier (described in section III.D.2), a three stage dye amplifier was developed. The experimental setup is shown in figure 3-37 and has been more fully described elsewhere.³⁶ Each stage was a flowing cell of kiton red with concentrations of 2×10^{-4} in the first two stages and 1×10^{-4} in the third. The solvent used in these dye stages was water, used for its low threshold to stimulated Raman generation. The fast molecular reorientation of water did not pose a problem, since our synchronism between the pump and input pulses was much better than the reorientation time as well as the fact that at these pump intensities ASE was the dominant factor in the storage time of the gain. Dichroic mirrors were used which reflected 99% of the green pump light and transmitted 95% of the input pulse. Both beams were focussed by the same lens to various spot sizes. In general the spot size of the green beam was made smaller on the input lens so that at focus the green spot was bigger than the dye. This tended to optimize the gain and preserved the beam quality of the input pulse. After the second stage, a saturable absorber of DODCI or malachite green was used to absorb the small amounts of ASE generated in the first two stages before they had a chance to be further amplified in the final stage. The input pulse, however, had enough intensity to bleach through the absorber with a loss of only a factor of 2. Amplified-pulse-to-ASE-energy contrasts of 1000:1 were typical when the saturable absorber was adjusted properly. This amplifier gave gains of 10^4 in the first stage, 10^2 in the second stage and 10^1 in the third stage. The reduced gain in the later stages is due to the fact that the input pulse is

getting bigger for these stages and the gain must be reduced to prevent running in a saturated regime. The total gain was therefore 10^7 which amplified the 100 pJ input pulses into 1 mJ output amplified pulses. Since the pump pulses and the input pulses were made collinear, the output beam profile of the amplified dye pulses matched that of the input pump pulses. It was for this reason that great care was taken to make an extremely clean pump beam profile.

Typical input and output pulse autocorrelations are shown in figures 3-38 and 3-39. One can see that the pulse has not been broadened by the amplification process.

A second dye amplifier has been developed for use with pump pulses from the output of the CW pumped regenerative amplifier (see section III.D.4). This amplifier chain is diagrammed in figure 3-40. The pump pulse energy is less than the previous case (300 μ J) but is at a repetition rate more than 100 times greater (1kHz). A two-stage amplifier was developed that is very similar to the previously described dye amplifier. Ten percent of the pump pulse was used to pump the first stage while the rest pumped the second stage. Mirrors which had one side cut flat were used to make the pump and input beams parallel and therefore focus to the same spot. These were used instead of dichroics to allow flexibility in the wavelengths of the input as well as preventing any dispersion present in the dichroic mirror coatings from broadening the ultrashort dye pulse. At an input wavelength of 615 nm, cells of DCM mixed in a mixture of propylene carbonate, methanol, and water were used at a concentration of 1×10^{-4} M. DCM was chosen for its high gain at the input

Figure 3-38 CW autocorrelation of the input to the 10 Hz dye amplifier chain.

Figure 3-39 Pulsed autocorrelation of the output from the 10 Hz dye amplifier chain.

Figure 3-40 Dye amplifier experimental configuration for the high-repetition-rate dye amplifier.

wavelength, while having a large Stokes shift allowing for considerable absorption at the pump wavelength. A saturable absorber was used between the two stages to control the ASE generation as well as trim the front edge of the amplified pulse if it had been saturated slightly in the first stage. Gains of 10^3 and 10^1 were typical in the first and second stages respectively, giving a total gain of 10^4 and an output energy of $1 \mu\text{J}$. Optimization of this amplifier led to much larger spot sizes than used in the 7 Hz dye amplifier, implying that if larger spot sizes were used with the 7 Hz amplifier, a much higher output energy could have resulted.

The input pulses used with this amplifier were on the order of 90 fsec. This short pulse required a great deal of bandwidth and this in turn required attention to dispersion in the amplifiers. Dispersion can come from all elements in the amplifier chain: coatings,⁴⁷ dyes, glasses, and air. In order to take out the effects of dispersion broadening the input pulses, a grating pair⁴⁸ was employed to induce a negative group velocity dispersion (opposite to that of all the other elements in the amplifier chain) and compensate exactly for their effects. The input pulse had a pulse width of 110 fsec assuming a hyperbolic secant squared pulse shape. The output pulse after pulse compression in the grating pair is shown in figure 3-41 and has a pulse width, assuming the same hyperbolic secant squared shape, of 65 fsec. This pulse compression in amplification is probably due to the self-phase modulation in the amplifier chain which generates new frequencies in the amplified pulse and the grating pair which reconstructs them into the shorter

Figure 3-41 Pulse autocorrelation of the amplified and compressed output of the high-repetition-rate dye amplifier.

output pulse.

By focussing this pulse into a cell of water, a white-light continuum can be generated⁴⁹ due to the extremely high intensities in the water cell. The white light generation is an extremely non-linear phenomenon and has an intensity threshold that must be exceeded. The output pulses from each of these dye amplifier chains produced a healthy white-light continuum. The energy threshold for white light generation was measured to be only 250 nJ which illuminates the fact that the excellent beam quality present on the amplified output beam can be translated into an extremely tight beam at focus -- an irregular, amplified beam could not be focussed as well.

III.F LASER SOURCE CONSIDERATIONS

In order to perform the required experiments a stable laser source of pulses with a duration between 1 and 2 psec was needed. The pulses could not be shorter than 1 psec for fear that the Fourier transform bandwidth would become so large that it would start to cause direct line excitation of the D lines. Pulses longer than 2 psec were undesirable since they could no longer be called "short" pulses for the Na-Ar collision system. In order to achieve this pulse a Lyot filter was added to the dye oscillator cavity. This cavity was more completely described in section III.C.3. In addition to the Lyot filter used to restrict the lasing bandwidth, the

concentration of the DODCI saturable absorber dye was reduced to allow the laser to run at the required wavelengths.

Longer pulses were also needed for some experiments. Pulses with temporal durations up to 50 psec were obtained in a straightforward manner. These longer pulses were generated by adding more Lyot filters to the laser cavity as well as, for the longest pulse durations, Fabry-Perot etalons. Each of these laser configurations gave stable, short, nearly Gaussian shaped pulses.

In order to amplify these pulses a dye amplifier similar to that described in section III.D was constructed. Since the wavelengths used were near 590 nm rather than 620 nm, the dye kiton red was used for the gain medium and DODCI used for the saturable absorbers. Using the same geometry as that previously described, pulse energies of up to 16 μJ were obtained at a repetition rate of 1 kHz. This is, in fact, the most energy ever obtained out of a high repetition rate picosecond dye amplifier with an efficiency of >5%. The major reason for this additional energy is the superb alignment of the wavelength desired for the experiment to the peak of the gain curve of the dye kiton red. At 620 nm the amplifier described in section III.E is forced to run on the edge of the gain curve of DCM.

Great care was also needed to ensure that the pulse duration was not modified during the amplification process. Figure 3-42 illustrates the autocorrelation of the input pulse (solid line) to the output pulse (dots). As one can see, the pulse duration is indeed preserved although the wings of the dye pulse are greater. Recompression of the dye pulse using a grating pair compressor⁴⁹ to

Figure 3-42 CW and pulsed autocorrelations of the amplified 1.6 psec pulses at 5916 Å. The solid line is the CW autocorrelation while the dots are the amplified autocorrelation. Some pulse reshaping is evident due to the amplification process however the pulse width is approximately the same for both cases.

attempt to eliminate these wings was undesirable due to the large energy losses (>50% loss) associated with the grating pair. Similar checks were made for the longer pulse duration with superior results (in terms of the preservation of the pulse shape) to that described for the short pulse.

III.G SODIUM VAPOR CELLS

Two sodium vapor cells were constructed to provide the environment for the collision studies. The first was a stainless and sapphire cell used without a buffer gas to investigate Na+Na collisions. The second was a vapor cell in a heat pipe design used to study Na+Ar collisions. Each of the cells construction and operation will be described.

III.G.1 Na+Na VAPOR CELL

A cell was constructed which allowed the experimenter to view solely Na + Na collisions since a buffer gas was not used. Very few reliable cells have been made which provide the high vapor density of pure Na necessary for collision studies. The reasons for this are obvious: sodium is a highly corrosive material (especially in the vapor phase) and finding materials which it does not attack is difficult.

In every vapor cell, the vapor condenses on the coldest point in the cell chamber. When one does not use a buffer gas to protect the windows used for viewing the reactions (as is true in the case of a heat pipe) then the window itself must be heated to the high temperatures required to maintain a high vapor pressure. This places the most difficult engineering task on determining the window and window seal which can not only withstand the attack of the sodium vapor, but also maintain a high vacuum seal (<1 mTorr) during a temperature cycling up to 450°C . Although all these requirements may seem mutually exclusive, an attempt was made to build such a cell.

The cell built was a near copy of one built by Huennekens⁵⁰ at the University of Colorado. A general diagram of the cell and vacuum handling is shown in figure 3-43a with a detailed drawing of the cell chamber itself illustrated in figure 3-43b. The cell has two chambers, one which is maintained at a stable temperature with a "puddle" of sodium in it providing the Na vapor for the cell and the other, at a slightly higher temperature, equipped with windows to inject the laser and monitor the output fluorescence. The viewing chamber is a cross drilled in a block of 304 stainless steel with a vacuum connector to the sodium puddle. Sapphire rods are placed in three of the sections of the cross. Two of the rods are used to transport the laser beam into and out of the observation region while the third is used to minimize the effects of radiation trapping by transporting the fluorescence out of the cell without having it pass through great lengths of unexcited sodium vapor. Sapphire is used

Figure 3-43 Na-Na vapor cell. The top diagram illustrates the gas handling used with this cell. The bottom diagram is a detailed drawing of the cell chamber itself.

since it is somewhat resistant to attack by sodium vapor. The viewing chamber was typically maintained at a temperature 50 °C higher than that of the puddle to prevent the sodium from plating on the sapphire rods (which could be slightly cooler because of conductive cooling from the windows exposed to outside air) and to reduce the number of dimers in the viewing chamber by thermally breaking them down. The windows were also made of sapphire and were sealed to the stainless viewing chamber with metal O-rings 51 crushed by using a six-point stainless steel flange. A variety of different materials were used for the metal seals with silver surrounding an inconel spring providing the best leak seals given the temperature cycling.

