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알칼리 원자의 해석적 결맞음 제어에 관한 연구

Analytical coherent controls of Alkali atoms in strong field regime

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A thesis submitted to the faculty of KAIST in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Department of Physics . The study was conducted in accordance with Code of Research Ethics¹.

> 2010. 11. 25. Approved by Professor Ahn, Jaewook [Advisor]

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알칼리 원자의 해석적 결맞음 제어에 관한 연구

이상경

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ABSTRACT

The outcome of quantum-mechanical dynamics can be steered by application of light field shaping, and this concept is called "Coherent Quantum Control." Since Silberberg's seminal work of the coherent control of two-photon absorption process of atomic Cesium, many studies on the coherent control of multi-photon processes have been attempted in more complex systems. But these studies considered multi-photon processes in weak-field regime, where the energy level structure remains unchanged during the light-matter interaction. Recent studies are focused on coherent control in strong-field regime. The strong-field effects in a multi-photon process, such as dynamic Stark shift and power broadening, prevent efficient coupling of matter with an intense femtosecond laser field. Especially, the dynamic Stark shift is crucial to optimize multi-photon processes.

Hence we have developed a new coherent control method, in which the dynamic Stark shift is successfully compensated. In this thesis, we present experimental demonstration of the newly devised analytical coherent control method, in which the laser-induced energy level change is compensated by the frequency-shift of computer-programmed shaped laser pulses. In the newly developed analytical coherent control, laser pulses are shaped as a polynomial sum of frequency and/or time in an acousto-optic programmable dispersive filter. For the given light-matter interactions, the model Hamiltonians developed in the strong-field regime are analytically studied and the derived excitation probability formulas given as analytical solutions are compared with the experimental results.

In experiments with atomic Cesium, we have demonstrated the analytical coherent control of the two-photon absorption in a dynamically shifted energy level structure. We have obtained the transition probability of the two-photon broadband excitation as a function of pulse shape parameters, frequency detuning, and laser intensity, by solving the two-photon two-level model Hamiltonian. First, we have found out that transform-limited pulses are not optimal in the strong-field regime, and certain linearly chirped pulses with a lower peak intensity enhance the given non-linear process. Further, spectro-temporal laser-pulse shaping is applied up to the third order chirp to preserve the resonant absorption condition during the intense laser-field interaction. Experiments carried out in the strong-field regime of two-photon absorption reveal that the analytically obtained offset and curvature of a laser spectrum compensate the effect of both static and dynamic energy shifts of the given light-atom interaction.

Lastly, the analytical coherent control method for a 2+1 multi-photon energy ladder system in atomic Sodium has been developed. We have coherently controlled the interference among sequential and non-sequential excitation paths from the ground 3s state to the 7p state in the strong-field regime. Both analytic formula and experimental results show that a negatively chirped pulse enhances the 7ppopulation because the sequential path is opened by a negatively chirped pulse. We expect that our study gives the possibility to realize quantum logic gates in multi-photon absorption system.

Keywords: Multi-photon absorption, Coherent control, Alkali atom

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Chapter 1. Introduction

Nature is not only the object of observations, but also the object of control. After appearance of quantum mechanics, many observed phenomena which can not be explained by classical mechanics have been successfully described by quantum mechanics. At the same time, control of quantum dynamics has attracted public attention. One can understand an unknown quantum system deeply and also obtain desired outcomes of quantum mechanical processes by control of quantum dynamics. The desire for control of quantum system led to the appearance of 'coherent control'.

The term 'coherent control' refers to the idea of coherent manipulation of quantum states. The state of an object is controlled quantum mechanically, by altering the dynamics of the amplitude (probability distribution) and the phase (quantum coherence) of the quantum wavefunction. The coherent preparation of light has been considered relatively easy compared to that of matters, and therefore, in a light-matter interacting system, the quantum states of the matter is controlled by controlling the coherent nature of interacting light. In this regards, both the laser development and the light control itself have played important roles in the the study of "coherent control". Lately, ultrafast lasers, which produce coherent superposition of broadband frequency components in optical region, are widely used in various settings of coherent control experiments. For example, the optimization problem of various light-matter interactions has been tried with shaped laser pulses that are prepared in special waveform designed either by solving the Schrödinger equation or by feed-back adaptation.

In recent decades, a variety of coherent controls, for example, quantum control, ultrafast optical control, or optimal control methods, have been developed to change a quantum-mechanical system towards desirable final states in a more systematic fashion. One of the important problems in coherent control is the selective bond breaking of molecules. One of the early approaches to the selective bond breaking was the use of tunable narrow-band lasers. The high intensity laser field of which the frequency is resonant with one of the vibration frequencies of molecules was used to produce selective molecular fragments [1]. Unfortunately, the intra-molecular vibrational redistribution (IVR)¹, which occurs in a sub-picosecond time scale, prevents selective bond breaking. As a result, this simple idea of coherent control failed, and alternative coherent control schemes have been required.

Two different coherent control methods have been proposed in 1980's. One is Brumer-Shapiro method, which is based on the control interference among the several quantum passages towards a desired target state proposed in 1986 [3]. In this method, the final-state probability can be controlled via constructive or destructive interference between the excitation passages, by adjusting the relative phase between two CW (continuous wave) lasers, which was experimentally demonstrated in 1990. The other scheme proposed by Tannor, Kosloff, and Rice in 1985 [4], known as Tanner-Kosloff-Rice method controls the time-evolution of a wavepacket. The wave packet is induced by an ultrashort laser pulse on the excited potential energy surface (PES). After the free evolution of wave packets, the molecule arrives at a certain position which prefers the desired reaction outcome. Then, a second ultrashort laser pulse transfers the wavepacket to the desired state, and therefore the time delay between the two pulses is the control parameter. This control scheme was experimentally demonstrated in early 1990's by the groups

¹IVR is known as the phenomena that the energy for breaking the certain bond of a molecule is redistributed to many eigenstates of the molecular system [2].

of Gerber and Zewail. These two schemes were operated in simple molecules whose Hamiltonians are known.

With the development of the femtosecond laser, many studies have been attempted to use various degrees of freedom of a femtosecond pulse: the complex pulse shaping technique has been developed. In complex systems like huge molecules, it is not possible to obtain the full Hamiltonian, nor to describe the time evolution of wave packets. Without a priori information of the system Hamiltonian, it is not possible to design the optimal pulse shape for the desired state manipulation. To overcome this difficulty, Judson and Rabitz proposed the closed-loop coherent control method in 1992. [5] This method uses a feed-backed learning algorithm to provide iteratively different electric light fields, towards an improvement of the predetermined outcome. After many iterations, the optimal electric field can be obtained. Because this scheme needs a complex pulse shaping technique, the laser pulses were programmed either by liquid crystal arrays or acousto-optic modulators, placed in frequency domain. In a complex pulse shaping example, for each frequency component of a laser pulse, not only the amplitude and phase but also the polarization states were programmed [6].

Gerber and coworkers first demonstrated the closed-loop control experiment on the photo-dissociation in 1998 [7]. Also, in the same year, Meschulach and Silberberg used the complex pulse shaping for manipulating two-photon absorptions [8]. These two experiments were followed by many coherent control experiments that were performed using similar methods. This novel concept of ultrafast coherent control has been applied to, for example, the optimization of nonlinear processes(e.g. second-harmonic generation, third-harmonic generation [9, 10], and multi-photon absorption [8, 11, 12, 13, 14]), nonlinear Raman spectroscopy (CARS) [15], *in-vivo* fluorescence microscopy [16], frequency comb [17], and the manipulation of electronic wavefunction [18].

In particular, ultrafast coherent control in multi-photon absorptions has been studied widely in the weak-field regime where the energy level structure of the matter remains unchanged. An analytic form obtained from the perturbation theory has well described multi-photon processes with pulses of π phase step, sinusoidal phase [8, 11, 12, 13, 14, 17], and linear chirping [19, 20, 21]. For the enhancement of multi-photon processes, the strong field are required. In the strong field regime, strong-field effects, such as the dynamic Stark shift and the power broadening have to be considered. It is now well known that, in general, the control schemes devised in the weak-field regime are not directly applied for the strong field coherent control, although the partial information of the weak-field solution is still useful to cancel the power broadening [22]. In recent years, there have been many studies towards strong-field coherent control. For optimization of strong-field two-photon absorptions, Weinacht and his coworkers proposed the strong-field atomic phase matching [23]. Also Silberberg's group phenomenologically studied strong-field two-photon absorptions by using the piecewise linear chirped pulse [25]. For the selection of the desired state among accessible states, the selective population of dressed states (SPODS) was demonstrated, based on terms of adiabatic passages and photo locking [26].

We have considered the analytical coherent control method for multi-photon absorption processes in strong-field regime. In this study, we have theoretically analyzed the strong-field two-photon absorption of linearly-chirped optical pulses in atomic Cesium [27]. Then, we have experimentally demonstrated a new analytical control scheme in which the static and dynamic energy level shifts are compensated by a polynomial sum of the laser spectral phase for the optimization of the two-photon absorption [28]. Further, we have applied our method for the 2+1 multi-photon absorption in Sodium, and the experimental results were found in a good agreement with the calculations from the theoretical analysis [29].

This thesis contains 7 chapters. In Chapter 2, we provide a review of coherent quantum control, which includes the typical approaches in coherent control and pulse shaping techniques. We introduce the theory of multi-photon controlled absorption in Chapter 3. The experimental apparatus used to verify the devised coherent control method are explained in Chapter 4, and the experimental results and theoretical analysis of Cesium vapor experiment is given in Chapter 5. In Chapter 6, the analytical control experiment of the 2+1 multi-photon absorption process in Sodium is explained with the calculation and experimental results. The conclusion is given in Chapter 7.

Chapter 2. Review of coherent quantum control

In this chapter, we review the early studies on coherent quantum control. Especially, we pay attention to the coherent control of multi-photon processes. Many studies have been performed to manipulate atomic or molecular states utilizing the coherence of laser fields. Two approaches were proposed: one is frequency domain coherent control, also known as Brumer-Shapiro scheme, and the other is time-domain coherent control, Rice-Tannor scheme.

2.1 Coherent control in frequency domain

Brumer-Shapiro control scheme [3], or the coherent control in frequency domain is based on controlling the interference between two several passages leading to the desired target state: one passage is a multi-photon process, and the other is an one-photon process. In this case, the relative phase between photons for the two processes needs to be well maintained. And the angular frequencies of each processes have to satisfy $\omega_1 = n\omega_n$, where ω_1 and ω_n are angular frequencies of the photons for the one-photon and multi-photon processes. For the well maintained relative phase, the electric field for the multi-photon process is obtained from the electric field for the one-photon processes via high harmonic generation. The well-known example is the experimental demonstration of the interference between three-photon process and one-photon process in Hg atoms [30]. Subsequent experiments were performed in other systems, such as HCl, HI, and CH₃I molecules. When the third harmonic photons are used for the one-photon process, the probability of the desired state P_e is given by

$$P_{e} = |A_{1ph} + A_{3ph}|^{2}$$

$$= P_{1ph} + P_{3ph} + A\cos(3\theta_{1} - \theta_{3} + \delta_{1,3}),$$
(2.1)

where A_{1ph} and A_{3ph} are the probability amplitudes induced by the one-photon and three-photon processes, respectively. θ_1 and θ_3 represent the phase of the photons for the one-photon and three-photon processes, respectively. The intrinsic phase $\delta_{1,3}$ is originated from the structure of atoms or molecules. The relative phase $\Delta \theta = 3\theta_1 - \theta_3$ can be controlled by varying the thickness of a dispersive optical medium. The probability of the desired state shows the sinusoidal behavior as a function of the relative phase as shown in Fig. 2.1. By observing this evidence of quantum interference, this Brumer-Shapiro scheme has proven the principle of coherent quantum control.