Four 500 Watt cartridge heaters were used to heat the viewing chamber while heating tape was wrapped around all of the other sections of the vapor cell. Separate voltage controllers were used on each of the tapes to maintain an independent temperature at each of the different points in the cell.

Sodium was cut up and placed into the puddle area under a constant backflushing of argon gas to prevent oxidation forming on the outside of the sodium metal. After the metal was inserted the cell was completely pumped down and baked out for several days to remove any impurities. The sodium itself could not be baked and pumped at the same time, but was cycled 4 or 5 times above its melting point to eliminate any impurities which happened to be on the surface. After the cell was fully baked out and proven to be completely leak tight, the high temperature valve was closed off and

the cell was slowly raised in temperature (over 10 hours) until the temperature of the sodium puddle was at the correct temperature to provide the desired vapor pressure, and the temperature of the viewing chamber was 50 °C hotter. If this raising of the cell temperature was successful without the metal O-rings used to seal the sapphire windows springing a leak, then the viewing chamber was not lowered in temperature for any reason. The vapor pressure was varied separately by adjusting the temperature of the sodium puddle and was cooled when not in use to minimize the corrosive effects of the sodium vapor on the sapphire.

In practice this cell could only be raised in temperature up to 350 °C (on the viewing chamber) before the O-ring seals on the windows started to leak or the sapphire windows shattered. The reasons for this are unclear. Careful studies of the expansion coefficients for the sapphire, O-ring seals, 304 stainless, and the stainless bolts used for compression indicate that the thermal expansion should have been easily taken up by the spring nature of the O-ring seal itself. Different flange designs were tested to minimize the stress on the sapphire, with little success. Numerous studies over a 2 and 1/2 year period did not solve these problems despite several marathon attempts. Discussions with Huennekens and others at the University of Colorado did reveal, however, that out of five cells constructed with this design, only the one used by Huennekens performed reliably.

In addition to the low temperature (and therefore low vapor pressure) of this cell, there was a great deal of laser scatter

inside the cell which made Rayleigh scattering measurements impossible. This scatter came from many little pits eaten into the sapphire rods by the sodium vapor which were visible by the fluorescence collection system.

For these experimental difficulties and because initial experiments made on the collision system indicated that higher collision densities were required, this cell was abandoned for the majority of the experimental studies made. A vapor cell using argon as buffer gas as well as collision partner was used to solve all of these problems.

III.G.2 Na-Ar VAPOR CELL

A second vapor cell was implemented in order to study Na-Ar collisions. There were two reasons for this change of cell. First, the Na-Na cell did not work reliably and had a great deal of laser scatter which disrupted the fluorescence collection. Second, preliminary indications were that a much higher collision rate was needed to view the processes at the higher laser inputs. The collision rate is proportional to the number densities of both the active atoms and its perturbers. Since we couldn't raise the number density of either in the previous cell, the decision to change to Na-Ar collisions allowed the flexibility to increase the Ar (perturber) density as high as desired.

The Na-Ar cell was of a modified heat pipe design as

diagrammed in figure 3-44. A stainless steel cross was used with plate glass windows sealed with viton O-rings on all four ports. Inside the cell, a nickel wick was placed 8 cm out from the center on all four ports. The middle of the cell was heated using nichrome wire wrapped around a ceramic wire mold. The ends of each of the ports were cooled by water flowing through copper tubing welded to the sides. In practice, sodium metal was placed into the center of the cross and heated in a high pressure (200 Torr) of Ar. The sodium vapor then flowed around in all directions with some atoms heading out towards the windows. Multiple collisions between the Ar atoms and the Na atoms changed their trajectory sufficiently to make the Na atoms hit the wall a distance away from the center of the cross. At this point the wall is cool and the Na atom sticks to the wall. The nickel wick then "wicks" the sodium metal liquid back towards the heat source. The sodium then makes this cycle over and over with no sodium ever reaching the cold windows and plating there. The pipe was run in oven mode, with argon uniformly distributed throughout.

For the experiments reported in this work the temperature of the cell was maintained at about 300 °C as measured on the outside of the cell. Measurements made of the vapor pressure inside the cell indicated, however, that the temperature inside the cell was nearly 75 °C cooler due to the large temperature gradient between the center of the cell and the ends of each port. The argon used was a high-purity research grade and was maintained at a 250 Torr pressure for most experiments. Figure 3-44 illustrates the gas handling used with this cell.

Figure 3-44 Na-Ar vapor cell. The design of this cell is a heat pipe design however it is not run in heat pipe mode. The pressure of the Ar buffer gas is much higher than the Na vapor pressure and as such the Ar is present throughout the cell. Two pressure gauges are used. The Baratron gauge is used to measure the argon pressure while the thermocouple gauge is used for leak checking.

This cell solved many of the reliability problems of the previous cell. It did require observing foreign-gas-broadened atoms rather than resonance-broadened atoms, however computer simulations indicated that this did not cause significant changes in the intensity required to view the desired effects. A drawback to this cell was the collection efficiency and the presence of radiation trapping; the long and narrow arms of the cross allowed for only a $f/7$ collection system (compared to the $f/4$ of the Na-Na cell) and a long path with unexcited Na atoms which introduces radiation trapping. Both of these effects reduced the signal output significantly.

III.H DATA ACQUISITION HARDWARE AND SOFTWARE

In order to accumulate and reduce the data taken for these experiments a data acquisition system had to be assembled to not only accept and record data, but also perform at the high (1 kHz) repetition rate of the laser. A small mini-computer was interfaced to several input/output devices and software was written optimized to run at the high data rates.

The data acquisition system is illustrated in figure 3-45. A Digital Equipment Corp. LSI 11/23 minicomputer with 256K of user addressable memory was used to run the experiments. There are four RS232 I/O ports for serial communication with terminals, experimental equipment, and a large mainframe computer for more extensive data

Figure 3-45 Block diagram of the computer used to perform the experiments.

manipulation in a multi-user environment. A CAMAC⁵² crate is interfaced to the LSI backplane and is used for all data I/O other than the serial lines. CW and pulsed A/D's, a D/A, and several logic interface modules are used in the crate. Two 8 inch single density disk drives are used for program and data storage. An XY plotter is driven by three ports of the D/A for rudimentary plotting. Three stepper motor controllers have been interfaced through CAMAC, as well as an OMA⁵³ I used for one-dimensional image analysis. The data is displayed on an ADM3A terminal with a Retrographics card which allows it to act as a Tektronics 4010 emulator. An auxiliary keyboard can simultaneously be used elsewhere in the lab (such as right next to the sodium experimental setup) through a different serial line. The video lines to the terminal's CRT have been tapped and manipulated into standard video so that the output can be viewed from anywhere in the lab on several different television monitors.

The software used has been written in the language Forth, chosen for its high speed, easy interfacing with I/O devices, and support at the Laser Energetics Lab. The Forth used is UR/LLE v. 3.01, a version which conforms roughly to the Forth/79 standard⁵⁴. Programming is done in a file system with text compression which frees one from the restrictions made due to blocks (hardware defined units of memory). This is particularly important for program clarity and the ability to add a plethora of comments and visually appealing space to the code without the worry of using excessive disk space. Details of this particular file system have been described elsewhere.^{55,56} A data file management system has also been

developed for archival storage. This system is separate from the programming file system and places a premium on economy of storage space with little overhead. A typical data disk holds 100 data runs of varying file lengths.

A great deal of software has been written for experiments done in this laboratory. A typical experiment is to drive a stepper motor (attached to a linear stage to change a time delay or a wavelength control knob on a monochromator) while monitoring the signal output on an A/D. To perform this experiment the computer moves the stepper motor which drives the linear translator to the proper location, accumulates data, stores and displays the data, and then moves to the next location. A user-designed display can lead to a closer interaction with the data. One consequence of this is that if an experiment is for some reason not performing properly the run can be aborted in mid-run, saving valuable laser time. By averaging a number of laser shots at each translation stage position one can reduce the short term (shot to shot) noise and by taking multiple scans of the entire length of the translator one eliminates long term drift. By appropriately balancing these two methods of data acquisition one can reduce nearly all sources of noise. The software can also be set to monitor the laser energy and reject a particular laser shot if it isn't within certain user-defined bounds.

Although the "step-average" method of data collection just described is used most often, there are times when one desires to acquire data in a manner similar to that of a signal averager. An experiment which can best use a signal averager type of operation is

one which has low shot-to-shot noise but high long-term drift. In this method the stepper motor is set at one end of the scan range and then started towards the end of the scan. While it is moving, the data is sampled at a user-determined rate. By dividing the total travel by the number of data points obtained and by ensuring that the stepper travel is linear and continuous, one can later calculate the stepper position at each data point. This change in data collection scheme offers a clear example of the power and flexibility that one obtains when using a standard collection routine and vectored execution. An example of this type of data acquisition is shown in figure 3-46. In this experiment the stepper motor was controlling the wavelength control on the monochromator observing the different wavelengths of light emitted from the sodium vapor cell pumped by pulses at 100 MHz detuned to the red side of the Na D lines. The feature on the left is Rayleigh scattering of the laser, while the two on the right are the fluorescence from the two sodium D lines caused by collisional redistribution. This data represents the first observation of collisional redistribution using picosecond laser pulses.

Additional software has been written specifically for the experiments carried out in this thesis. One package, in particular, was used to monitor the laser energy focussed into the sodium vapor cell and to tabulate this result along with the resultant fluorescence output. By taking 150,000 shots and calculating the average fluorescence output for a given laser-input energy, the functional behavior of the output with respect to the input can be

Figure 3-46 An example of the "signal averager" method of data acquisition. In this example a stepper motor was used to control the wavelength control on the monochromator observing the different wavelengths of light emitted from the sodium vapor cell. The feature on the left is Rayleigh scattering of the input laser, while the two on the right are the fluorescence from the two sodium D lines.

studied with a high degree of statistical significance.

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CHAPTER IV

EXPERIMENTAL RESULTS

The experimental apparatus described in the previous chapter was applied to the study of picosecond optical collisions. In this chapter the results of the first use of picosecond pulses to study the dynamics of atomic collisions will be detailed.

To perform these experiments two cells had to be built and calibrated. In order to measure the atomic number density at a given measured temperature the method of equivalent width was used. This technique will be described for both the Na-Na collision cell and the Na-Ar collision cell. Measurements of the dependence of the laser detuning on both the Rayleigh scattered component and the collisionally redistributed signal at both D_1 and D_2 will also be described. The dependence of the excited state population versus peak input laser intensity will be presented for three ranges of intensity: low, intermediate and high intensities. Other measurements made to understand the propagation of picosecond pulses in a resonantly absorbing medium will also be described. Finally, the intensity dependence of the excited state population for different values of the laser pulse width will be experimentally

tested.

IV.A METHOD OF EQUIVALENT WIDTH

The method of equivalent width was used to determine the Na atomic density in the two cells constructed. This method was preferred since it does not require either a single-mode cw dye laser or an extremely high resolution spectrometer to determine the exact absorption profile. A relatively crude spectrometer is all that is needed to determine with some accuracy the number density in the atomic cells.

Mitchell and Zemansky have described¹ the area under a measured absorption curve as being independent of the resolution of the instrument being used. Therefore, no matter what the wavelength resolution is of the particular spectrometer used, the area measured is exactly the same as that measured when a spectrometer with sufficient resolution to measure the actual lineshape is used. The equivalent width is related to the absorption lineshape and is defined in terms of the absorbed intensity $I_\lambda(L)$ after propagation through a vapor cell of length L . Given $I_\lambda(0)$, the input light intensity at a given wavelength, the equivalent width is defined as:

$$W_\lambda = \int \frac{I_\lambda(0) - I_\lambda(L)}{I_\lambda(0)} d\lambda \quad (4-1)$$

The value for the equivalent width can be related through a

one-to-one correspondence to the atomic number density^{2,3} The curves are diagrammed in figure 4-1. In this figure the value a is defined as:

$$a = \frac{\Gamma/2}{\Delta\omega_0} \quad (4-2)$$

where Γ is the collisional Lorentzian full width at half height and $\Delta\omega_0$ is the doppler half width at e^{-1} height. The y axis units are:⁵

$$\frac{W_\lambda}{2\pi\Delta\omega_0} \quad (4-3)$$

where W_λ is the equivalent width. The x-axis is in units of:

$$\log_{10} [N L f \lambda^2 \sqrt{\pi} e^2 / (m_e c^2 \Delta\lambda_0)] \quad (4-4)$$

where N is the atomic number density, L is the optical path length in the cell, f is the oscillator strength of the measured line, e is the electron charge, and m is the electron mass. Thus, by measuring the equivalent width and calculating the collisional and doppler widths at the measured temperature, the atomic number density can be determined.

As can be seen in figure 4-1, there are three regions in these curves. One at low values of the number density which has a constant slope and is determined by the natural linewidth, a second at moderate values of the density with a variable slope due to the

Curve of Growth

Figure 4-1 Curve of growth used to convert equivalent width measurements into atomic number density. Graph taken from reference 4.

increasing presence of a doppler broadened lineshape, and the third at the highest values of the number density with a constant slope is due to a collisionally broadened lineshape. As the value a becomes larger the collisional component is stronger, overwhelming the doppler region until at $a=\infty$ one moves directly from the natural line to the collisional line regions (or at very high perturber gas pressures is always in the collisionally broadened region).

The values of the slopes in the purely natural region and the purely collisional region can be determined using some simple approximations.⁶ At low atomic number densities the absorption depth is very long so the absorption can be defined as:

$$I_{\nu}(L) = I_{\nu}(0)e^{-k_{\nu}L} \quad (4-5)$$

where k_{ν} is the absorption coefficient at frequency ν , and $I_{\nu}(0)$ is the input light intensity. Using the approximation:

$$1 - e^{-k_{\nu}L} \approx k_{\nu}L \quad (4-6)$$

one can obtain⁶ the slope of the density curve at low densities:

$$W_{\nu} = \frac{\lambda^3}{8\pi} \frac{g_2}{g_1} A_{21}LN \quad (4-7)$$

where g_2 and g_1 are the statistical weights of the upper and lower levels, N is the number density, A_{21} is the Einstein A

coefficient for the transition, and L is the length of the absorbing region. At high densities the slope is given (using other approximations⁶) by:

$$W_0 = \frac{\lambda^2}{8\pi^2} \frac{g_2}{g_1} N_A N_p A_{21} L k_{br} \quad (4-8)$$

where N_p is the number density of the perturbing atoms, N_A is the number density of the absorbing (target) atoms, and k_{br} is the broadening parameter related to the collisionally broadened line width Γ by:

$$k_{br} = \frac{\Gamma}{N_p} \quad (4-9)$$

Both Eq (4-8) and Eq (4-9) can be transformed into W_λ through the use of the equation:

$$W_\lambda = \frac{\lambda^2}{c} W_0 \quad (4-10)$$

As is evident in figure 4-1 the method of equivalent width is accurate in the collisionally and naturally broadened regions but is not very sensitive in the doppler region. For the experiments done in this dissertation the densities used were primarily in the collisionally broadened region.

IV.B Na-Na EXPERIMENTAL STUDIES

IV.B.1 CALIBRATION OF Na-Na CELL

The method of equivalent widths was used to measure the Na density in the Na-Na collision cell. The experimental arrangement used to make these measurements is illustrated in figure 4-2. An ordinary light bulb was used as the white light source. Since the range of wavelengths used was small, the intensity at a given wavelength was uniform across the trace. The light transmitted through the cell was focussed into a .3m spectrometer used with 10 μ m slits (.3 $\text{\AA}$ resolution) and a photomultiplier tube (PMT) was used to monitor the intensity of the transmitted light. Since there are four arms in this cell there are two possible pathways with two different lengths L to traverse. Measurements were made through both initially to check the results of one with the other, however most measurements were made along the same axis through which the laser will be entering, and which has an optical path length of $L=6.5$ mm. The absorption as a function of wavelength was measured by using the computer to both move the stepper motor driven wavelength adjustment on the spectrometer (which had been previously calibrated) and to measure the output of the PMT used to view the transmitted white light. A representative trace is shown in figure 4-3. The computer was then programmed to measure the area under the curve and obtain a value for W_{λ} . The temperature of the sodium puddle (see figure

Figure 4-2 Experimental configuration used to make the white light absorption measurements.

Figure 4-3 White light absorbed due to the D_2 line. The area absorbed by the resonance line was measured to determine the equivalent width.

3-43) could be changed to alter the sodium vapor pressure in the cell. From this temperature one can approximate the sodium vapor pressure either by using vapor pressure curves or the equation⁶:

$$T[K] = -5134.86 / (\log_{10} N[\text{cm}^{-3}] - 23.35) \quad (4-11)$$

By varying the temperature of the sodium puddle and using Eq. (4-11) to determine the horizontal axis, the data shown in figure 4-4 was obtained. In order to compare our results to those of others, the line used for calibration was the D₂ line of the sodium doublet. One can see clearly the move from the doppler broadened line to a collisionally broadened line. Huennekens⁶ has generated a set of curves for the case of Na self broadened D₂ line, which are shown in figure 4-5. The curves in figures 4-4 and 4-5 do not correspond at the given absorption length used, indicating that the temperature measured on the outside of the sodium puddle is probably cooler than that inside the stainless steel cylinder (as should have been expected since on both sides of the puddle the temperature is kept hotter than the puddle and while the thermocouple itself acts as a mild heatsink). If there is an error of 20°C in the measurement of the temperature then the curves overlap completely. A further indication of the discrepancy is shown by the solid line in figure 4-4 which is the position and slope of the W_λ vs N curve at high values (collisionally dominated line shape region) of the atomic density obtained by using Eq. (4-8). A difference of 20°C would make the experimental points asymptotically approach the solid line.

Figure 4-4 Equivalent width versus atomic number density for Na-Na vapor cell. The number density was calculated by measuring the temperature of the sodium puddle and using the vapor pressure tables.

Figure 4-5 Equivalent width versus Na density for different cell lengths in cm. Graph taken from reference 6.