2.2 Coherent control in time domain (Semi-classical)

In time domain, Rice and Tannor developed the wavepacket modulation approach [4], which scheme is often called 'pump-dump control scheme'. A sequence of ultrafast pulses creates a wavepacket of a certain molecular state. Then the wavepacket evolves into a predetermined intermediate state. After some time delay, another sequence of dump pulses excites the wavepacket to the predetermined final state. The original version of this 'pump-dump' scheme was proposed for coherent control of molecules, but can be applied to atomic systems as well. For example, in a simple atomic system interacting with a) 3-photon and 1-photon scheme

b) experimental deminstration in Hg atoms



Figure 2.1: (a) Schematic diagram of coherent control based on the interference between 3-photon and 1-photon passages. (b) Experimental data in Hg atoms [30]. The relative phase is determined by the pressure of the Ar buffer gas.

the pulses via one-photon process, the total laser field can be expressed as

$$E_t = \sum_p E_p(t), \tag{2.2}$$

where p indicates the 'p'th pulse. In weak field regime (perturbative regime), the wavefunction after a time t is given as

$$\begin{aligned} |\psi(t)\rangle &\simeq |g\rangle + \frac{i}{\hbar} \sum_{k} \sum_{p} \int_{-\infty}^{t} \mu_{kg} E_{p}(t') e^{i\omega_{k}(t'-t)} dt' |k\rangle \\ &= |g\rangle + \frac{i}{\hbar} |\psi_{e}(t)\rangle, \end{aligned}$$

$$(2.3)$$

where μ_{kg} is the transition dipole moment between the $|g\rangle$ and $|k\rangle$ states. Then the wavefunction of excited states is given by

$$|\psi_{e}(t)\rangle = \sum_{p} |\psi_{e,p}(t)\rangle$$

$$= \sum_{p} \sum_{k} \int_{-\infty}^{t} \mu_{kg} E_{p}(t') e^{i\omega_{k}(t'-t)} dt' |k\rangle.$$
(2.4)

The wavefunction is the sum of individual wavepackets each created by constituent pulses in the sequence. The wavepacket created by the first pulse p = 1 evolves freely, and interferes with the wavepacket induced by the second pulse p = 2, and so on. At the end of the sequence of interactions, the wavefunction of the excited states is given as

$$|\psi_e(t)\rangle = \sum_p \sum_k \mu_{kg} e^{-i\omega_k t} \epsilon_p(\omega_k) |k\rangle, \qquad (2.5)$$

where $\epsilon_p(\omega_k)$ is the result of the Fourier transform of $E_p(t)$. The probability of the state k is obtained as follows

$$P_k = \left| \mu_{kg} \sum_{p} \epsilon_p(\omega_k) \right|^2.$$
(2.6)



Figure 2.2: (a) Schematic diagram of coherent control based on pump-dump scheme; L1(L2) indicates two-photon transitions by the pulse 1(2). L12 represents the passage through the intermediate state $|i\rangle$. (b) Experimental data of Cs₂ [19].

For example, when the time delay between two identical pulses is given by τ , the probability of the state k is given as $P_k(\tau) \propto [1 + \cos(\omega_k \tau)]$. The time dependence of the probability shows the oscillation of the period $2\pi/\omega_k$. The oscillation due to the interference shows that the operation of the coherent control scheme is possible. In this scheme, the time delay needs to be shorter than the life time of the excited states, because the decay of the probability amplitude toward a third intermediate state should not avoided. It is noted that, when the two pulses are overlapped, the optical interference dominates the quantum interference. Figure 2.2 shows the pump-dump scheme and the experimental results by Girard's group [19]. Here, the optical interference (L12 passage) prevails when the two pulses are overlapped. This pump-dump scheme has been developed to the quantum state measurement in terms of coherent transient [31] and two-dimensional Fourier transform optical spectroscopy [32].

2.3 Coherent control of multi-photon process in Alkali atoms

2.3.1 Non-resonant two-photon absorption in Cesium

The multi-photon process is a typical example in which the coherent control scheme based on the interference between several passages can be applied. For the coherent control of multi-photon process, broad band pulses, such as femtosecond pulses generated by a Ti:Sapphire oscillator, are used. The coherent control of two-photon process was demonstrated using femtosecond pulses in Cesium by Silberberg's group [8]. They controlled the probability of the desired state by applying certain phase functions, such as a π phase step or sinusoidal function, as shown in Figs. 2.3(a) and 2.3(b). In the experiment performed in the perturbative regime, by varying the spectral phase, the interference between the passages of the multi-photon absorption(MPA) was controlled. In the perturbative regime, the analytic approach is available for the given multi-photon process. The transition probability amplitude



a) π phase step

b) Experimental data

Figure 2.3: (a) π step phase function for the non-resonant two-photon absorption. (b) Experimental results [11]



Figure 2.4: Schematic diagram of the two-photon absorption. (a) Non-resonant two-photon absorption (b) Resonant two-photon absorption assisted by the third resonant state $|r\rangle$.

of the two-photon absorption (TPA) is given by [11]

$$a_e(t) = -\frac{1}{\hbar^2} \sum_n \mu_{en} \mu_{ng} \int_{-\infty}^t \int_{-\infty}^{t_1} E(t_1) E(t_2) \exp(i\omega_{en} t_1) \exp(i\omega_{ng} t_2) dt_2 dt_1,$$
(2.7)

where μ_{ij} is the transition dipole moment between the *i* and *j* states. Indices *e* and *g* represent the excited and ground states, respectively. If all the intermediate states are far from the pulse spectrum, the probability of the excited state can be approximated by

$$a_f \simeq -\frac{1}{i\hbar^2} \sum_n \frac{\mu_{en}\mu_{ng}}{\omega_{ng} - \omega_{eg}/2} \times \int_{-\infty}^{\infty} \epsilon(\omega)\epsilon(\omega_{eg} - \omega)d\omega, \qquad (2.8)$$

where $\epsilon(\omega)$ is the Fourier transform of E(t). Several passages are available in the given two-photon absorption, as shown in Fig. 2.4. The photon pairs, if their energy sum equals the energy difference between the ground and excited states, (i.e., $\omega_1 + \omega_2 = \omega_{eg}$), construct quantum passages to the excited state. By adjusting the spectral phase, we can control the phase of each passage. In the non-resonant TPA case (i.e., a third resonant level does not exist as shown in Fig. 2.4(a)), the constructive interference occurs if the spectral phase is constant. Thus, the transform-limited pulse, which has the shortest temporal duration among the pulses of the same bandwidth, maximizes the two-photon absorption. However, a transform-limited pulse is not always optimal in two-photon absorptions.

2.3.2 Resonant two-photon absorption in Rubidium

If a resonant level exists within the laser spectrum, as shown in Fig. 2.4(b), a transform-limited pulse is not optimal anymore [13]. In Alkali atoms, Rubidium has such resonant intermediate level, 5p states, in the two-photon absorption processes between 5s and 5d states. The two resonant wavelengths for the 5s-5p transitions are 780 nm (D_2 line) and 795nm (D_1 line), and they are within the spectrum of a typical Ti:sapphire laser. The probability amplitude of the excited state is given by

$$a_f = -\frac{\mu_{ei}\mu_{ig}}{2\hbar^2} \left[i\pi E(\omega_{ig})E(\omega_{ei}) + \wp \int_{-\infty}^{\infty} d\omega \frac{E(\omega)E(\omega_{eg} - \omega)}{\omega - \omega_{ig}} \right],\tag{2.9}$$

where indices e, g, and i denote the excited 5d, the ground 5s, and the resonant 5p states. \wp is the sign for Cauchy principal value calculation. As shown in Eq. (2.9), the denominator in the integral, $\omega - \omega_{ig}$, determines the phase of individual passage. The non-resonant passages above the resonant level $(\omega - \omega_{ig} > 0)$ and the ones below the resonant level $(\omega - \omega_{ig} < 0)$ have opposite phase (0 and π). On the other hand, the resonant passage has $\pi/2$ phase if a transform-limited pulse is used. Therefore, the non-resonant parts destructively interfere. To enhance the resonant TPA, the spectrum which contributes to the destructive interference needs to be blocked. Figure 2.5 shows the corresponding experimental result, where, as the cutoff wavelengths approached the resonant transition wavelengths, the average power was reduced by 71%, whereas the TPA rate was doubled.

Therefore, a transform-limited pulse is not optimal for the two-photon resonant transition in the weak-field interaction regime. However, in a non-resonant two-photon transition (no intermediate state), a transform-limited pulse is believed to be optimal. However, we found in the strong-field regime that the transform-limited pulse is also not an optimal pulse, even in this non-resonant TPA. We discuss this phenomenon in chapter 5.

2.3.3 Ladder climbing structure in Sodium

In a 3s-3p-5s ladder system of Sodium, Girard's group demonstrated the interference between the direct and sequential paths by using linearly chirped pulses. They observed two oscillations in experiment



Figure 2.5: Enhancement of the resonant TPA by blocking parts of spectrum. (a) A slit blocks the pulse spectrum symmetrically around $\omega_{eg}/2$. (b) The average power transmitted through the slit (diamonds) together with the experimental (circles) and calculated (line) normalized fluorescence intensities as a function of the higher cutoff wavelength. (c) Calculated temporal intensities of the optimal shaped-pulse (solid curve) and the initial 50 fs FWHM transform-limited pulse (dashed lines). The optimal pulse is wider by almost a factor of 8 (390 fs FWHM), and its peak intensity is reduced by a factor of 38. [13]



b)

a)

Figure 2.6: (a) Dressed states picture of a ladder system with two intermediate states. The arrows indicate the different paths. (b) 4p-3s fluorescence as function of chirp. [21]

with negatively chirped pulses. The fast one is originated from the interference between the sequential and direct paths, (1) and (2) with (3) in Fig. 2.6, and the slow one is caused by the interference between the two sequential paths, (1) and (2). Note that a negatively-chirped pulse enables the sequential excitation paths. From Eq. (2.9), the contributions of both paths are given by [21]

$$a_s = -\frac{\mu_{ei}\mu_{ig}}{\hbar^2} E(\omega_{ei}) E(\omega_{ig}) \frac{1 - \operatorname{sgn}(a_2\delta)}{2}, \qquad (2.10)$$

$$a_{d} = -\frac{\mu_{ei}\mu_{ig}}{\hbar^{2}} \frac{E^{2}(\omega_{0})}{\delta\sqrt{2\pi T_{0}T_{p}}} e^{-i(\theta+\pi)},$$
(2.11)

where a_s and a_d are the sequential and direct contributions, respectively, and $\tan\theta = -2a_2/T_0^2$. T_0 is the pulse duration of a transform-limited pulse. T_p is determined by $T_p = T_0\sqrt{1 + (2a_2/T_0^2)^2}$. The one-photon detuning δ is given by $\delta = \omega - \omega_{ig}$. The direct contribution has the chirp rate dependence, $e^{-i\theta}$. It induces the fast oscillation as a function of linear chirp rate a_2 . $\operatorname{sgn}(a_2\delta)$ has a crucial role to determine whether the sequential path is available or not. In Sodium 3s-3p-5s system, δ is positive $(\delta \simeq 0.070 \text{ rad/fs}^{-1})$. Therefore, the sequential path has nonzero contribution to the probability of the excited state only if a negatively-chirped pulse is used. A negatively chirped pulse enhance the excitation of the 3s-3p-5s ladder system of Sodium better than a positively chirped pulse. This experiment also showed that a transform-limited pulse is not optimal in a ladder system. In chapter 6, we adopt this interference scenario of sequential and direct transition paths for a different multi-photon transition in Sodium, which is the 2+1 multi-photon absorption process in 3s-4s-7p system.

2.4 Pulse shaping in coherent control

In this section, we describe pulse shaping devices for coherent control experiments. In the recent decades, more complex pulse shapes have been required to control complex systems than before. Complex temporal shapes are obtained by modulating the amplitude and phase in Fourier domain, which has been achieved by applying a spectral filter, such as a liquid crystal spatial light modulator (SLM) [6], an



Figure 2.7: Principle of Fourier domain pulse shaping using a SLM. Gratings and lenses are arranged in a 4f configuration

acousto-optic modulator (AOM) [34], or a deformable mirror [35].

Figure 2.7 shows the pulse shaping in Fourier domain. A typical pulse shaping setup in Fourier domain is a 4f configuration which consists of two gratings, two lenses, and a modulator, separated by the focal length of lenses as shown in Fig. 2.7. The first grating spatially separates spectral components, and the phase and amplitude of spatially separated spectral components are controlled by the spatial modulator. After that, the controlled spectral components are converged by the second grating, and we obtain a shaped ultrafast pulse. In the next subsections, two widely used components in a pulse shaper, the liquid crystal array and the acousto-optic modulator (AOM), are introduced in more detail.