IV.B.2 LASER DETUNING DEPENDENCE IN Na-Na SYSTEM

Studies were made of the dependence of the magnitude of both the collisionally redistributed signal at D_1 and D_2 and the Rayleigh scattered signal on the laser detuning. The picosecond laser was tuned to the red side of the D_1 line and the spectrometer was used to view the scattered signal. The sodium number density was kept at $5 \times 10^{14} \text{ cm}^{-3}$ for all runs, indicating that the lines are almost completely dominated by collisional broadening. A representative trace is shown in figure 4-6. The area under the D_1 , D_2 , and Rayleigh components evident in this figure were measured and tabulated for different detunings.

The dependence of the Rayleigh scattering case is shown in figure 4-7. The circles indicate the raw data taken. Because of the geometry of the cell the ends of the input and output sapphire rods for the laser were visible from the spectrometer. Sodium vapor slowly attacked the sapphire creating pits in the ends of the rods that could scatter laser light into the spectrometer. Despite many efforts, the influence of this scatter on the measurements of the Rayleigh scattered intensity could not be eliminated. If one assumes that the amount of laser light scattered is constant with detuning and that the Rayleigh scattered component is very small at large detunings then the scattered component due to the pits on the ends of the rods can be measured and subtracted out of the signal. In order

Figure 4-6 Scattered signal from the Na-Na collision cell. The feature on the left is Rayleigh scattering from the input laser detuned 17 Å to the red side of the Na D₁ line. The Na D lines, populated due to collisional redistribution, are evident on the right.

Figure 4-7 Rayleigh scattered signal strength versus detuning from the D₁ line.

to take this scatter out, the data was replotted in a different form. If the signal varies as given by:

$$S = K\Delta^2 \quad (4-12)$$

then the signal with an additional background is given by:

$$S+B = K\Delta^2 + B \quad (4-13)$$

If one then plots Δ^{-2} vs $S+B$ then there should be a straight line fit with an intercept equal to the background scatter. This is plotted in figure 4-8 and indeed is a good straight line fit indicating a Δ^{-2} dependence of the Rayleigh scattering. The expected dependence of the Rayleigh intensity with laser detuning for resonance collisions is $\Delta^{-2.7}$

The magnitudes of the D_1 and D_2 lines do not suffer from the same problem of laser scatter interference due to the large detuning between the laser wavelength and the resonance line. The detuning dependence of the D lines is shown in figure 4-9. The slopes are also -2 as expected for resonance broadening although the signal becomes quite noisy at large values of the detuning. The reason that the ratio between the D_1 and the D_2 line is smaller than expected (about a factor of 2; see Table 1) is due to the presence of radiation trapping absorbing the D_2 line more efficiently than the D_1 line (see section IV.C.4).

Figure 4-8 Data from figure 4-7 replotted. Rayleigh scattered signal strength versus the inverse of the detuning squared.

Figure 4-9 D₁ and D₂ signal strength versus laser detuning from the D₁ line. The solid lines have a slope of -2.

IV.B.3 REASONS FOR DISCONTINUING STUDY OF Na-Na COLLISIONS

There were two main facts that indicated a need to discontinue the use of the Na-Na collision cell. Preliminary experiments performed on this cell suggested that a higher intensity pulse was needed together with a stronger output fluorescence signal. This required that the number density be increased, which was impossible using this Na-Na cell. The second was the extreme unreliability of the sodium vapor cell. The O-ring seals lasted about 4-5 days before they leaked and everytime there was a leak it took a full two days to slowly cool down, fix the seal, and burn out impurities that had leaked into the cell. Although experiments could have been done with this cell, the continual breaking of vacuum made the observed results somewhat unreliable.

IV.C Na-Ar COLLISIONS

IV.C.1 REASONS FOR CHOICE OF Na-Ar COLLISIONS

As was shown in section II.M, the magnitude of the collisionally redistributed signal is proportional to both the target atom number density and also the perturber atom number density. In the Na-Na collision case, the target and the perturber were both Na so in order to increase either population one had to increase both.

If one uses a different perturber, e.g. Ar, then its number density can be raised independently of the target atoms and thus increase the fluorescence signal. Computer simulations (such as those illustrated in section II.N) indicated that Na-Ar collisions with their R^{-6} interaction potentials could reveal the same optical collision dynamics as the Na-Na resonance collisions with R^{-3} potentials, albeit with a smaller signal and a smaller value of R_c . As a result, the move was made to a different cell which offered a much more reliable operation and an ability to raise the perturber atomic number density as high as needed to view the desired effects.

The schematic potential curves for the Na-Ar collision system are shown in figure 4-10. The ground 3S state is a $\sum_{\frac{1}{2}}$ state with the two excited state curves which shift to the red side of the resonance lines being π states. Another $\sum_{\frac{1}{2}}$ state from the D_2 line only shifts to the blue side of the resonance lines. The laser will be detuned to the red side of the D lines and since the $\pi_{\frac{1}{2}}$ state which comes from the D_1 line comes in contact with the dressed level at a larger value of R there will be more collisions involving this curve crossing. Thus we expect the D_1 line to be somewhat preferred over the D_2 line.

IV.C.2 CALIBRATION OF Na-Ar CELL

The calibration of the Na-Ar cell was made in a manner very similar to that of the Na-Na cell. The method of equivalent widths

Figure 4-10 Na-Ar potential curves. Graph redrawn from reference 9.

was used to determine the Na atomic number density versus the measured cell temperature. Argon buffer gas densities were measured using a Baratron pressure gauge.⁸ Through a knowledge of the vapor temperature in the illuminated region, the argon number density could be calculated using the ideal gas law.

The experimental arrangement used to make the equivalent width measurements was similar to that in figure 4-2 with the different cell installed. The length L of the Na vapor region was assumed to be equal to 10 cm, which is 60% of the length of the nickel wick at $L=10$ cm. By varying the temperature of the cell one can change the number density of the sodium while keeping the argon density relatively constant. The change in equivalent width with changing sodium number density is plotted in figure 4-11. The argon pressure was kept constant (to within the effects due to the ideal gas law) during this plot at $P_{Ar}=243$ Torr. One sees that the slope is constant which indicates that at this argon pressure the Na D lines are primarily collisionally broadened. One can compare this curve to the curves in figure 4-1. In this case the value of $a=1.43$ should give a number density of $8 \times 10^{12} \text{ cm}^{-3}$ at an equivalent width of $W_{\lambda}=1 \text{ \AA}$. Using Eq. (4-11) to determine the vapor pressure at the measured temperature, the density at a measured value of $W_{\lambda}=1 \text{ \AA}$ is at 2.5×10^{14} a factor of 30 higher. This indicates that the temperature measurement is too high by 80°C . This is a large factor but has been somewhat corroborated through other measurements. The Na-Ar cell has a large temperature gradient between the hot central section of the cell to the cool room

Figure 4-11 Equivalent width versus Na atomic number density for the Na-Ar collision system. The Ar pressure was maintained at 200 Torr. The Na atomic number density was calculated from the measured cell temperature and vapor pressure curves.

temperature windows. The difference in temperature between these two points is approximately 300°C. Using a stainless steel cell with its low thermal conductivity, measuring the temperature on the outside of the cell, and the large temperature gradient present could all contribute to this large discrepancy. All further discussions of the Na atomic number density will take this measured temperature difference into account. During all experiments performed on this cell, the equivalent width was measured to ensure an accurate knowledge of the Na density.

The variations in the D_1 and D_2 line fluorescence intensities with atomic number density have been measured. The result is presented in figure 4-12. The experimental configuration and technique is similar to that used in section IV.A. The argon pressure was maintained at 243 Torr and the laser detuning at 20 Å from the D_1 line. The data shows a slope of 1 for both the D_1 and D_2 lines. This is as theoretically expected from Eq. (2-38).

The variation in the equivalent width (W_λ) with changing argon pressure is displayed in figure 4-13. For these experiments the Na density was kept at $N=8 \times 10^{12} \text{ cm}^{-3}$. The variation in the value of W_λ is due to the changing value of the collisionally broadened linewidth and thereby the value of a in Eq. (4-2). At lower values of the argon pressure the value of a is less and one moves down the different curves of figure 4-1.

Figure 4-12 Sodium D line fluorescence signal strength versus Na atomic number density. The laser was detuned 20 Å from the D₁ resonance line. The Na atomic number density was calculated from the measured cell temperature and vapor pressure curves.

Figure 4-13 Equivalent width versus argon pressure.

IV.C.3 LASER DETUNING DEPENDENCE FOR Na-Ar COLLISIONS

The fluorescence intensities of the D_1 and D_2 lines were measured versus laser detuning. Since the interaction potential for Van der Waals collisions goes as R^{-6} the variation in line intensity with detuning⁷ should vary roughly as $\Delta^{-3/2}$. The actual absorption profile of the Na D_1 line when broadened by the effects of various perturbers is shown in figure 4-14. The line intensity dependence versus detuning is shown in figure 4-15. For this plot the argon pressure was set at 243 Torr and the sodium number density was $6 \times 10^{12} \text{ cm}^{-3}$. The two curves are for the intensity of the D_1 versus the detuning from that line and the intensity of the combined doublet versus the detuning from the center of mass of the doublet. Both curves show a slope of $-3/2$ as expected for Van der Waals collisions.

Measurements were not made of the detuning dependence of the Rayleigh scattered signal. This is because of the extremely small Rayleigh signal (when compared to the D_1 line intensity) generated in a strongly collisionally broadened system. The ratio of the intensity of the collisionally redistributed signal to the Rayleigh signal is given by:¹⁰

$$\frac{I_{\text{RAY}}}{I_{D_1}} \equiv \frac{A_{21}}{\Gamma} \quad (4-14)$$

For our experimental conditions the ratio is approximately $(250)^{-1}$

Figure 4-14 Measured absorption curves for the red side of the Na D₁ line when perturbed by four different perturbers. In our case only Ar was used as a perturber. Graph taken from reference 7.