2.4.1 Liquid crystal arrays

The refractive index of the liquid crystal can be controlled by applying the voltage into the liquid crystal [6]. Liquid crystal arrays consist of three layers: a thin layer of a liquid crystal and two pieces of glass coated with electrically conducting films (Additionally, one piece is patterned into separate electrodes or pixels.) The liquid crystal consists of long thin rod-like molecules, which are originally aligned with their long axis (for example, along the y direction). When an electric field perpendicular to the long axis of molecules (z direction) is applied, the molecules incline to the z direction, causing a refractive index change for y-polarized light. A maximum phase change of at least 2π is required for complete phase control. In order to apply the electric field, the inside surface of each glass is coated with a thin, transparent, electrically conducting film of indium tin oxide. One piece is patterned into separate electrodes or pixels. A typical device has 128 (256, 640) pixels of liquid crystals which are individually controlled electronically. Thus the phase of the light passing through the liquid crystal is a function of the voltage applied to the electrodes. If the liquid crystal array is used alone in a 4f configuration, only the phase shaping is possible. But the combination with a pair of polarizers enables the amplitude shaping as well. Generally, pulse shapers based on liquid crystal arrays need two liquid crystal arrays to manipulate amplitude and phase simultaneously. It has the advantage for realization of discontinuous phase steps. The waveform update time depends on the reorientation time of the liquid crystal molecules,



Figure 2.8: Principle of Fourier domain pulse shaping using an AOM: gratings and lens is arranged in a 4f configuration

50 ms, longer than the update time of an AOM. The liquid crystal arrays has smaller power loss than the AOM, and can be used for high repetition lasers. But, it has disadvantages of complex alignment, pulse shape distortion, due to pixel gaps and pixel calibration, and linkage of on-off pixel operation with neighbor pixels.

2.4.2 Acousto-optic modulator

In a 4f configuration, liquid crystal arrays can be replaced by an acousto-optic modulator as shown in Fig 2.8. This type of pulse shaper controls both the phase and amplitude of each frequency components [34]. The acousto-optic modulator generates an acoustic wave by an arbitrary waveform generator synchronized with the kHz repetition rate of the laser. The acoustic wave propagates perpendicular to the direction of the pulse propagation, and generates an index pattern in the acousto-optic crystal. This index pattern determines the amplitude and phase of spectral components which are diffracted. The AOM uses a large aperture TeO₂ (visible) or InP (Infrared) AOM crystal. The acoustic wave propagates at the speed of sound in the crystal, $4.2mm/\mu s$ for TeO₂. The speed of light in the crystal is about $2.0 \times 10^5 mm/\mu s$. Thus the acoustic wave is regarded as a fixed diffraction grating. Because of using the acoustic wave, the acousto-optic pulse shaping is suitable for laser systems of a kHz repetition rate. Other type, an acousto-optic programmable dispersive filter (AOPDF) allows the pulse shaping without Fourier transform optics [36]. It also utilizes an acousto-optic interaction but acoustic and optical waves propagate collinear, not perpendicular. It is easy to align and obtain the smooth spectral modulation. It is also possible to modulate both amplitude and phase. Usually, it is used to compensate the dispersion for an amplifier system. More details of AOPDF will be explained in chapter 4.



Figure 2.9: Schematic diagram of the closed loop coherent control

2.5 Closed-loop and open-loop control schemes

The purpose of coherent quantum control is to optimize certain light-matter interaction. In a simple system whose Hamiltonian can be written as a simple form, one can easily obtain an analytic solution to describe the probability amplitude. Such an analytic solution allows a direct determination of the pulse shape. This scheme is called 'open-loop optimal control'. This scheme is suitable to find physical meanings of pulse shapes. In a complex system, it is hard to obtain an analytic solution of the system, hence it is impossible to predict an optimal pulse shape for the desired target state. For practical applications such as determination of a chemical channel, an optimization using a learning algorithm and a feedback are required as shown in Fig. 2.9. This scheme is called 'closed-loop optimal control' [5]. It is composed of iterative measurements and complex pulse shaping: both processes are connected with a learning algorithm, such as genetic algorithm. A learning algorithm steers a pulse shape to the direction of maximizing a certain outcome, without a priori information of Hamiltonian. After many iterations, one can obtain an optimal pulse shape. Bardeen used this technique to demonstrate the control of molecular electronic population transfer in dye molecules [33]. Gerber and co-workers tried to optimize the branching ratios of photo-dissociation channels by using this technique [7]. This scheme is used practically but it is hard to find physical meaning of optimal pulse shapes due to unknown Hamiltonian.

In our study, we will use open-loop coherent control to investigate multi-photon absorption processes of Alkali atoms such as Cesium and Sodium. These absorption processes can be modeled by twolevel and three-level Hamiltonians, respectively. By analyzing the Hamiltonians, we have successfully obtained analytical solutions of the given light-matter interaction as a function of pulse shape parameters, especially in the strong-field regime. In the next chapter, we will describe the theory of multi-photon processes in the presence of non-negligible dynamic Stark shifts.

Chapter 3. Theoretical background and calculation

In this chapter, we describe the theory of two-level two-photon absorption process in the strong field regime. First, we introduce two-photon spectrogram picture to explain the two-photon absorption in the presence of dynamic Stark shift, and then we obtain analytic optimization pulse-shape solutions for the given absorption process by compensating the time-dependent transition level shift. Finally, we expand the theory developed for the two-level two-photon absorption into a three-level three-photon absorption problem.

3.1 Semi-classical approach to one-photon transition in a twolevel system

We provide a brief review of the problem of a dipole-coupled two-level system interacting with a laser pulse. The semi-classical approach to the given problem is one of the fundamental examples of quantum mechanics. The theory introduced in this section is used as an analogy to the approach in multi-photon absorption problems to be discussed in the following sections. First, we consider a two-level system that interacts with a short laser field as shown in Fig. 3.1. We note that a dipole transition is allowed between the two states which have different angular momentum numbers ℓ , for example $|5s, \ell = 0 >$ and $|5p, \ell = 1 >$ in Rubidium, according to the selection rule. After we apply the dipole approximation, the interaction Hamiltonian is given as

$$V(t) = -\vec{\mu} \cdot \vec{E}(t), \qquad (3.1)$$

where E(t) is the electric field and μ is the electric dipole moment. To estimate the population of the states, we solve the Schrödinger equation in a density matrix form [38], i.e.

$$\dot{\rho}_{ij} = -(\gamma_{ij} + i\omega_{ij})\rho_{ij} - \frac{i}{\hbar}[V,\rho]_{ij}, \qquad (3.2)$$

where $\hbar \omega_{ij}$ is the energy difference between the energy levels *i* and *j*, and γ_{ij} is the decay constant. γ_{ij} is assumed very small and then the density matrix equation is given by

$$\dot{\rho}_{gg} = \frac{i}{\hbar} E(t) \mu_{ge} \rho_{eg}, \tag{3.3}$$

$$\dot{\rho}_{ee} = \frac{i}{\hbar} E(t) \mu_{eg} \rho_{ge}, \qquad (3.4)$$

$$\dot{\rho}_{eg} = -i\omega_{eg}\rho_{eg} + \frac{i}{\hbar}E(t)\mu_{eg}\rho_{gg}, \qquad (3.5)$$

where μ_{eg} is the transition dipole moment between the ground excited states. The angular frequency difference between these two states is ω_{eg} . We denote $\rho_{eg} = \Re_{eg} e^{-i\omega_{eg}t}$, where \Re_{eg} is the slowly varying envelope that changes little during the optical pulse duration. Then, Eqs. (3.2) become

$$\dot{\rho}_{gg} = \frac{i}{\hbar} E(t) e^{-i\omega_{eg}t} \mu_{ge} \Re_{eg}, \qquad (3.6)$$

$$\dot{\rho}_{ee} = \frac{i}{\hbar} E(t) e^{i\omega_{eg}t} \mu_{eg} \Re_{ge}, \tag{3.7}$$

$$\dot{\Re}_{eg} = \frac{\imath}{\hbar} E(t) \mu_{eg} \rho_{gg}, \tag{3.8}$$



Figure 3.1: Schematic diagram of broadband laser pulse interaction with a two-level atom via one-photon transition

and can be written further in a simple form as

$$\dot{\rho_{tr}}(t) = \frac{i}{\hbar} \left[\rho_{tr}(t), \mathbf{H_{tr}}(t) \right], \tag{3.9}$$

where the Hamiltonian H_{tr} is given by

$$H_{tr} = \begin{bmatrix} 0 & \mu_{ge} E(t) \exp(-i\omega_{eg}t) \\ \mu_{eg} E(t) \exp(i\omega_{eg}t) & 0 \end{bmatrix}.$$
(3.10)

The transformed Hamiltonian is written in the interaction picture and the probability amplitudes are given by

$$\dot{a}_g = i\mu_{ge}E(t)\exp(-i\omega_{eg}t)a_e, \qquad (3.11)$$

$$\dot{a_e} = i\mu_{eg}E(t)\exp(i\omega_{eg}t)a_g. \tag{3.12}$$

We assume that the atom is initially in the ground state and the interaction with a shaped laser pulse is in the weak field regime. Then the probability amplitude of the excited state, given as a result of the first-order perturbation, is obtained as

$$a_e^{(1)} \simeq -\frac{i}{\hbar} \mu_{eg} \int_{-\infty}^{\infty} dt E(t) \exp(i\omega_{eg} t)$$

$$= -\frac{i}{\sqrt{2\pi\hbar}} \mu_{eg} \epsilon(\omega_{eg}),$$
(3.13)

where $\epsilon(\omega)$ is the Fourier transform of E(t). Therefore, $a_e^{(1)}$ is determined by the spectral amplitude and phase of the photons with angular frequency ω_{eg} , only, and not dependent on any other frequency components. Figure 3.2 shows the excitation probability, $|a_e^{(1)}|^2$, of Rubidium D_1 transition, plotted as a two-dimensional function of linear chirp rate and pulse energy. Here we assume that the interacting laser pulse is linearly chirped rate but the population of the excited state is proportional to $|\epsilon(\omega_{eg})|^2$ which is constant. Hence $|a_e^{(1)}|^2$ is not dependent on the phase of the shaped pulse and, therefore, becomes a constant for a linearly-chirped pulse. This result in the weak field regime is well illustrated in Fig. 3.2, which shows a good agreement between the experimental data and the calculation based on the first-order perturbation theory.



Figure 3.2: (a) Calculation and (b) measurement of Rubidium D1 fluorescence at 794.7 nm. The fluorescence is proportional to the excited $5P_{1/2}$ -state population.

The second order perturbation calculates the two-photon transition rate of a ground-state atom back to the ground-state via the excited state, given by

$$a_{g}^{(2)} \simeq -\frac{\mu_{ge}\mu_{eg}}{\hbar^{2}} \int_{-\infty}^{\infty} dt E(t) \exp(-i\omega_{eg}t) \int_{-\infty}^{t} dt_{1}E(t_{1}) \exp(i\omega_{eg}t_{1})$$

$$= -\frac{\mu_{eg}^{2}}{\hbar^{2}} \left[i\pi\epsilon^{*}(\omega_{eg})\epsilon(\omega_{eg}) + \wp \int_{-\infty}^{\infty} d\omega \frac{\epsilon^{*}(\omega)\epsilon(\omega)}{\omega_{eg} - \omega} \right],$$
(3.14)

where \wp is the Cauchy principal value and ω_c is the spectrum center. If $\epsilon(\omega)$ is symmetric about the angular frequency ω_{eg} , the integral vanishes and only the first term with the resonant component $\epsilon^*(\omega_{eg})\epsilon(\omega_{eg})$ is nonzero. If laser spectrum is far off from the resonant frequency ω_{eg} , the probability amplitude can be written as

$$a_g^{(2)} \simeq -\frac{1}{\hbar^2} \frac{\mu_{eg}^2}{\omega_{eg} - \omega_c} \int_{-\infty}^{\infty} d\omega \epsilon^*(\omega) \epsilon(\omega), \qquad (3.15)$$

and, as a result, the laser laser spectral function affects the transition probability in this two-photon process.

3.2 Semi-classical approach to non-resonant two-photon absorption in a two-level system

In this section, we consider two-photon transitions of a two-level atom interacting with an ultrafast laser field, as depicted in Fig. 3.3. In particular, we treat non-resonant two-photon absorption, which means that there are no extra resonant levels within the laser spectrum. The transition selection rule does not allow one-photon transitions between the ground ($\ell = 0$) and excited ($\ell = 0$) states, but twophoton transitions are still possible. This is a good model for a two-photon resonant transition between the $|6s\rangle$ and $|8s\rangle$ states of atomic Cesium. The resonant laser wavelength $\lambda(= 2\pi c/\nu)$ for this two-photon transition, $\nu = \omega_{6s-8s}/2$, is 822 nm. If we take the fact that the typical wavelength width $\Delta \lambda = 20$ -30 nm for an ultrafast laser pulse into account, the resonant one-photon transition to $|6p\rangle$, from $|6s\rangle$, is



Figure 3.3: Schematic diagram of a two-photon transition with no intermediate one-photon resonant states

far off-resonant from the laser field. We note $\lambda_{6s-6p} = 852$ nm. A more careful validation for two-level modeling is given in section 5.5.

Also, we employ a semi-classical Hamiltonian. For this problem, the semi-classical description is perfectly adequate, because the number of photons in the laser pulse is huge. Thus, the laser field is written classically as:

$$\mathbf{E}(t) = \frac{1}{2} E_0 \sqrt{g(t)} e^{i\phi(t)} e^{i\nu t} \hat{\varepsilon} + c.c., \qquad (3.16)$$

where g(t) and $\phi(t)$ are the temporal intensity profile and temporal phase, respectively. ν is the center angular frequency of the pulse. $\hat{\varepsilon}$ is the polarization vector and E_0 is the field strength. The energy of the dipole-field interaction is given by

$$V_{ij} = -\frac{1}{2}\mu_{ij}E_0\sqrt{g(t)}e^{i\phi(t)}e^{i\nu t} + c.c., \qquad (3.17)$$

where μ_{ij} is the transition dipole moment defined as $\mu_{ij} = \langle i | e\mathbf{r} \cdot \hat{\varepsilon} | j \rangle$. In the case of one-electron atoms, or alkali atoms, the electric dipole only connects states of opposite parities, $\ell + \ell' = \text{odd}$ [39]. Thus s states are only coupled with p states and the p states are not coupled with other p states. As a result, the couplings between p intermediate states are negligible in alkali atoms. By solving the generalized Schrödinger equation (3.2), the coherence between the intermediate and ground (excited) states is obtained as the following differential equations:

$$\dot{\rho}_{jg} = -(\gamma_{jg} + i\omega_{jg})\rho_{jg} + \frac{i}{2\hbar}E_0 \left[\sqrt{g(t)}e^{i\phi(t)}e^{i\nu t} + c.c\right] \left[\mu_{jg}\rho_{gg} + \mu_{je}\rho_{eg}\right],\tag{3.18}$$

$$\dot{\rho}_{je} = -(\gamma_{je} + i\omega_{je})\rho_{je} + \frac{i}{2\hbar}E_0 \left[\sqrt{g(t)}e^{i\phi(t)}e^{i\nu t} + c.c\right] \left[\mu_{jg}\rho_{ge} + \mu_{je}\rho_{ee}\right],\tag{3.19}$$

We assume that the decay rate $\gamma_{jg,e}$ is negligible. As ρ_{eg} is rapidly oscillating with the frequency of ω_{eg} , we set $\rho_{eg} = \Re_{eg} e^{-i\omega_{eg}t}$. For the following condition, or if the electric field envelope is a slowly varying function,

$$\frac{d}{dt} \left| \frac{\mu_{jg,ej} E_0}{\hbar} \sqrt{g(t)} \right| << \left| \omega_{jg,ej} - \nu \right|.$$
(3.20)

Then, the coherence between the intermediate and ground (excited) state can be written as:

$$\rho_{jg} = \frac{i}{2\hbar} E_0 \int_{-\infty}^t dt' \left[\sqrt{g(t')} e^{i\phi(t')} e^{i\nu t'} + c.c \right] e^{-i\omega_{jg}(t-t')} \left[\mu_{jg} \rho_{gg} + \mu_{je} \Re_{eg} e^{-i\omega_{eg}t'} \right]$$

$$= \frac{1}{2\hbar} E_0 \left[\frac{\sqrt{g(t)} e^{i\phi(t)} e^{i\nu t}}{\omega_{jg} + \nu} + \frac{\sqrt{g(t)} e^{-i\phi(t)} e^{-i\nu t}}{\omega_{jg} - \nu} \right] \mu_{jg} \rho_{gg}$$

$$- \frac{1}{2\hbar} E_0 \left[\frac{\sqrt{g(t)} e^{i\phi(t)} e^{i(\nu - \omega_{eg})t}}{\omega_{ej} - \nu} + \frac{\sqrt{g(t)} e^{-i\phi(t)} e^{-i(\nu + \omega_{eg})t}}{\omega_{ej} + \nu} \right] \mu_{je} \Re_{eg},$$
(3.21)

$$\rho_{je} = \frac{i}{2\hbar} E_0 \int_{-\infty}^t dt' \left[\sqrt{g(t')} e^{i\phi(t')} e^{i\nu t'} + c.c \right] e^{-i\omega_{je}(t-t')} \left[\mu_{jg} \Re_{ge} e^{i\omega_{eg}t'} + \mu_{je} \rho_{ee} \right]$$
(3.22)
$$= \frac{1}{2\hbar} E_0 \left[\frac{\sqrt{g(t)} e^{i\phi(t)} e^{i(\nu+\omega_{eg})t}}{\omega_{jg} + \nu} + \frac{\sqrt{g(t)} e^{-i\phi(t)} e^{i(-\nu+\omega_{eg})t}}{\omega_{jg} - \nu} \right] \mu_{jg} \Re_{ge}$$
$$- \frac{1}{2\hbar} E_0 \left[\frac{\sqrt{g(t)} e^{i\phi(t)} e^{i\nu t}}{\omega_{ej} - \nu} + \frac{\sqrt{g(t)} e^{-i\phi(t)} e^{-i\nu t}}{\omega_{ej} + \nu} \right] \mu_{je} \rho_{ee}.$$

If either a resonance condition between $\nu \simeq \omega_{jg}$ and $\nu \simeq \omega_{ej}$ is satisfied, the coherence between the intermediate state and the ground (or excited) state is not negligible, and the population ρ_{jj} of intermediate states can not be ignored. Then, a full consideration requires at least three energy states, and a three-level system is described in the next section. Otherwise, by substituting Eqs. (3.21) and (3.22) into Eq. (3.2) and by neglecting the rapidly oscillating terms such as $e^{i2\nu t}$, $e^{i\omega_{eg}t}$, and $e^{i(2\nu+\omega_{eg}t)}$, we have the set of the following differential equations:

$$\dot{\rho}_{gg} = \frac{i}{4\hbar^2} E_0^2 g(t) \frac{e^{i(\Delta t + 2\phi(t))}}{\omega_{je} + \nu} \mu_{gj} \mu_{je} \Re_{eg} - \frac{i}{4\hbar^2} E_0^2 g(t) \frac{e^{-i(\Delta t + 2\phi(t))}}{\omega_{je} + \nu} \mu_{ej} \mu_{jg} \Re_{ge}, \tag{3.23}$$

$$\dot{\rho}_{ee} = \frac{i}{4\hbar^2} E_0^2 g(t) \frac{e^{-i(\Delta t + 2\phi(t))}}{\omega_{jg} - \nu} \mu_{ej} \mu_{jg} \Re_{ge} - \frac{i}{4\hbar^2} E_0^2 g(t) \frac{e^{i(\Delta t + 2\phi(t))}}{\omega_{jg} - \nu} \mu_{gj} \mu_{je} \Re_{eg},$$
(3.24)

$$\dot{\Re}_{eg} = \frac{i}{4\hbar^2} E_0^2 g(t) \frac{e^{-i(\Delta t + 2\phi(t))}}{\omega_{jg} - \nu} \mu_{ej} \mu_{jg} \rho_{gg} + \frac{i}{2\hbar^2} E_0^2 g(t) \frac{\omega_{je}}{\omega_{je}^2 - \nu^2} \mu_{ej} \mu_{je} \Re_{eg}, \tag{3.25}$$

$$-\frac{i}{2\hbar^2}E_0^2g(t)\frac{\omega_{jg}}{\omega_{jg}^2-\nu^2}\mu_{gj}\mu_{jg}\Re_{eg}-\frac{i}{4\hbar^2}E_0^2g(t)\frac{e^{-i(\Delta t+2\phi(t))}}{\omega_{je}+\nu}\mu_{ej}\mu_{jg}\rho_{ee},$$

where Δ is the two-photon detuning, given as $\Delta = 2\nu - \omega_{eg}$. The summation index j is omitted. From Eqs. (3.23)-(3.25), we reconstruct the Hamiltonian of 2 by 2 square matrix:

$$\mathbf{H}(\mathbf{t}) = \hbar \begin{pmatrix} S_g(t) & \frac{1}{2}\Omega(t)e^{i[\Delta t + 2\phi(t)]} \\ \frac{1}{2}\Omega(t)e^{-i[\Delta t + 2\phi(t)]} & S_e(t) \end{pmatrix},$$
(3.26)

and the set of differential equations can be written as

$$\dot{\rho}(t) = \frac{i}{\hbar} \left[\rho(t), \mathbf{H}(t) \right], \qquad (3.27)$$

where $S_{g,e}$ represent the dynamic Stark shift of the ground and excited states.

In the presence of intermediate states of angular frequencies ω_j , an intense laser field temporally shifts the angular frequencies of the ground and excited states, and the dynamic Stark effect is given by

$$S_{e,g}(t) = -\sum_{j} \frac{|\mu_{e,gj}|^2}{2\hbar^2} E_0^2 g(t) \frac{\omega_{je,g}}{\omega_{je,g}^2 - \nu^2},$$
(3.28)

where $\mu_{ej(jg)}$ are the transition dipole moments between the excited (ground) and intermediate states. $\Omega(t)$ is the two-photon Rabi frequency,

$$\Omega(t) = -\sum_{j} \frac{\mu_{ej} \mu_{jg}}{2\hbar^2} E_0^2 g(t) \frac{1}{\omega_{jg} - \nu} \simeq -\sum_{j} \frac{\mu_{gj} \mu_{je}}{2\hbar^2} E_0^2 g(t) \frac{1}{\omega_{je} + \nu},$$
(3.29)

representing the coupling between the excited and ground states for the small two-photon atom-field detuning. Consistently, the two-photon rotating wave approximation is given that $|\Delta| \ll \omega_{jg} - \nu \simeq \omega_{je} + \nu$. The Hamiltonian of the system can be transformed to a more simple form. We define $\Re_{eg} = \wp_{eg} e^{-i \int_{-\infty}^{t} S(t') dt'}$, where S(t) is the difference of the dynamic Stark shifts defined as $S(t) = S_e(t) - S_g(t)$. It simplifies the Eqs. (3.23) and (3.24) as:

$$\dot{\rho}_{gg} = -i\frac{1}{2}\Omega(t)e^{i\alpha(t)}\wp_{eg} + i\frac{1}{2}\Omega(t)e^{-i\alpha(t)}\wp_{ge}, \qquad (3.30)$$

$$\dot{\rho}_{ee} = -i\frac{1}{2}\Omega(t)e^{-i\alpha(t)}\wp_{ge} + i\frac{1}{2}\Omega(t)e^{i\alpha(t)}\wp_{eg}, \qquad (3.31)$$

$$\dot{\varphi}_{eg} = -i\frac{1}{2}\Omega(t)e^{-i\alpha(t)}\rho_{gg} + i\frac{1}{2}\Omega(t)e^{-i\alpha(t)}\rho_{ee}, \qquad (3.32)$$

where $\alpha(t)$ is the induced phase accumulated by the atom-field interaction, obtained as $\alpha(t) = -\int_{-\infty}^{t} S(t')dt' + \Delta t + 2\phi(t)$. With $\alpha(t)$, the simplified Hamiltonian can be written as

$$\mathbf{H}^{(\mathbf{tr})}(\mathbf{t}) = \hbar \begin{pmatrix} 0 & \frac{1}{2}\Omega(t)e^{i\alpha(t)} \\ \frac{1}{2}\Omega(t)e^{-i\alpha(t)} & 0 \end{pmatrix}.$$
(3.33)

We note that the complete transfer is achieved by both the condition of the minimum pulse area, or $\int dt \Omega(t) = \pi$, and the phase matching condition

$$\dot{\alpha}(t) = -S(t) + \Delta + 2\dot{\phi}(t) = 0,$$
(3.34)

that needs to be satisfied for the pulse duration. With this phase matching, the resonance condition is maintained, or the dynamic Stark shift is compensated by temporal change of laser frequency.