Figure 4-15 Na D line signal strength versus laser detuning. The top curve is the sum of the D_1 and D_2 line strengths versus the detuning from the weighted average wavelength of the two lines. The bottom curve is the D_1 signal strength versus the detuning from the D_1 line. The solid lines have a slope equal to 1.5.

so the intensity of the Rayleigh component would be this fraction of the intensity found for the D_1 line. Experimentally this would be nearly impossible to view even if there was no laser scatter from the windows or other reflections contaminating the cell.

IV.C.4 EFFECTS OF RADIATION TRAPPING

When resonance radiation is emitted in the middle of an optically thick vapor, there is a finite probability (sometimes quite large) that the radiation will be reabsorbed before it escapes from the vapor cell. This effect, called either radiation trapping or radiation imprisonment, can lead to a loss of signal, scrambling of the emitted polarization, and an increase in the decay time of the observed fluorescence.

Due to reabsorption of the fluorescence photons, the effective decay rate Γ_{eff} decreases as:¹¹

$$\Gamma_{\text{eff}} = gA_{21} \quad (4-15)$$

where g is the average probability that a photon emitted from the center of the cell will escape without reabsorption. The value g is termed the "escape factor" and depends on the optical depth at line center, the geometry of the cell, and the type of broadening mechanisms present. Holstein and others¹²⁻¹⁹ have long attempted to evaluate the escape factor g theoretically as a measure of the

degree of radiation trapping, with success only on unrealistic or idealized geometries. For our particular cell geometry, there have been no known theoretical models made to describe the effects of radiation trapping.

AC-stark shifting of atomic lines has been used in the past to overcome the effects of radiation trapping.²⁰ When a laser pulse is used to address the atoms has a pulse duration which is as long as the decay time of the atoms, then the atoms fluoresce at the Stark-shifted wavelengths. Atoms which are outside the laser beam are not effected by the stark effect and therefore do not absorb the stark-shifted fluorescence. In our case, however, the laser pulse duration is much shorter than the natural decay time of the Na atoms and therefore the amount of ac-Stark-shifted fluorescence is negligible.

Experimentally there are three radiation trapping effects which occur and must be noted when interpreting the experimental data. The first is the loss of signal. When light is emitted from the center of the cell it must traverse a long cylindrical section of unexcited Na atoms (see figure 3-44). As it passes down this tube it can be absorbed by the ground state Na atoms present. At some later time the atom fluoresces in a completely random direction. This randomizing of the direction of propagation decreases the amount of signal collected in two ways. First, there is fluorescence that is trapped and reradiated in a direction such that it hits the wall of the cell and is lost. The second is that as it reradiates it propagates in a different direction and thus is not imaged onto the

slit of the spectrometer by the collection optics. Visually the sharp, crisp image of the fluorescence that is easy to image at low atomic densities turns into a blurred line that is impossible to completely collect without a severe loss of spectral resolution. This loss of signal is, of course, dependent on the absorption coefficient of the line being observed. Weaker lines do not absorb as well and have less of a problem with radiation trapping. One must be extremely careful, therefore, when comparing the intensities of two lines of different absorption cross section under experimental conditions which include radiation trapping.

The second effect which must be noted is the increase in the decay time of the emitted fluorescence. The time delay which occurs as the photons are absorbed and reemitted causes the observed decay time to increase by the factor g . Monte-Carlo techniques²¹ have been used to further describe this increase for small values of g (<10) and have indicated that the time dependence becomes decidedly non-exponential for values of g greater than 3. These effects have been clearly observed experimentally.^{18,22}

The third effect involves the scrambling of the emitted polarization by reabsorption of the emitted fluorescence. The reemitted photons can have a random polarization orientation compared to the absorbed photons. Therefore any polarization information is lost by the absorption-reemission sequence. Studies of the polarization of the emitted fluorescence must be carried out in an experimental arrangement which does not include the effects of radiation trapping.

Simple estimates of whether or not radiation trapping will be a problem can be made by calculating the peak absorption coefficient of the resonance line and calculating the Beer's absorption depth for different atomic densities. The absorption profile for a collisionally broadened line is given as:²³

$$\sigma(\omega - \omega_0) = \frac{4\pi^2 \omega d^2}{\hbar c} \frac{1}{\pi \Gamma_2'} \frac{1}{\Delta^2 + (\frac{1}{\Gamma_2'})^2} \quad (4-16)$$

The peak absorption crosssection can be evaluated and simplified to give:^{24,25}

$$\sigma(0) = \frac{3e^2 f}{mc^2 \Delta \nu_c} \quad (4-17)$$

where f is the oscillator strength of the transition and $\Delta \nu_c$ is the collisional half width in cm^{-1} . Using the values for Na and Na-Ar collisions found in Table 2-1, we calculate the peak absorption crosssection as $2.9 \times 10^{-12} \text{ cm}^2$ for the D_1 line and $5.8 \times 10^{-12} \text{ cm}^2$ for the D_2 line. By multiplying by the number density one can determine the absorption depth. This is plotted in figure 4-16. Indications are that in order to have an absorption depth of less than 1 for the D_2 line with the 5 cm path length of our cell a density of less than $3.4 \times 10^{10} \text{ cm}^3$ must be maintained.

In order to view the effects of radiation trapping two experiments were performed. As stated previously, the signal loss due to radiation trapping depends on the peak cross section, making

Figure 4-16 Line center absorption depth versus Na atomic number density of the Na D_1 line (bottom) and the Na D_2 line.

the ratio of fluorescence intensities between two lines of differing magnitude difficult when radiation trapping is present. The results in figure 4-12 were further manipulated by noting the ratio of the D_2 and the D_1 lines as a function of the sodium number density. This is displayed in figure 4-17. Since the ratio of the emission oscillator strengths is 2, one would expect the ratio of the intensities to also be 2 when radiation trapping is not present and the argon buffer gas is very dense. As is seen in figure 4-17, at densities below $6 \times 10^{10} \text{ cm}^3$ this is indeed the case. Above this density this ratio decreases as the D_2 line has a greater signal loss, due to its higher absorption cross section, than the D_1 line. Thus experiments done to monitor the ratio of the two lines should be done at low densities.

The second experiment performed was to measure the decay time of the fluorescence signal. Experimentally, this was somewhat difficult since the signal generated was so small that only single photon events were recorded. In order to measure the decay time a gated integrator was used to measure the total signal over the $1 \mu\text{s}$ gate width used. By delaying the gate with respect to the signal the decay time could be determined. The result is shown in figure 4-18 for the D_1 line fluorescence at a Na density of $2.2 \times 10^{12} \text{ cm}^3$. A relatively slow exponential decay is found with a e^{-1} decay time of 41 nsec. This indicates a value of $g=2.5$ (as given by Eq. (4-15)), lower than would be expected by considering the absorption depth of the D_1 line at this density. This could be partially due to the fact that fluorescence is emitted at all of the wavelengths in

Figure 4-17 Ratio of the signal strengths of the two Na D lines versus atomic number density. The ratio of the emission line strengths is 2. This value is only evident at low values of the Na number density. This plot clearly illustrates the effect of radiation trapping.

Figure 4-18 Measured fluorescence decay time from the Na D₁ line. A decay time of 41 nsec was measured at a Na density of $2.2 \times 10^{12} \text{ cm}^{-3}$. The natural decay time of Na is 16 nsec. This illustrates the effect of radiation trapping lengthening the decay time due to absorption and reemission of the resonance radiation.

the lineshape, with only the ones that occur at line center undergoing the strong reabsorption indicated by figure 4-16.

The effects of radiation trapping have been shown to be important in understanding experimental results taken in high-absorption-depth vapors. In experiments performed in the present work, where only the intensity dependence of the line versus the input laser is examined, the effects of radiation trapping do not alter the dependence -- rather they only alter the amount of signal obtained. In this case a high atomic number density can be used in order to increase the observed signal. If, however, the ratio of the D_2 to D_1 lines or the polarization is being examined, then the number density must be kept low to eliminate radiation trapping effects.

IV.C.5 EXPECTED SIGNAL ESTIMATES

Several attempts have been made to estimate the magnitude of the fluorescence signal expected (neglecting radiation trapping effects). Knowledge of the collisional broadening rates and the lineshape in the wings allows us to calculate the linear absorption coefficient far off-resonance, and from that the number of atoms excited per cubic centimeter.

McCartan and Farr⁷ have carefully measured the lineshape far in the wings of the sodium D lines. Their result for the red wing of the D_1 line is shown in figure 4-14 for sodium perturbed by various

noble gases. By using the value of the collisional broadening rate together with Eq. (4-16), the absorption cross section at detunings within the impact regime (small detunings) can be determined. Using the graph for Na-Ar collisions on figure 4-14, the cross section at larger detunings can also be determined.

Using the method described above, the absorption cross section at a detuning of 45 cm^{-1} has been calculated to be $9.88 \times 10^{-17} \text{ cm}^2$. This value for the cross section agrees well with the results of the numerical solution in Sec. II.N. A 2 psec pulse at 10^9 W/cm^2 has 6×10^{15} photons per pulse per cm^2 . Therefore, at a sodium density of 10^{12} cm^{-3} the excited state number is expected to be $5 \times 10^{11} \text{ cm}^{-3}$ while at a density of 10^9 cm^{-3} it is expected to be $5 \times 10^8 \text{ cm}^{-3}$. The number excited is half of the total density of atoms. This is unrealistic and indicated the inadequacy of using a linear equation to make signal estimates of a non-linear problem.