3.2.1 Weak-field formula

In the weak-field regime, the probability amplitude of the excited state can be written up to the first order of Dyson series:

$$A_e \simeq -i \int_{-\infty}^{\infty} dt' \frac{1}{2} \Omega(t') \exp[i\alpha(t')] dt'$$
(3.35)

$$= -i \int_{-\infty}^{\infty} dt' \frac{1}{2} \Omega(t') \exp[i(\Delta t' + 2\phi(t'))] dt'$$
(3.36)

$$= i \sum_{j} \frac{\mu_{ej} \mu_{jg}}{4\hbar^2} \frac{1}{\omega_{jg} - \nu} \int_{-\infty}^{\infty} E_0^2 g(t') \exp[i(2\nu - \omega_{eg})t' + 2i\phi(t')] dt'.$$
(3.37)

The dynamic Stark shift S(t), contained in the phase $\alpha(t)$, is proportional to the pulse peak intensity I, and is considered to be zero in the weak-field limit. With the electric field in frequency domain represented as the Fourier transform

$$\frac{1}{2}E_0\sqrt{g(t)}e^{i\nu t}e^{i\phi(t)} = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty}\epsilon(\omega)e^{i\omega t}d\omega,$$
(3.38)

Eq. (3.35) can be rewritten in frequency domain [47] as

$$A_{e} = i \sum_{j} \frac{\mu_{ej} \mu_{jg}}{\hbar^{2}} \frac{1}{\omega_{jg} - \nu} \int_{-\infty}^{\infty} d\omega_{1} \int_{-\infty}^{\infty} d\omega_{2} \epsilon(\omega_{1}) \epsilon(\omega_{2})$$

$$\times \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{i(\omega_{1} + \omega_{2} - \omega_{eg})t}$$

$$= i \sum_{j} \frac{\mu_{ej} \mu_{jg}}{\hbar^{2}} \frac{1}{\omega_{jg} - \nu} \int_{-\infty}^{\infty} d\omega_{1} \int_{-\infty}^{\infty} d\omega_{2} \epsilon(\omega_{1}) \epsilon(\omega_{2}) \delta(\omega_{1} + \omega_{2} - \omega_{eg})$$

$$= i \sum_{j} \frac{\mu_{ej} \mu_{jg}}{\hbar^{2}} \frac{1}{\omega_{jg} - \nu} \int_{-\infty}^{\infty} d\omega_{1} \epsilon(\omega_{1}) \epsilon(\omega_{eg} - \omega_{1}),$$
(3.39)

Thus the probability amplitude of the excited state is determined by the sum of pairwise interference between ω and $\omega_{eg} - \omega$ photons. In that regards, a transform-limited pulse induces fully constructive interference and maximizes the two-photon absorption in the weak-field regime.

3.2.2 Strong-field solutions

In a strong-field regime, dynamic Stark shifts and higher-order transitions have to be considered. When phase matching is satisfied or $\dot{\alpha}(t) = 0$, the Hamiltonians at different time commute. In this case, we obtain an analytic form of the probability amplitude.

The time-evolution operator is given as

$$U(t) = \hat{I}\cos(\int_{-\infty}^{t} \frac{1}{2}\Omega(t')dt') - i\hat{\sigma}_x\sin(\int_{-\infty}^{t} \frac{1}{2}\Omega(t')dt'), \qquad (3.40)$$

where \hat{I} is a 2 by 2 identity matrix, and $\hat{\sigma}_x$, $\hat{\sigma}_y$, and $\hat{\sigma}_z$ denote Pauli matrices. The excited-state population can be written as

$$P_e(t) = \sin^2\left(\int_{-\infty}^t \frac{1}{2}\Omega(t')dt'\right),\tag{3.41}$$

which behavior is the Rabi oscillation given as a function of pulse intensity. The final probability can be maximized by the condition $\int_{-\infty}^{\infty} \Omega(t') dt' = \pi$. From this condition, we determine the minimum energy to induce the population inversion (i.e. $P_e = 1$), to be terms as the minimum π -pulse energy. Above the minimum π -pulse energy, the excited-state population is decreased because the stimulated emission is larger than the absorption. When $\dot{\alpha}(t) \neq 0$, the Hamiltonians at different-time do not commute anymore. The Hamiltonian can be rewritten as

$$H = \frac{1}{2}\Omega(t)\cos(\alpha(t))\hat{\sigma}_x - \frac{1}{2}\Omega(t)\sin(\alpha(t))\hat{\sigma}_y.$$
(3.42)

Then, the time-evolution operator U(t) satisfies the following differential equation,

$$\frac{\partial}{\partial t}U(t) = -i\frac{1}{2}\Omega(t)\cos(\alpha(t))\hat{\sigma}_x U(t) + i\frac{1}{2}\Omega(t)\sin(\alpha(t))\hat{\sigma}_y U(t).$$
(3.43)

The time evolution of the states can be obtained by solving the Schrödinger equation. Even the Schrödinger equation is numerically solvable, for example, by using the fourth order Runge-Kutta method, it is in general very hard to obtain an exact formula for the excited state. To approach the excited population analytically, we introduce a new parameter, complex pulse area defined as

$$\theta = \int_{-\infty}^{\infty} \frac{1}{2} \Omega(t) \exp(i\alpha(t)) dt, \qquad (3.44)$$



Figure 3.4: Estimation of the excited population as a function of pulse peak intensity and linear chirp rate. (a) Numerical solution of Schrödinger equation. (b) $|\theta|^2$. Two results show a good agreement.

which is also the result of the first order Dyson series of Eq. (3.43). When $|\Omega(t)\tau/2| \ll 1$, $|\theta|^2$ approximate the state probability.

In Cesium, we estimate that the maximum amplitude of $\Omega(t)$ is -54 Trad/s at $I = 1.0 \times 10^{11}$ W/cm². In the experiments, the pulse duration τ is ranging from 37 fs to 90 fs. Thus, $2.0 < |\Omega(t)\tau/2| < 4.86$ at $I = 1.0 \times 10^{11}$ W/cm², and the experiment is in non-perturbative regime, if the spatial-intensity effect is not taken into account. However, due to the spatial-intensity effect, the effective $\Omega(t)^{-1}$ is reduced and the perturbative approach turns out valid. As a result, $|\theta|^2$ is the excited-state population in our experiment. For example, Fig. 3.4 shows the excited-state populations: (a) the numerical solution of Schrödinger equation; (b) the complex pulse area. They show a good agreement, and that the complex pulse area is a good estimate for the excited-state population. More specification about the spatial intensity averaging effect is given in section 3.4.

The atom-field phase $\alpha(t)$ represents the accumulated detuning given as a function of time, and the nonzero phase occurs as a result of accumulated off-resonances. When $\dot{\alpha}(t)$ is large (large detuning case), the pulse area becomes zero, because $\Omega(t)\exp(i\alpha(t))$ becomes a rapidly oscillating function. If $\dot{\alpha}(t) = 0$, on the other hand, the pulse area can be maximized (zero detuning case). The degree of off-resonance can be written as $\dot{\alpha}(t) = E_e(t)/\hbar - 2\omega(t)$ where the shifted energy is given as $E_e(t) = \omega_{eg} + S(t)$ and the instant frequency of the pulse is represented as $\omega(t) = \nu + \dot{\phi}(t)$.

Spectrogram is useful to represent the characteristic of laser pulses in phase space, which is an analogy of the Wigner quasi-probability distribution for a classical field. The Wigner quasi-probability distribution has been widely used in quantum optics [44]. The spectrogram given as

$$W(\omega,t) = \int_{-\infty}^{\infty} E^*(t+\tau)E(t-\tau)e^{2i\omega\tau}d\tau,$$
(3.45)

shows the probability distribution of the pulse in time-frequency domain. Especially, a chirped pulse is well described by the spectrogram (e.g. the slope of the distribution depicts the linear chirp rate.) We

 $^{^{1}}$ The effective two-photon Rabi frequency is an average of two-photon Rabi frequencies of individual atoms, which is usually 6-12 times smaller than the two-photon Rabi frequency given at the pulse-peak intensity.

define a two-photon spectrogram as

$$P(\Omega, t) = \int_{-\infty}^{\infty} W(\omega, t) W(\Omega - \omega, t) d\omega, \qquad (3.46)$$

where Ω denotes the sum frequency. For a two-photon absorption, the two-photon resonance is satisfied at $\Omega = \omega_{eg}$, and therefore, the overlap between the time-dependent resonance frequency and the two-photon spectrogram represent an absorption probability distribution given as a function of both time and Ω , in spectral-temporal two-dimensional space. Figure. 3.5 shows spectrogram and two-photon spectrogram of two different chirped pulses. The density of two-photon spectrogram overlapped with the shifted energy level depicted as dashed lines in Figs. 3.5(b) and (d) determines the total magnitude of two-photon absorptions. It is useful to qualitatively estimate two-photon absorptions induced by a shaped pulses. The instant frequency in a two-photon spectrogram is determined by $2\omega(t) = 2\nu + 2\dot{\phi}(t)$, and therefore $\dot{\alpha}(t)$ is the difference between the shifted energy level and two-photon spectrogram, depicted as the black arrows in Figs. 3.5(b) and (d).

In this section, a simple form of transition probability amplitude is obtained for strong-field twophoton absorption. From the formula used in the low laser intensity limit, we successfully construct the well-known weak-field formula for the given two-photon absorption. We verify this derived theoretical results experimentally in chapter 5, where examples of laser excitation with more complex shapes are considered.

3.3 Two-photon and one-photon transitions in a three-level ladder system

In this section, we expand the theory of two-photon absorption, derived in the previous section, into a three-level ladder system. We consider a 2+1 multi-photon process in alkali atoms, where '2+1' means that the system consists of a two-photon and a one-photon transitions. The schematic diagram of the 2+1 multi-photon ladder system is shown in Fig. 3.6. In this case, a higher energy state $|r\rangle$ is added to the previously considered two-level system of $|g\rangle$ and $|e\rangle$, and the newly added state $|r\rangle$ is one-photon coupled to the excited state $|e\rangle$.

In Sodium, the ground-state atom is excited to the 4s excited state by non-resonant two-photon absorption of 777 nm photons. Simultaneously, the 4s-excited atom can be further excited by absorbing a 781-nm photon. Because the energy difference between the 4s and 7p state is 781 nm in wavelength scale, the 4s-7p transition is resonant with the laser field. Thus, the 7p state has to be treated explicitly, which is different from the non-resonant intermediate states. Then, the probability amplitudes are given by the differential equations as

$$\begin{aligned} \dot{a}_{g}(t) &= -iS_{g}(t)a_{g}(t) - i\frac{1}{2}\Omega(t)e^{i[\Delta_{1}t+2\phi(t)]}a_{e}(t), \\ \dot{a}_{e}(t) &= -i\frac{1}{2}\Omega(t)e^{-i[\Delta_{1}t+2\phi(t)]}a_{g}(t) - iS_{e}(t)a_{e}(t) - i\frac{1}{2}\Omega_{er}(t)e^{i[\phi(t)+\Delta_{er}t]}a_{r}(t), \\ \dot{a}_{r}(t) &= -i\frac{1}{2}\Omega_{er}(t)e^{-i[\phi(t)+\Delta_{2}t]}a_{e}(t) - iS_{r}(t)a_{r}(t), \end{aligned}$$
(3.47)

where indices e, g, and r indicate the $|4s\rangle$ excited state, $|3s\rangle$ ground state, and $|7p\rangle$ resonant state, respectively. Δ_1 is the two-photon detuning defined as $\Delta_1 = 2\nu - \omega_{eg}$. Δ_2 is the one photon detuning, $\Delta_2 = \nu - \omega_{re}$. $\Omega_{er}(t)$ is the one-photon Rabi frequency, $\Omega_{er}(t) = -\mu_{er}E_0\sqrt{g(t)}/\hbar$. S_g , S_e , and S_r are the dynamic Stark shifts of the ground, excited, and resonant levels, respectively. S_g and S_e can be written



a) pulse spectrogram at a_2 =-2000 fs²

b) two-photon spectrogram at a_2 =-2000 fs²

Figure 3.5: (a) Spectrogram of a linearly-chirped pulse with $a_2 = -2000 f s^2$. (b) Two-photon spectrogram a linearly-chirped pulse with $a_2 = -2000 f s^2$. (c) Spectrogram at $a_2 = 2000 f s^2$. (d) Two-photon spectrogram at $a_2 = 2000 f s^2$. When the phase of the pulse is given by $\Phi(\omega) = \frac{a_2}{2} (\omega - \omega_0)^2$, a_2 is called linear chirp rate. More specific refers to the subsection 4.2.1.


Figure 3.6: Schematic diagram of 2+1 multi-photon transition

by

$$S_{e,g}(t) = -\sum_{j \neq r} \frac{|\mu_{e,gj}|^2}{2\hbar^2} E_0^2 g(t) \frac{\omega_{je,g}}{\omega_{je,g}^2 - \nu^2}.$$
(3.48)

The only difference of Eq. (3.48) from Eq. (3.28) is that the contribution of the r state is extracted. Also, $\Omega(t)$ is the two-photon Rabi frequency given by

$$\Omega(t) = -\sum_{j \neq r} \frac{\mu_{ej} \mu_{jg}}{2\hbar^2} E_0^2 g(t) \frac{1}{\omega_{jg} - \nu} \simeq -\sum_{j \neq r} \frac{\mu_{gj} \mu_{je}}{2\hbar^2} E_0^2 g(t) \frac{1}{\omega_{je} + \nu},$$
(3.49)

which does not include the contribution of the r state. At 777 nm center-wavelength, two-photon rotating wave approximation² is satisfied, because the two-photon detuning is nearly zero. At 800 nm center-wavelength, the two-photon detuning is calculated as $|\Delta| \approx 0.14 \times 10^{15}$ rad/s. The calculated values of $\omega_{jg} - \nu$ and $\omega_{je} + \nu$ are larger than 0.7×10^{15} rad/s, and therefore the two-photon rotating wave approximation condition in Sodium 3s-4s transition is valid in the experimental situation. We can solve Eq. (3.47) by numerical method such as Runge-Kutta method. In a matrix representation, atomic Sodium interacting with an electric field can be described by the three-level Hamiltonian:

$$\hat{H}(t) = \begin{pmatrix} S_g(t) & \frac{1}{2}\Omega(t)e^{i[\Delta_1 t + 2\phi(t)]} & 0\\ \frac{1}{2}\Omega(t)e^{-i[\Delta_1 t + 2\phi(t)]} & S_e(t) & \frac{1}{2}\Omega_{er}(t)e^{i[\Delta_2 t + \phi(t)]}\\ 0 & \frac{1}{2}\Omega_{er}(t)e^{-i[\Delta_2 t + \phi(t)]} & S_r(t) \end{pmatrix}.$$
(3.50)

This Hamiltonian can be transformed by using a matrix \hat{T} given as $\hat{T} = e^{i[\Delta_1 t + 2\phi(t)]}|g\rangle \langle g| + |e\rangle \langle e| + e^{-i[\Delta_2 t + \phi(t)]}|r\rangle \langle r|$. After the transformation, the Hamiltonian can be written alternatively as $\hat{H}^{(T)} = \hat{T}^{\dagger}\hat{H}\hat{T} - i\hbar\hat{T}^{\dagger}d\hat{T}/dt$. Finally, we obtain the following transformed Hamiltonian describing the

²Two-photon rotating wave approximation is given by $|\Delta| \ll \omega_{jg} - \nu \simeq \omega_{je} + \nu$ where $\Delta = 2\nu - \omega_{eg}$

system,

$$\hat{H}^{(T)}(t) =$$
 (3.51)

$$\begin{pmatrix} S_g(t) + \Delta_1 + 2\dot{\phi}(t) & \frac{1}{2}\Omega(t) & 0\\ \frac{1}{2}\Omega(t) & S_e(t) & \frac{1}{2}\Omega_{er}(t)\\ 0 & \frac{1}{2}\Omega_{er}(t) & S_r(t) - \Delta_2 - \dot{\phi}(t) \end{pmatrix}$$
(3.52)

The diagonal terms can be transformed into the phase of the off-diagonal terms. The transform matrix is given as $\hat{T'} = e^{-i\left[\int^t S_g(u)du + \Delta_1 t + 2\phi(t)\right]} |g\rangle \langle g| + e^{-i\int^t S_e(u)du} |e\rangle \langle e| + e^{-i\left[\int^t S_r(u)du - \Delta_2 t - \phi(t)\right]} |r\rangle \langle r|$. As a result, the final form is obtained as,

$$\hat{H}^{(T')}(t) = (3.53)$$

$$\begin{pmatrix}
0 & \frac{1}{2}\Omega(t)e^{iQ_{1}(t)} & 0 \\
\frac{1}{2}\Omega(t)e^{-iQ_{1}(t)} & 0 & \frac{1}{2}\Omega_{er}(t)e^{iQ_{2}(t))} \\
0 & \frac{1}{2}\Omega_{er}(t)e^{-iQ_{2}(t)} & 0
\end{pmatrix},$$

where $Q_1(t) = -\int^t S_{eg}(u)du + \Delta_1 t + 2\phi(t)$ and $Q_2(t) = -\int^t S_{re}(u)du + \Delta_2 t + \phi(t)$. The level shift parameters are given as $S_{eg}(t) = S_e(t) - S_g(t)$, and $S_{re}(t) = S_r(t) - S_e(t)$. The resonant 7p level is shifted by couplings with s ($\ell = 0$), d ($\ell = 2$) and continuum states. We note that the excited state can be coupled with the continuum. The Ponderomotive energy can be expressed as [41]:

$$S_r(t) = \frac{e^2 E^2(t)}{4m\nu^2},$$
(3.54)

where ν depicts the laser frequency and *m* is the mass of an electron. In the Sodium 3s-4s-7p system, the Ponderomotive energy is 8.5 Trad/s and is roughly two-orders of magnitude above the dynamic Stark shift, because of the presence of lower-lying s and d states [42]. From Eq. (3.53), the probability amplitude of the 7p state can be calculated. The probability amplitude of 7p state is the result of the second order Dyson series, or

$$A_{7p} \simeq \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega_{er}(t) e^{-iQ_2(t)} \int_{-\infty}^{t} dt' \frac{1}{2} \Omega(t') e^{-iQ_1(t')}, \qquad (3.55)$$

where $Q_1(t)$ and $Q_2(t)$ are the atomic phases induced from the level shift, detuning, and phase of the laser pulse. We use the truncation, keeping terms up to $O(t/\tau)$, or

$$\int^{t} S_{eg,re}(u) du = \int^{t} C_{eg,re}g(u) du = \delta_{eg,re}t + O(t^{2}/\tau^{2}).$$
(3.56)

Hence, the probability can be rewritten by

$$A_{7p} = -\sum_{j \neq r} \frac{\mu_{re} \mu_{ej} \mu_{jg}}{8i\hbar^3} \frac{1}{\omega_{jg} - \nu} \int_{-\infty}^{\infty} dt E_0 \sqrt{g(t)} e^{-i[(\nu - \omega_{re} - \delta_{re})t + \phi(t)]}$$

$$\times \int_{-\infty}^{t} dt' E_0^2 g(t) e^{-i[(2\nu - \omega_{eg} - \delta_{eg})t + 2\phi(t)]}.$$
(3.57)

Then, finally, the probability amplitude of the 7p state can be written in frequency domain [47] as

$$a_r = i\pi E(\omega_{re} + \delta_{re}) \int_{-\infty}^{\infty} E(\omega) E(\omega_{eg} + \delta_{eg} - \omega) d\omega, \qquad (3.58)$$

$$a_{nr} = \wp \int_{-\infty}^{\infty} \frac{E(\omega)}{(\omega - \omega_{re} - \delta_{re})} \times \int_{-\infty}^{\infty} E(\omega') E(\omega_{rg} + \delta_{rg} - \omega - \omega') d\omega d\omega'.$$
(3.59)

where \wp is the Cauchy principal value. Indices 'r' and 'nr' denote the resonant and non-resonant parts. δ_{eg} and δ_{re} are the maximum amplitudes of the total level shifts of the 3s-4s and 7p-4s transitions, respectively, when a transform-limited pulse is illuminated. We note that the factor $\sum_{j\neq r} \pi \mu_{re} \mu_{ej} \mu_{jg} / (4\hbar^3(\omega_{jg} - \nu))$ is omitted. The probability amplitude of 4s state can be written up to the third order Dyson series, as

$$A_{4s} = -i \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega(t) \exp(-iQ_1(t))$$

$$+ i \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega(t) \exp(-iQ_1(t)) \int_{-\infty}^{t} dt' \frac{1}{2} \Omega(t') \exp(iQ_1(t')) \int_{-\infty}^{t'} dt'' \frac{1}{2} \Omega(t'') \exp(-iQ_1(t''))$$

$$+ i \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega_{er}(t) \exp(-iQ_2(t)) \int_{-\infty}^{t} dt' \frac{1}{2} \Omega_{er}(t') \exp(iQ_2(t')) \int_{-\infty}^{t'} dt'' \frac{1}{2} \Omega(t'') \exp(-iQ_1(t'')).$$
(3.60)

In the perturbative regime $(\Omega(t)\tau \ll 1 \text{ and } \Omega_{er}(t)\tau \ll 1)$, the first-order term dominates the thirdorder terms. The first-order term is also shown in the two-photon two-level system described in the previous section.

In this section, the probability amplitudes of excited states $|e\rangle$ and $|r\rangle$ in the 3-level Hamiltonian are obtained in pertabative regime. In particular, the probability amplitude of the $|e\rangle$ state can be separated into two parts, resonant and non-resonant contributions. We will compare these probability amplitudes with the experimental results of Sodium 3s-4s-7p ladder system in chapter 6.

3.4 Spatial intensity averaging effects

In this section, we discuss the spatial intensity averaging effect. In the experiment, we use vapor cells of alkali atoms, and because the laser beam has the spatial intensity distribution, many atoms in spatially distributed intensity region interact with the laser pulses. Thus, a measured atom response is the sum of responses from individual atoms interacting with different pulse peak intensities. At the focus, the intensity distribution is given by

$$I(r) = I_0 \exp\left(-\frac{r^2}{d^2}\right),\tag{3.61}$$

where d is the beam radius at the focus. If we define the response from an individual atom at the pulse intensity I as P(I). The total response can be expressed as

$$P_{total} = \int_0^\infty P(I(r)) 2\pi r dr \qquad (3.62)$$
$$= \pi d^2 \int_0^{I_0} \frac{P(I)}{I} dI,$$

where, we substituted the radius r with the peak intensity I. The contribution to the total response depends on the inverse of the pulse peak intensity I. Thus, the response is mainly determined by the weak-field interactions, even though strong-field effects is still present. In order to compare the simulation with the experimental results, the simulation results under I_0 has to be integrated. For example, Fig. 3.7 shows the calculated excited-state population as a function of linear chirp rate a_2 and pulse peak intensity I/I_0 , (a) with and (b) without the intensity averaging effects. At zero chirp rate, the Rabi oscillation as a function of intensity at zero chirp rate is presented in the simulation, but the Rabi oscillation were not observed in our experiment due to the spatial averaging effect. The contribution of weakly interacting



Figure 3.7: Excited-state population plotted as a function of linear chirp rate a_2 and pulse peak intensity I/I_0 , (a) if no spatial intensity averaging effect is considered, and (b) if spatial intensity averaging effect is considered.

atoms is larger than that of strongly interacting atoms 3 . The dip structure caused by level shifts is shown even after the spatial intensity averaging, which is discussed experimentally verified in chapter 5.

³The number of atoms in the interval from I to I + dI depends on the inverse of the pulse peak intensity I. The Rabi oscillation is shown when the pulse area exceeds the minimum π pulse. The pulse area of weakly interacting atoms is much smaller than π .

Chapter 4. Experimental apparatus

In this chapter, we describe the experimental apparatus used in the experiments. The apparatus consists of a Ti-sapphire laser amplifier system, a pulse shaper(DAZZLER), atomic vapor cells, and a fluorescence collecting system, as shown in Fig. 4.1. We use vapor cells of Cesium (TT-CS-75-V-Q-CW, Triad technology) and Sodium (CP25075-NA, Thorlab Inc.). For the investigate of multi-photon absorption in strong-field regime, the density of Cesium vapor, 2.89×10^{16} m⁻³ is sufficient to observe fluorescence by photomultiplier tube (PMT) in room temperature (20° C.). However, the density of Sodium in room temperature is 3.76×10^{11} m⁻³ and the Sodium vapor cell needs to be heated. For the heating, the Sodium vapor cell was prepared as shown in Fig. 4.2. The gradient heating of the vapor cell prevents metal deposition on the view windows, where the temperature of the sides of the cell is kept higher than that of the center. Thus, the vapor is driven into the center of the cell. In the experiments, the Sodium cell is heated about 130° C for the atomic density of 1.92×10^{11} m⁻³. The temperature of the Sodium cell is measured by a thermistor (PTC100, Thorlab Inc.) of which the resistance is a function of temperature. The temperature controller (TC200, Thorlab Inc.) converts the resistance to digitized numbers.