The collection efficiency of our detection system can be estimated to determine the estimated signal at our detector. With $f/18$ collection optics a factor of 1280 is lost. A factor of 2 loss through the spectrometer has been measured, while our photomultiplier tube has a quantum efficiency of 12%. Combining all of these the total collection efficiency from photons generated to photoelectrons produced is $(21000)^{-1}$. The focal region can be varied somewhat to increase the signal, however for a $55 \mu\text{m}$ spot at the focus of the input laser, a volume of $1.2 \times 10^{-4} \text{ cm}^3$ can be observed giving an excitation population of 6×10^7 . Given our collection efficiency

this means that on each shot 5000 photoelectrons should be generated. As will be seen, the method used to make the signal strength approximation is far from ideal, however it did give us confidence that the signal could be observed.

IV.D OTHER EXPERIMENTAL CONSIDERATIONS

Several details of the experiment which are important for its success should be completely described. These include the problems of ASE causing direct atomic line excitation, the different methods of data acquisition used, and the methods used to reduce the data.

IV.D.1 ABSENCE OF ASE-INDUCED FLUORESCENCE

Amplified spontaneous emission generated in the dye amplifiers can be a serious problem when performing off-resonance studies. Collision-induced fluorescence signals are already extremely weak, and ASE which is generated at on-resonance wavelengths can excite the atoms efficiently without the benefit of an optical collision. Eliminating any ASE is therefore a prime concern. In the dye amplifier configuration used, a saturable absorber is employed to absorb any ASE generated in the first stage. Proper adjustment of the absorber jet at focus is critical to ensure that the ASE is completely removed. There is little ASE generated in the second

stage (since there is a mild focus used there) and what is generated is not very directional. Two pinholes are used to aperture the beam and spatially clean any ASE away.

These two methods have eliminated any effects of ASE in perturbing the experiments. In order to demonstrate this, three experiments were performed to illuminate the presence of any ASE. The first was to block the laser input to the amplifier so that only ASE is generated. All fluorescence signal then disappeared. Therefore all the signal generated was entirely due to the ultrashort laser pulse. Second, a variation of the argon buffer gas pressure should only change the collisionally redistributed signal and not any direct line excitation. A linear dependence of the signal on the argon pressure was found experimentally as is expected from the optical collision theory (see in particular Eq. (2-38)). And third, the variation with laser detuning was measured. Direct line excitation due to ASE would not cause a decrease in signal with increased detuning, however this decrease was observed. Given all of these results, ASE causing excitation without the benefit of a collision seems to have been ruled out as an experimental difficulty. This is the most sensitive probe used to measure the amount of ASE generated and illustrates again the superior qualities of the synchronous dye amplifier chain in producing a clean, noise-free, pulse for experiments.

IV.D.2 CONFIRMATION OF STABLE PICOSECOND PULSE PROPAGATION

As demonstrated by Nakatsuka and Grischkowsky,²⁶ when a near-resonant picosecond pulse propagates through an absorbing medium it can be reshaped and distorted due to the group velocity dispersion (GVD) and attenuation caused by the vapor. We experimentally examined this effect for the cell and vapor pressures used for our experiments. It was found not to have an effect on the 1.6 psec pulse used. There are two reasons for this. First, the path length of our cell is 10 cm rather than 50 cm used by Nakatsuka and Grischkowsky. Second, our atomic number density is 10^3 times less than theirs, decreasing the GVD by this factor.

IV.D.3 METHODS OF DATA ACQUISITION AND REDUCTION

The experimental arrangement used to measure the optical collision signal dependence on the input laser intensity is shown in figure 4-19. The laser pulse passes through a set of crossed polarizers with a half-wave plate between them. Rotation of the wave plate varies the amount of pulse energy transmitted by the second polarizer and therefore focussed into the cell. The second polarizer is oriented such that transmitted light is vertically polarized. This polarization was used exclusively for all of the experiments performed. The transmitted pulse energy is monitored by using a 4%

Figure 4-19 Experimental configuration used to measure the fluorescence intensity dependence of the input laser intensity. The laser pulse is passed through a pair of crossed polarizers with a half-wave plate between them. By rotating the half-wave plate the amount of light transmitted through the second polarizer could be modulated. The energy of the input laser pulse was monitored by using a 4% pickoff mirror focussed on a calibrated photodetector. The laser pulse is focussed into the Na-Ar oven with the fluorescence at the focus imaged out of the cell onto the slit of a monochromator. The fluorescence intensity at a given scattered wavelength was then measured using a photomultiplier tube.

splitter to send part of the beam to a calibrated photodiode. The output of the photodiode is sent to the computer so that for each laser shot the exact energy can be recorded. The laser beam is then focussed into the center of the vapor cell using various lenses. Different lenses are used to adjust the spot size at focus and thereby the input laser intensity in the experimental region. Spot sizes were accurately determined by measuring the diameter of the beam at focus with a $3\text{ }\mu\text{m}$ pinhole.²⁷ This pinhole is raster scanned using two orthogonal stepper motor driven translators. These stepper motors are controlled by computer with the intensity of the transmitted light also measured using a photodiode interfaced to the integrating A/D. A contour plot of the resulting scan is shown in figure 4-20. The beam is remarkably smooth and symmetric with a slight amount of astigmatism due to the focussing of the beams in the dye amplifier through slightly tilted dye cells. The beam is estimated to be no more than 10% greater than diffraction limited. Knowledge of the pulse duration, the spot diameter, and the pulse energy all combine to allow an accurate recording of the laser input intensity for each shot.

The fluorescence is imaged onto the slit of a .3m spectrometer used in conjunction with a high speed, high gain photomultiplier tube²⁸ to measure the fluorescence intensity at any wavelength. The output of the PMT is sent to the A/D with the amount of charge generated being proportional to the amount of scattered light at the given wavelength.

Two methods were used to determine the dependence of the

Figure 4-20 Contour plot of the amplified dye beam profile measured by raster scanning a 3 μm pinhole across the laser beam at the focus of the lens used to focus the beam into the Na-Ar oven. The contours are separated by 10% of the peak laser intensity.

fluorescence intensity upon the input intensity. The first was to fix the laser intensity (by accepting only those shots which fell within a given energy range) and scan the spectrometer wavelength to observe the scattered light. By measuring the area under the D_1 , D_2 , and Rayleigh components the scattered intensities at each laser input could be obtained. This technique was used for the first experiments, however it proved to be extremely time consuming. The second method overcame this by carefully fixing the spectrometer to only look at the peak value of the D_1 , D_2 or Rayleigh component which is proportional to the area under the component. A large 2-dimensional histogram was then created in the computer's memory. For each laser shot 1 was added to the bin which represented the measured fluorescence intensity (for the line of interest) at the measured laser pulse energy. One hundred fifty thousand shots were taken (2 1/2 minutes at the 1 kHz repetition rate) for each run which allowed for good statistics to be obtained. After each run the number of shots and the average fluorescence intensity were calculated for each laser energy by examining the 2-dimensional histogram. The average value and the number of shots for one representative run are shown in figures 4-21a and 4-21b. The number of shots was then used with a threshold value to accept only those results which had enough laser shots to generate the required statistical significance. Generally 500 shots per laser energy "bin" were required. The accepted data that is recorded is shown in figure 4-21c. In order to test whether this technique introduced any non-linear dependence, the intensity of scattered laser light was

Figure 4-21 Data reduction method used. The data shown in a.) is the average value of the fluorescence intensity at each value of the laser input intensity. The data in b.) is an energy histogram of the input laser intensity. The energy distribution is spread out due to the rotation of the half-wave plate between the crossed polarizers. The data in c.) is the reduced data. In order to reduce the data a threshold number of shots per laser energy unit was determined (usually 500 shots) to give satisfactory signal statistics. Average fluorescence values are only accepted if the number of laser shots in the particular laser energy unit was more than this threshold.

measured over the full range of the histogram and was found to be linear (as it should be) with input laser intensity.

At lower values of the atomic number density the fluorescence signal became so small that photon counting proved to be the desirable method of data acquisition. The particular photomultiplier used²⁸ could not adequately resolve the difference between one and two photoelectron signals however it could distinguish the difference between zero photoelectrons and one photoelectron. Figure 4-22 illustrates a histogram of the charge output from the PMT for 32000 consecutive laser shots. In figure 4-22a a large zero charge component due to a laser shot which generates zero photoelectrons is evident. By eliminating this zero component the one photoelectron component becomes visible as seen in figure 4-22b. Knowledge of the gain of the PMT and the sensitivity of the A/D further corroborate the fact that this is indeed a one photoelectron event. Two photoelectron events could also be observed as a second "bump" in the histogram of the charge output however they were not distinct enough such that one could clearly determine whether there was a one or two photoelectron event.

If the probability of obtaining one photoelectron of signal on a given laser shot is 20% then the probability of obtaining a laser shot with two photoelectrons generated can be assumed to be 2% (a Poisson photoelectron distribution is assumed). Thus an error of 2% is introduced if one can only detect zero or one photoelectron events. This error occurs since all two photoelectron events are assumed, using this method, to be one photoelectron events. This is

a.)

b.)

Figure 4-22 Charge histogram from the photomultiplier tube. The histogram in a.) illustrates the large number of laser shots with zero photoelectrons generated. If one removes these shots and rescales the data then one can view the charge distribution due to a one photoelectron event as seen in b.).

clearly seen in figure 4-23, where the photon-counted signal is presented for three different fluorescence intensities changed by adding ND filters in front of the spectrometer. The optical density of an NG 11 is about 2, but one can see that this factor is not found in the results. At the peak value of the 0 ND trace 45% of the laser shots have a photoelectron event. Therefore 10% of the shots have 2 photoelectron events which are assumed to be 1 photoelectron events. The difference between the NG11 curve and the NG5 curve should also be a factor of 2 and it is, indicating that at this value of photoelectrons per shot the chance of getting a "2 photoelectron error" is reduced. This was the criterion used to determine whether the photon counting technique could be used.