4.1 Pulse generation and measurement

We use a home-built femtosecond laser amplifier system, which produces short pulses of 35-90 fs pulse width and $100\mu J$ pulse energy at 1 kHz repetition rate. The seed beam for the amplifier system is generated from the oscillator (KM-LAB) operating at 90 MHz, and its pulse width is 35-50 fs. The wavelength of laser pulses is chosen in the range from 782 to 830 nm. The pulses from the amplifier system are characterized by a second harmonic frequency-resolved optical gating (SHG FROG) [43], and also by an autocorrelator (Mini autocorrelator, APE). The intensity of the SHG is given as

$$I^{SHG}(\omega,\tau) = \left| \int_{-\infty}^{\infty} E(t)E(t-\tau)e^{-i\omega t}dt \right|^2.$$
(4.1)

By measuring the SHG spectra as a function of delay τ , the corresponding FROG trace is generated. We measure the SSG spectrum in a spectrometer (USB2000, Ocean Optics Inc.) of which the spectral resolution is 0.3 nm. The spectral (or temporal) amplitude and phase are retrieved from the FROG trace using a FROG algorithm. Figure. 4.3 shows the schematic diagram of a FROG setup to measure the pulse characteristics. The BBO crystal used in the experiments has angle cuts at $\theta = 29.2^{\circ}$ and $\phi = 90^{\circ}$, and the thickness is 100μ m. As an example, the FROG signal of a linearly chirped pulse generated from the laser system is shown in Fig. 4.4. After applying the FROG algorithm, the amplitude and phase, in spectral and temporal domains, are retrieved as shown in Figs. 4.4(b) and (c), respectively.

4.2 Pulse shaping

We use an acousto-optic programmable dispersive filter (AOPDF) [36] as a pulse shaper which is used to program the phase and amplitude of ultrafast pulses. The structure of the AOPDF (DAZZLER)



Figure 4.1: Experimental setup for shaped-pulse two-photon excitation of Alkali atoms



Figure 4.2: Schematic diagram of gradient temperature heating structure of Sodium cell



Figure 4.3: Schematic diagram of FROG (frequency-resolved optical gating) setup.

used in the experiments is shown in Fig. 4.5. AOPDF overcomes the two limitations of spatial light modulators: the coupling between the spatial and temporal aberrations of laser beam, and small dispersion compensation ranges. Figure 4.6 shows the schematic diagram of AOPDF, which is based on a collinear acousto-optic interaction. Acoustic waves are generated by the transducer, and propagates along the z-axis in a 25-mm-long TeO₂ crystal. The speed of sound in the crystal is 1000 m/s, and the index difference is $\Delta n = 0.04$. Along the z-direction, an acoustic spatial grating is produced. The frequency component of the pulse that travels a certain distance meets a phase-matched frequency of an acoustic wave, and diffracted into an extraordinary mode at the phase-matched position. The traveling distance determines the phase of each frequency component, and the amplitude of each frequency component is determined by diffraction efficiency, that is controlled by the acoustic power at the phase-matched position. In the following, we provide coupled-wave analysis of AOPDF. The dielectric tensor in a TeO₂ crystal is given by

$$\epsilon(z) = \epsilon + 2\epsilon_1(z)\cos(\psi(z)), \tag{4.2}$$

and the acoustic phase given by

$$\psi(z) = \int_0^z K(z)dz, \qquad (4.3)$$

where K(z) is the instant spatial frequency of the acoustic wave, usually a monotonic function of z. ϵ is the dielectric tensor in absence of an acoustic wave. $\epsilon_1(z)$ is the perturbed dielectric tensor resulted from the optical mode coupling. We note that ϵ_1 is in general much smaller than ϵ . The optical pulses can be represented as a Fourier sum of spectral components, i.e.,

$$\mathbf{E}(t,\mathbf{r}) = \int \mathbf{E}(\omega,\mathbf{r})\exp(i\omega t)d\omega.$$
(4.4)

When no acoustic wave is applied, $\epsilon_1 = 0$. Then,

$$\mathbf{E}(\omega, \mathbf{r}) = \sqrt{S(\omega)} \left[A_1(z) \mathbf{e}_1 \exp(-ik_1(\omega)z) + A_2(z) \mathbf{e}_2 \exp(-ik_2(\omega)z) \right], \tag{4.5}$$



Figure 4.4: (a) FROG tracer of a linearly-chirped pulse of 20000 fs^2 ; (b) Retrieved spectral amplitude and phase; (c) Retrieved temporal amplitude and phase.

where $k_{1,2} = n_{1,2}(\omega)\omega/c$, and

$$\mathbf{e}_{1,2} = \left(\frac{2\mu_0\omega}{k_{1,2}(\omega)}\right)^{1/2} \mathbf{p}_{1,2}$$
(4.6)

where $\mathbf{p}_{1,2}$ are the unit polarization vector of optical waves (e.g. ordinary and extraordinary axes). In the crystal, the wave equation is given by

$$\frac{d^2}{dz^2}\mathbf{E}(\omega,\mathbf{r}) + \omega^2 \mu_0 \epsilon(z) \mathbf{E}(\omega,\mathbf{r}) = 0.$$
(4.7)

By inserting Eq. (4.5) into Eq. (4.7), the optical wave satisfies the following coupled-wave equation [37]:

$$\frac{d}{dz}A_{1}(z) = -i\kappa(z)A_{2}(z) \left[\exp(-i\phi_{+}(z)) + \exp(-i\phi_{-}(z))\right], \qquad (4.8)$$
$$\frac{d}{dz}A_{2}(z) = -i\kappa(z)A_{1}(z) \left[\exp(i\phi_{+}(z)) + \exp(i\phi_{-}(z))\right],$$

where

$$\phi_{\pm}(z) = [k_2(\omega) - k_1(\omega)]z \pm \psi(z), \qquad (4.9)$$

$$\kappa(z) = \frac{\omega^2 \mu_0}{2[k_1(\omega)k_2(\omega)]^{1/2}} \mathbf{p_1} \cdot \epsilon_1(z) \cdot \mathbf{p_2}.$$
(4.10)

It is well known that energy transfer is possible between ordinary and extraordinary modes if phase matching is satisfied between optical and acoustic waves [40], i.e.,

$$\frac{d}{dz}\phi_{\pm}(z) = 0 = k_2(\omega) - k_1(\omega) \pm K(z).$$
(4.11)

The choice of the sign depends on the sign of $n_2(\omega) - n_1(\omega)$, the difference between refractive indices of the two optical modes. For example, when $n_2(\omega) - n_1(\omega) < 0$, there exists an unique position $z(\omega)$ where the energy transfer occurs between the two optical modes, given by

$$K(z(\omega)) = \frac{\omega}{c} (n_1(\omega) - n_2(\omega)), \qquad (4.12)$$

which explains that an angular frequency ω is diffracted at position z of which the spatial frequency of the acoustic wave is determined by $\frac{\omega}{c}(n_1(\omega) - n_2(\omega))$. In other words, the energy transfer for a certain optical frequency occurs at a position where a proper acoustic wave is generated. The phase of each frequency component is determined by the traveling distance to the phase matching position. Also, the acoustic power at the phase matching position determines the amplitude of the diffracted optical frequency component. Hence the output electric field can be written in frequency domain as

$$E_{out}(\omega) \propto E_{in}(\omega)S(\alpha\omega),$$
 (4.13)

where α is the scaling factor, which satisfies $\alpha = \Delta n(V/c)$. V is the speed of acoustic wave, and Δn is the index difference between the ordinary and extraordinary waves. The arbitrary output pulse is programmed by a proper acoustic wave function $S(\alpha\omega)$. After the Fourier transform, we obtain the output electric field in time domain. The output signal in time domain is proportional to the convolution of the input electric field and the scaled acoustic wave [36], or

$$E_{out}(t) \propto E_{in}(t) \otimes S(t/\alpha).$$
 (4.14)

In the experiment, α is about 10^{-7} . We note that the spectral resolution of DAZZLER in wavelength scale is 0.2 nm at 822 nm. We generate complex shape pulses, such as linearly-chirped and quadratically-chirped pulses from the DAZZLER. The specifications are following in the next subsections.



Figure 4.5: Top view of Dazzler. The output beam is diffracted by 1 degree from the input beam, in the 90-degree-rotated linear polarization.



Figure 4.6: Schematic diagram of acousto-optic programmable dispersive filter (AOPDF) [36]

4.2.1 Linearly-chirped pulses

Linearly-chirped pulses are widely used in coherent control experiments. The instant frequency of a linearly-chirped pulse is given as a linear function of time t. The pulse shaper provides three modes: 'dial and polynomial', 'file', and 'both'. For an output electric field is given by $E_{out}(\omega) = E_{in}(\omega)A(\omega)e^{i\Phi(\omega)}$, where

$$A(\omega) = A_0 \exp(-(\omega - \omega_0)^2 / \delta \omega_0^2), \qquad (4.15)$$

$$\phi(\omega) = -a_1(\omega - \omega_0) - \frac{a_2}{2}(\omega - \omega_0)^2 - \frac{\tilde{a}_3}{6}(\omega - \omega_0)^3 - \frac{a_4}{24}(\omega - \omega_0)^4,$$

a chirped pulse can be obtained by giving a_1 , a_2 , \tilde{a}_3 , and a_4 parameter in the 'polynomial' mode. Here, ω_0 is the center frequency and $\delta\omega_0$ is the bandwidth of pulse shaper modulation function, $S(\alpha\omega)$. a_1 is time delay. a_2 , \tilde{a}_3 , and a_4 are, respectively, the second, third, and fourth order chirps in frequency domain. The out electric field in time-domain is obtained by Fourier transformation of $E_{out}(\omega)$. For example, a_1 , \tilde{a}_3 , and a_4 are fixed at zero and only a_2 varies, to make a linearly chirped pulse.

We consider that the input electric field is given by $E_{in}(\omega) = E_0 \exp(-(\omega - \omega_0)^2/\Omega^2)$, and assume that the bandwidth of the input electric field (20-30 nm) is much smaller than the bandwidth of modulating signal $A(\omega)$ (50-70 nm) i. e., $\Omega \ll \delta\omega_o$. Then, the amplitude modulation $A(\omega)$ can be regarded as constant. Hence the Fourier transform of $E_{out}(\omega)$ is given by

$$E_{out}(t) = E_0 A_0 \sqrt{\frac{\pi}{\frac{1}{\delta\omega_0^2} + i\frac{a_2}{2}}} \exp\left(-\frac{t^2}{4\delta\omega_0^2 \left(\frac{1}{\delta\omega_0^4} + \frac{a_2^2}{4}\right)}\right) \exp\left(i\omega_0 t + i\frac{a_2 t^2}{8\left(\frac{1}{\delta\omega_0^4} + \frac{a_2^2}{4}\right)}\right)$$
(4.16)
= $E_1 \exp\left(-t^2/\tau^2\right) \exp(i\omega_0 t) \exp(i\beta t^2).$

From the above equation, a set of relations are obtained:

$$\tau_{0} = \frac{2}{\delta\omega_{0}}, \quad \tau = \tau_{0}\sqrt{1 + 4\frac{a_{2}^{2}}{\tau_{0}^{4}}}, \quad \beta = \frac{2a_{2}}{\tau_{0}^{4} + 4a_{2}^{2}},$$

$$E_{1} = E_{0}A_{0}\sqrt{\frac{2}{\tau_{0}^{2} + i2a_{2}}}, \quad |E_{1}|^{2}\tau = \text{constant.}$$

$$(4.17)$$

Additionally,

$$I_0 \tau_0 = I\tau, \quad \beta \tau^2 = \frac{2a_2}{\tau_0^2}, \tag{4.18}$$

where I_0 and I are the pulse peak intensities of the unshaped pulse and of the linearly-chirped pulse, respectively. τ_0 and τ are the temporal pulse widths of the unshaped and the linearly-chirped pulses, respectively. β is the time-domain linear chirp parameter. In the spectrogram of a linearly chirped pulse, 2β corresponds to the slope of frequency sweeping.

The pulse shaper has the damage threshold at 100 MW/cm^2 , which is significantly lower than the laser peak power from the laser amplifier. To avoid the damage, the pulse shaper is inserted in the laser amplifier itself, before the compressor, and the pulse is stretched to pass through the pulse shaper. This method keeps the pulse peak intensity below the damage threshold, and requires a new calibration of chirp rates because the pulse is stretched.

In general, the modulation and phase functions are not controlled independently in a pulse shaper. For example, the laser power is significantly reduced at the lime of zero linear chirp, times smaller than laser powers at non-zero input values. To keep the laser power at constant for various shaped pulses, the 'constant gain' function of the pulse shaper is used, which maintains the diffraction efficiency for the output beam nearly constant.



Figure 4.7: Theoretical and measured pulse durations plotted as a function of linear chirp rate a_2 . Gaussian width is fitted by $\tau = \tau_0 \sqrt{1 + (2a_2/\tau_0^2)^2}$.