By inspection of figure 4-22b a threshold could be set to determine whether a one photoelectron or zero photoelectron event had occurred. For the photon counting case, two one-dimension histograms were used: one was used to count the number of photoelectrons generated at each input laser energy, while the other was used to count the number of shots at each laser energy. After a run of one hundred fifty thousand shots the number of photoelectrons in each bin was divided by the number of shots in each bin to determine the average number of photoelectrons generated per shot for each input laser energy. The number of shots in each bin was again used to choose the acceptable data given the statistical requirements.

By using the three methods of data acquisition in determining the intensity dependence (scanning the wavelength at a fixed energy, fixing the wavelength and measuring the fluorescence intensity, or

Figure 4-23 Photon-counted signal from the D₁ line versus input laser intensity for 1.6 psec pulses. The output fluorescence intensity was altered by placing a NG11 (approximately a factor of 2) or NG5 (approximately a factor of 4) filter in front of the spectrometer to view the linearity of the photon-counting process. Photon counted signals appear to be acceptably linear if the signal is less than .2 photoelectrons generated per laser shot.

fixing the wavelength and measuring the number of photoelectrons generated) all the experiments could be carried out. Signals as small as 1 photoelectron every one hundred shots were observed for a variety of different intensity ranges, argon pressures, sodium pressures, and laser detunings.

IV.E INTENSITY DEPENDENCE RESULTS

Studies were made of the dependence of the input laser intensity on the observed fluorescence from the D_1 and D_2 lines for different sodium densities, argon pressures, laser pulse widths, and laser detunings. Because of the extremely small signal generated great care was taken to rule out competing excitation mechanisms and optimize the signal levels. These were described in the previous section. In this section we present the principal experimental data obtained in this study.

Theoretical estimates were made in estimating the shape and magnitude of the collisionally redistributed signal for different input laser intensities. These indicated that I_{crit} for the case of Na-Ar collisions 20 Å detuned would have a value of 5 MW/cm², which for psec pulses is relatively easy to obtain. By cavity dumping the dye oscillator one could achieve the required dye pulse energy while separating the time between pulses sufficiently to discount the effects of optical pumping. The operation and output characteristics of the cavity-dumped dye laser are described in

detail in section III.C.3. These pulses at 5 MHz were applied to the Na-Ar collision system. Seven-hundred femtosecond pulses were detuned 20 Å to the red side of the D₁ line and used with a peak intensity of 35 MW/cm² when focussed with a 330 mm lens. A Na density of 4x10¹¹ cm⁻³ was maintained with an argon density of 4x10¹⁸cm⁻³. The spectrometer slits were opened to 300 μm to collect as much light as possible with a 3 Å resolution. The result is shown in figure 4-24 for the cases of the D₂, D₁ and Rayleigh scattered signal. As shown in the figure there is no rollover of the scattered intensity at the highest values of the intensity, for either the D₁ or the D₂ components. The intensity ratio of the two lines indicates that there is severe radiation trapping as would be expected at these Na densities. We believe that the "Rayleigh" scattered component in this case is not Rayleigh scattering at all but rather laser scatter off of the windows of the vapor cell. Neither higher energies nor shorter pulses (either would increase the input laser intensity) were obtainable using the cavity-dumped dye laser. These results indicated that higher pulse energies would be required to view the rollover of the fluorescence intensity at intensity values greater than I_{crit}. The repetition rate was reduced from 5 MHz to 1 kHz in order to use the high repetition rate dye amplifiers with a comparable loss of average power and signal to noise ratio.

The second set of experiments were done at 1 kHz using the dye amplifiers to produce an input laser pulse focussable to much higher intensities than that available with the cavity dumped output. The

Figure 4-24 Fluorescence intensity of the D₁ and D₂ lines versus input laser intensity; low power.

output of the dye oscillator (used without the cavity dumper installed) was amplified in the dye amplifier chain producing outputs of up to $1 \mu\text{J}$ in a 1.6 psec pulse. Various lenses were used to vary the intensity in the cell. Sodium densities were kept at $1.8 \times 10^{13} \text{ cm}^{-3}$ with the argon densities at $4 \times 10^{18} \text{ cm}^{-3}$. The fluorescence output was measured by scanning the spectrometer in wavelength while keeping the laser energy constant and measuring the area under the D_1 , D_2 and "Rayleigh" components. With the small average power available out of the dye amplifier ($< 1 \text{ mW}$) it was necessary to accumulate data for 10 minutes (600,000 shots) to obtain each statistically significant data point. The results are compiled in figure 4-25. The data is relatively linear up to approximately 10^9 W/cm^2 where one begins to observe a rollover. The intensity at which this occurs is 200 times higher than predicted and is near the end of the intensity range available in this data. In order to fully characterize the effects, higher intensities were still needed.

Further development of the dye amplifier chain led to an increase in the output energy from 1 to $10 \mu\text{J}$. This energy increase, together with tighter focussing optics allowed us to obtain laser intensities of greater than 10^{11} W/cm^2 . In addition, high-speed software was written to allow us to fix the wavelength seen through the spectrometer to the peak of either the D_1 or D_2 line and vary the intensity as described in the previous section. These advancements allowed the experiments to be performed for the entire available intensity range for one line in $2 \frac{1}{2}$ minutes.

The variation in fluorescence intensity for three different

Figure 4-25 Fluorescence intensity of the D_1 and D_2 lines versus input laser intensity; intermediate powers. The data in this curve was taken in a slightly different manner than the others. For each point many wavelength scans of the spectrometer were made to measure (by noting the area under the D_1 or D_2 lines) the signal strengths. This proved to be an extremely time consuming method and was abandoned for other intensity dependance measurements.

values of the laser pulse width are shown in figure 4-26. For this case the Na density was kept at a value of $4 \times 10^{12} \text{ cm}^{-3}$ while the argon density was set at $4 \times 10^{18} \text{ cm}^{-3}$. The y axis is defined in units of single photoelectrons per shot illustrating the extremely small signals. The photon counting techniques described earlier were not used here, but rather the charge output that is related to the fluorescence intensity was divided by the mean charge output for a one photoelectron event as determined by figure 4-22.

As predicted by the theory the output at a given intensity value is much higher for longer pulses than for short pulses due to the difference in the number of photons present in the two laser pulses. For this reason the fluorescence signal was divided by the pulse width for each curve and the results are plotted together in figure 4-27. This figure demonstrates for the first time the difference in absorption efficiency for "short" pulses versus "long" pulses. In the Na-Ar case the time between curve crossings is about 2.5 psec so a 1.6 psec pulse is a "short" pulse whereas the 8.5 psec and 51 psec pulses are "long" pulses. Because a 300 μm slit width was used to collect as much fluorescence as possible and due to the smearing effect of radiation trapping, the signal was spatially averaged over the entire beam profile. This caused the loss of the dramatic difference between the "short" and "long" types of pulses as discussed in section II.0 and illustrated in figure 2-35.

These experiments confirm the theoretical predictions that pulses which are "short" do indeed have a higher absorption than pulses which are "long". By comparing figures 4-27 and 2-35 we see

Figure 4-26 Fluorescence intensity of the D₁ line versus input laser intensity for three values of the pulse duration; high power.

Figure 4-27 Fluorescence intensity of the D_1 line divided by the laser pulse duration versus input laser intensity for three values of the pulse duration. The fluorescence intensity was divided by the pulsewidth to eliminate the bias caused by the greater number of laser photons in a longer pulse at the same intensity as a shorter pulse. These results are in qualitative agreement with the theoretical predictions in Figure 2-35.

that the intensity at which this difference is observable is, however, at a much greater intensity than is predicted theoretically. This could be due to the effect of temporally and spatially averaging, a lack of a full description of the potential curves, or the existence of multiple (rather than a single) excited states.

The ratio of the intensity of the D lines remained constant over this intensity range although affected by radiation trapping as illustrated in figure 4-28. Quantitative measurements of the value of D_2/D_1 were impossible due to the undetermined effect of radiation trapping on the two lines.

Other measurements were made of the rollover dependence on argon pressure and detuning. As illustrated in figure 4-29 the D_1 signal dependence on argon pressure was linear even at the highest intensities. This further demonstrated that the rollover effect was truly due to an optical collision rather than direct line excitation as discussed in section IV.D.2. The variation with laser detuning for the "short" pulse case and the "long" pulse case is shown in figures 4-30 and 4-31 respectively. In the "short" pulse case the signal does not decrease as $\Delta^{-3/2}$ as theoretically predicted. This could be due to the fact that at higher detunings the time between curve crossings (T_c) gets shorter and a 1.6 psec pulse moves from being a "short" pulse to being a "long" pulse. For the "long" pulse case the detuning dependence is closer to that theoretically predicted ($\Delta^{-3/2}$) since a "long" pulse is always a "long" pulse in this case.

Figure 4-28 Fluorescence intensity of the D₂ and D₁ lines versus input laser intensity; high power. The pulsewidth in this case was 1.6 psec.

Figure 4-29 Fluorescence intensity of the D_1 line versus input laser intensity for different values of the argon buffer gas pressure. The data has been replotted to illustrate the linearity of the signal with argon pressure. The fluorescence intensity versus argon pressure is plotted for three values of the laser intensity on the next page.

Figure 4-30 Fluorescence intensity of the D₁ line versus input laser intensity for different values of the laser detuning from the D₁ line. The data has been replotted on the next page versus laser detuning for two different laser intensities. The solid lines have a slope taken from the absorption data found in figure 4-14. In this case the laser pulse duration was maintained at 1.6 psec.

Figure 4-31 Fluorescence intensity of the D_1 line versus input laser intensity for different values of the laser detuning from the D_2 line. The data has been replotted on the next page versus laser detuning for two different laser intensities. The solid lines have a slope taken from the absorption data found in figure 4-14. In this case the laser pulse duration was maintained at 8.5 psec.

Studies were also made at low values of the Na vapor density in order to view the fluorescence without the detrimental effects of radiation trapping. The sodium number density was lowered to $9 \times 10^{10} \text{ cm}^{-3}$ where radiation trapping was shown not to be a factor (see section IV.C.4). At this density the ratio of D_2 to D_1 and the fluorescence emission polarization were observed. Since the signals were extremely small at the low sodium density, photon counting techniques described earlier (section IV.D.4) were employed. The pulse widths used in this study were "short" with a temporal duration equal to 1.6 psec. The result obtained for the ratio of D_1 to D_2 , as shown in figure 4-32, was surprising since it indicated that the ratio was approximately 1 even without radiation trapping effects. Havey et al.²⁹ measured the ratio to be 1.4 at this argon pressure with a CW laser. This may indicate that there is some state selection made in using picosecond optical collisions to excite the D lines.

Figure 4-32 Fluorescence intensity of the D₁ and D₂ lines versus input laser intensity at very low Na number density to eliminate any effects due to radiation trapping.

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CHAPTER V

CONCLUSION

In this thesis the results of the first study of optical collisions using a picosecond laser has been presented. During this study several major advancements in the development of laser sources and in the understanding of atomic collision physics on an ultrashort time scale have been made. These advancements will be reviewed here.

The development of the first Nd:YAG-pumped dye oscillator provided several advantages over previous methods of producing picosecond optical pulses. These include a more stable and rugged pump source, improvements in the amplitude stability, and an easier form of short pulse amplification. Improvements in the dye oscillator since this first demonstration have reduced the pulsewidth still further to 80 fsec where it remains the shortest pulse ever produced with a synchronously pumped laser. During this thesis work the ideas of synchronous amplification have been formulated, developed, and proven experimentally. The synchronous amplifier offers a method of highly efficient short-pulse amplification at high repetition rates with exceptionally low values of noise and amplitude fluctuations.

Theoretical models have been developed to further understand and predict the interaction of picosecond optical pulses and colliding atoms. This theory included effects due to realistic pulse shapes and beam profiles which have been shown to have a major impact on the intensity dependant behavior. Following the proposal of Lee and George,¹ a study of the variation of the cross section with "long" as opposed to "short" laser pulses was carried out. "Long" laser pulses have been defined as those whose duration is longer than the time between curve crossings, whereas "short" laser pulses are defined as being shorter than this time between curve crossings. Lee and George predicted an enhanced cross section for "short" laser pulses, which was theoretically verified using our theory (see Figure 2-35), albeit with a reduction in the magnitude of the effect when one uses realistic pulse shapes.

Using the new laser source the first study of optical collisions with picosecond pulses has been performed. Despite extremely weak signals, the pulse-width dependant intensity variation proposed by Lee and George and theoretically confirmed in this work has been experimentally verified (see Figure 4-27). Thus the first laser interactions with single curve crossings (or half a collision) have been observed. Several other experiments were also performed to understand the interaction of atoms and picosecond pulses. These preliminary experiments may have indicated that picosecond pulses can be used to perform state selection due to their ability to interact and modify the atomic collision dynamics during only part of the collision (see Figure 4-32).

Although the cross section dependance on "long" versus "short" pulses was indeed confirmed, the electric field strength needed was 15 times higher than that theoretically predicted. This is probably due to the effect of multiple excited-state levels, as first pointed out by Light and Szöke.² The excited-state levels of the sodium $3P_{1/2}$ and $3P_{3/2}$ levels each correspond to π molecular states which intersect the laser-dressed level. These levels are doubly degenerate levels which split under application of intense electric fields. Thus, there are four levels with which the dressed level can interact. Light and Szöke showed that, due to the effect of multiple levels, the fall off of the cross section at high laser intensities was not as fast as predicted by the Landau-Zener model (as well as the Lee and George model and our model). With the strong effects of temporal and spatial averaging present, the difference between "long" and "short" pulses could not be observed until even higher electric field strengths were applied. Besides the effects of multiple excited state levels, indications have been found that the shape of the Na D_1 line perturbed by Ar does not have a true R^{-6} potential shape.³ Since the shape of the curve plays a role in determining the probability of jumping across an avoided crossing, this could also cause a difference in the observed position of the non-linearity.

V.A FUTURE DEVELOPMENTS

Work performed as part of this thesis has indicated the directions that need to be explored in the future both in the areas of laser source development and atomic collision physics. Some of these new directions will be explained in more detail.

The development of the new cw-pumped regenerative amplifier has provided an intense source of $.532 \mu\text{m}$ pulses at high repetition rates. These pulses can be used to pump a synchronous dye amplifier. The principles of the regenerative amplifier do not prevent the adaptation of these techniques to much higher repetition rates. Currently the repetition rate is limited by the acoustic wave generation in the triode-driven pockels cell. If this engineering difficulty can be overcome the completely variable repetition rate source could be used for many new experiments. As one increases the repetition rate beyond the reciprocal of the storage time of the lasing medium, then the average power will remain the same while the pulse energy will decrease. For experiments such as those performed in this thesis, the pulse energy could be controlled not by introducing attenuation to the laser beam, but rather by increasing the repetition rate of the regenerative amplifier. The decreased output of the regenerative amplifier then will reduce the output of the dye amplifier as well. Although the pulse energy will decrease the repetition rate will increase, giving the same average power for the experiments and the same signal-to-noise characteristics as well.

By changing the lasing medium in the regenerative amplifier from Nd:YAG to Nd:YLF several other problems could be overcome. A large loss in the regenerative amplifier cavity is caused by the thermally induced radial birefringence in the laser rod. Nd:YLF has a much lower figure for this birefringence.⁴ In addition the stored energy in a Nd:YLF rod can be higher giving higher pulse energies in the same configuration.

Atomic experiments done with lasers generally need a stringent requirement on the wavelength used. Sodium has been the material of choice to study not only because of its single outer electron (which is easier to theoretically interpret) but also because the sodium D lines lie conveniently in the rhodamine 6G lasing range. Other more suitable atomic species should also be studied, however, to eliminate any bias associated with sodium and to attempt to study more complicated multiple electron atoms. New picosecond and femtosecond lasers do, however, have limited ranges of wavelengths available where stable, short pulse operation is possible. Attempts have been made recently to solve this problem.⁵ These involve producing enough energy in an ultrashort pulse to generate a white light continuum, using an interference filter to mask out the desired wavelengths, and then amplifying these wavelengths in a second dye amplifier chain. Although experimentally extremely challenging, this scheme has been used experimentally on semiconductor samples and could also be applied to atomic systems.

New vapor cell designs are required to overcome several experimental difficulties. Resonance collisions are desirable to

study, for they offer longer range collisions where a short pulse could influence a smaller part of the collision. Radiation trapping causes a loss of information and signal when using dense atomic vapors. Attempts have been made previously, as well as in this thesis work, to build cells which allow resonance collisions to be observed while reducing radiation trapping effects.^{6,7} None of these has proven to be reliable and reproducible.

If one could enhance the fluorescence signal without introducing radiation trapping effects, studies could be done with reduced argon pressures. When one reduces the argon pressure, the time between Na-Ar collisions can be increased to greater than the fluorescence time. This can then give a better view of state selection and polarization ratio without the smearing effect of multiple collisions before the atoms fluoresce.

Other perturbers could be used instead of Ar in studying Na-noble gas collisions. The element Xe is particularly appealing since the Na-Xe collision induces a level shift with a C_6 coefficient much larger than with Ar. Longer range collisions could therefore be studied even with a van der Waals collision rather than a resonance collision.

By synchronizing a white light continuum pulse to an atomic excitation pulse further understanding of the excited state population motion could be obtained. A white light pulse could be used as a probe a variable time after a pump pulse which is used to excite the atoms to the 3P level. The probe would then excite the atoms to even higher lying levels, such as the 4S or 5S. Knowledge

of the shape of the 3P potentials and the 4S potential would allow the motions of the excited colliding atoms to be examined directly. By noting the wavelength-dependant absorption of the white light pulse by these excited 3P atoms a more complete understanding of the collision dynamics could be obtained.

The use of super-cooled atoms at extremely low velocities⁸ together with picosecond pulses could allow study of the dynamics of these slow atoms. In addition the very slow velocities could allow one to probe, using a picosecond pulse, during a curve crossing to further understand the physics behind this fundamental process in collision theory.

The use of vapor cells allows high densities to be obtained, however with a loss of control of the velocities or geometry of the collision. Further studies could be made using atomic beams to carefully select several parameters of individual collisions. Picosecond optical collisions could then be used to study the dynamics of these selected collisions.

The work described in this thesis is only the first study of the interaction of picosecond pulses with colliding atoms. Our studies have shown that this is indeed a rich area to explore for atomic physicists. The use of picosecond pulses creates new challenges for the experimenter in terms of the laser source and the fluorescence collection techniques used. However, the ability to interact with single rather than multiple collisions offers a method to more clearly understand collision physics. It is hoped that this work will inspire others to further investigate the effects of ultrashort pulses on atomic systems.

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