4.2.2 Quadratically-chirped pulses

In quadratically-chirped pulses, the temporal phase of pulses has t^3 dependence, i.e. the frequency sweeping has t^2 dependence. Quadratically-chirped pulses are realized in 'file mode' in the pulse-shaper control program. First, two N×2 spreadsheets of amplitude and phase, respectively, are prepared as a function of wavelength. Figure 4.8 shows the examples of spectral phase and amplitude of quadraticallychirped pulses. Then, the necessary acoustic wave is calculated by the control program and the device loads the acoustic wave on the AOM crystal. For a quadratically-chirped pulse, the amplitude and phase of an electric field in frequency domain can be obtained by the Fourier transform of the time-domain electric field

$$E_{out}(t) = \frac{1}{2} E_0 \sqrt{g(t)} e^{i\nu t} e^{i(a_1 t + a_3 t^3)} + c.c., \qquad (4.19)$$

and $A(\omega)\exp(i\Phi(\omega))$ is calculated by using the following equation,

$$E_{out}(\omega) = A(\omega)\exp(i\Phi(\omega)) \times E_{in}(\omega), \qquad (4.20)$$

where $E_{in}(\omega)$ is the spectral amplitude of the input pulse, a Gaussian function of 30 nm width and $E_{out}(\omega)$ is the Fourier transform of $E_{out}(t)$. The spectral information, $A(\omega)\exp(i\Phi(\omega))$, is programmed into the pulse shaper, and then the cubic phase, $\phi(t) = a_1t + a_3t^3$, is obtained.

4.3 Fluorescence detection

Shaped pulses are focused into a vapor cell by a lens of 125 mm focal length. The focused beam has a transverse spatial intensity profile, so the laser intensity in the interaction region is spatially nonuniform. The PMT detects all the photons from the interaction region, and the detected fluorescence is



Figure 4.8: Examples of spectral phase and amplitude programmed for pulses with temporal cubic phase: (a) $a_3\tau^2 = 5$ Trad/s, (b) $a_3\tau^2 = -5$ Trad/s. (c) Spectrogram of shaped pulse with $a_3\tau^2 = 5$ Trad/s, and (d) with $a_3\tau^2 = -5$ Trad/s.

the sum of fluorescence from the all atoms in the interaction region as follows

$$P_{total} = \int_0^\infty dr \int_{-z_R}^{z_R} dz 2\pi r \rho P(I(r, z)),$$
(4.21)

where z_R is the Rayleigh range of the laser beam, and ρ is the atomic vapor density. Considering this intensity averaging effect, the beam intensity I(r, z) is given by

$$I(r,z) = I_0 \frac{\omega_0^2}{\omega^2(z)} \exp\left(-\frac{r^2}{\omega^2(z)}\right),$$
(4.22)

where the beam waist $\omega(z)$ is $\omega(z) = \omega_0 \sqrt{1 + (z/z_R)^2}$. In the experiment, the beam diameter at the focus is $50\mu m$ and the Rayleigh range is 2mm. The number of Cesium atoms in this interaction region is 4.5×10^5 in room temperature. The number of Sodium atoms in 423 K is 3.0×10^6 . The focused spot on the vapor cell is imaged by a two-lens telescope (2-inch f=50 mm and 100 mm), installed before the photomultiplier tube (PMT). Additionally, a band-pass spectral filter is inserted before the PMT to block undesired fluorescence. BG13 is used as a blue-color filter, and also, a 476.5 nm bandpass filter of 10 nm bandwidth (F10-476.5, CVI), and a 590 nm band pass filter (F10-590, CVI) are used in various experiments. An aperture of 1 mm diameter is used to block undesired fluorescence from outside of the interaction region. The fluorescence is detected by the PMT, in which current is generated proportionally to the number of incident photons. The PMT signal is amplified by a low-noise pre-amplifier (SR570, Stanford research systems), which converts the current signal into a voltage signal. Then, the converted voltage signal is digitized by the digital lock-in amplifier (SR810, Stanford research systems), with a modulation frequency of 1 kHz.

Chapter 5. Coherent quantum control of two-photon absorptions in Cesium

In this chapter, we describe the experimental results of shaped-pulse excitation in atomic Cesium. The experimental results with the cesium atom interacting with shaped laser pulses, either linearlychirped or quadratically-chirped, are compared with our formula obtained in chapter 3. Cesium is a widely used atom in multi-photon absorption experiments. Its melting point is 301.59 K and the boiling point is 944 K. In our experiments at room temperature 293K, Cesium is solid. The atomic number of Cesium is 55 and the ground state of Cesium is the 6s state. The laser used to excited the 8s state via two-photon absorption has the center wavelength at 822 nm and the bandwidth of 30 nm. The energy between the intermediate state 6p and the ground state is 852 nm in wavelegnth scale. Hence, atomic Cesium can be considered as a two-level system. The specific energy level diagram is given in Fig. 5.1. The life-time of Cesium 8s state is about 90 ns, which is much longer than the atom-pulse interaction time. The vapor-pressure of vapor cell at solid phase is given as [45]:

$$\log_{10} P_v = 2.881 + 4.711 - \frac{3999}{T},\tag{5.1}$$

where the vapor pressure P_v is in Torr and the temperature T is in K. At the room temperature 293 K, the vapor pressure is 8.78×10^{-7} Torr. The corresponding number density calculated by P/k_BT , is $2.89 \times 10^{16}/m^3$. The mean free path is a function of temperature T and pressure P:

$$l = \frac{k_B T}{\sqrt{2\pi} d^2 P},\tag{5.2}$$

where d is the diameter of the gas particles. The van der Waals radius of Cesium is 343 pm, the mean free path in the room temperature is 16.5 m, and the most probable speed is 191.4 m/s at 293 K. Thus, the collisional coherence time is obtained as 86 ms. Therefore, the collision does not break the coherence within the pulse-atom interaction time scale in a few tens of femtoseconds.. The ground 6s state and the excited 8s state are coupled to the intermediate states with angular momentum quantum number l > 0, which are far off resonance. Table. 5.1 shows transition dipole moments and transition energies for Cesium used in the calculation.

The couplings to the intermediate states with $\ell > 1$ are neglected because the ground and excited states are of $\ell = 0$ angular momentum. From the selection rules, $\ell_j - \ell_i = \pm 1$, the ground $6s_{1/2}$ and excited $8s_{1/2}$ states are only coupled with the *p* states. By Eq. (3.28), the calculated dynamic Stark shifts are given in Table 5.1.

5.1 Two-photon excitation induced by linearly-chirped pulses

For a simple picture of the strong-field two-photon absorption, we consider control experiments with shaped laser pulses. Figure 5.2 depicts the schematics of experiments, with (a) a transform-limited pulse, (b) a positively-chirped pulse, (c) a negatively-chirped pulse, and (d) a red-detuned pulse. The corresponding energy diagrams of Cesium are shifted temporally by the laser fields, as illustrated in the time and frequency plane. The spectrograms of the IR laser pulses are drawn in red and their two photon



transition	dipole moment	energy	DSS
	$(\times 10^{-29} \mathrm{Cm})$	$(\times 10^{15} \text{ rad/s})$	(Trad/s)
$6s_{1/2} - 6p_{1/2}$	2.70	2.11	65.21
$6s_{1/2} - 6p_{3/2}$	3.81	2.21	296.05
$6s_{1/2} - 7p_{1/2}$	0.17	4.10	-0.03
$6s_{1/2} - 7p_{3/2}$	0.35	4.10	-0.15
$6s_{1/2} - 8p_{1/2}$	0.05	4.85	0.00
$6s_{1/2} - 8p_{3/2}$	0.13	4.86	-0.02
$8s_{1/2} - 6p_{1/2}$	-0.62	-2.48	3.59
$8s_{1/2} - 6p_{3/2}$	-0.88	-2.38	15.11
$8s_{1/2} - 7p_{1/2}$	-5.57	-0.49	-10.28
$8s_{1/2} - 7p_{3/2}$	-8.43	-0.49	-23.54
$8s_{1/2} - 8p_{1/2}$	10.66	0.26	19.31
$8s_{1/2} - 8p_{3/2}$	14.72	0.27	38.27
$8s_{1/2} - 9p_{1/2}$	1.05	0.62	0.48
$8s_{1/2} - 9p_{3/2}$	1.79	0.63	1.41
$8s_{1/2} - 10p_{1/2}$	0.38	0.83	0.09
$8s_{1/2} - 10p_{3/2}$	0.70	0.83	0.30

Figure 5.1: Energy level of Cesium for two-photon absorption [46]

Table 5.1: Dipole moments and transition energies of atomic Cesium. The dynamic Stark shifts (DSS) are calculated for the laser intensity at 1×10^{15} W/m². The line-strengths are from NIST atomic database [46].



Figure 5.2: Schematics of strong-field two-photon excitation of ground-state atomic cesium with various shaped laser pulses. The spectrogram of the laser pulses and of their two-photon spectrogram are depicted in red and blue, respectively. The magenta arrows indicate the excitation paths for the strong interaction cases, while the yellow arrows do for the weak interaction cases.



Figure 5.3: (a) Measured 7p-6s fluorescence signal induced by two-photon absorption in atomic cesium is plotted in color (red is the biggest) as a function of linear chirp rate a_2 and scaled TL(Transform-Limited) peak intensity I_0/I_c . The equi-signal levels are traced by contour lines which are reconstructed by a model calculation with best fit parameters. The typical behaviors of the signals are shown in the strong- and weak-field regimes in (b) and (c), respectively. Note that $I_c = 0.25 \times 10^{11} W/cm^2$.

spectrograms are in blue. Figure 5.2(a) shows the case when an unshaped pulse (transform-limited pulse) strongly interacts with atoms. The dynamic Stark effect causes the two-photon excitation temporally off-resonant from the two photon energy of the laser field. As a result, the most photons at the peak of the laser pulse do not induce atomic excitation. The excitation is expected to happen not at the peak of the pulse, but more in both the head and tail of the temporal profile of the pulse. On the other hand, in Fig. 5.2(b) and 5.2(c), the laser pulses have positive and negative frequency chirps, but of the same pulse energies as in Fig. 5.2(a). These pulses shift the atomic energy levels less than Fig. 5.2(a)and, thereby, the atom-field resonant conditions for the two-photon absorption are better maintained. In particular, the Stark-shift energy levels can resonantly cross the spectro-temporal field densities in the two-photon spectrograms (drawn in blue). We expect, therefore, to achieve stronger atomic excitations by both positively- and negatively-chirped pulses, than by a transform-limited pulse in Fig. 5.2(a). We note that the atomic excitation is expected to occur earlier in time than the peak of the pulse for the positively-chirped pulse as in Fig. 5.2(b), and later for the negatively-chirped pulse in Fig. 5.2(c). The Stark shift can be pre-compensated by a frequency-detuning of the laser pulse as illustrated in Fig. 5.2(d). Then, the atom-field resonant condition is satisfied at the peak of the pulse, and the pre-detuned pulse excites more than a pulse of zero detuning.

To estimate the magnitude of the two-photon absorption, we calculate the pulse area θ_f

$$\begin{aligned} |\theta_f|^2 &= \left| \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega(t) \exp(i\alpha(t)) \right|^2 \\ &= \left| \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega_c g(t) \exp\left[i(-\int_{-\infty}^t \delta_c g(t') dt' + \Delta t + \beta t^2) \right] \right|^2, \end{aligned}$$
(5.3)

where the pulse envelope g(t) is given as a Gaussian function, $\exp(-t^2/\tau^2)$. Ω_c and δ_c are the amplitudes of two-photon Rabi frequency and dynamics shift, respectively, which are proportional to pulse intensity. The Stark shift S(t) induces phase accumulation given by

$$\int_{-\infty}^{t} S(t')dt' \simeq \int_{-\tau}^{t \le \tau} \delta_c g(t')dt' = \frac{\sqrt{\pi}}{2} \delta_c \tau \left[1 + \operatorname{erf}\left(t/\tau\right)\right],\tag{5.4}$$

where $\operatorname{erf}(x)$ is an error function, defined as

$$\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x \exp\left(-t^2\right) dt$$

$$= \frac{2}{\sqrt{\pi}} \sum_{n=0}^\infty \frac{(-1)^n}{(2n+1)n!} x^{2n+1}.$$
(5.5)

To simplify the integral in Eq. (5.3), we truncate $O(t^3/\tau^3)$ terms, which is valid for Gaussian pulses. After the truncation, the pulse area is calculated as

$$\begin{aligned} |\theta_f|^2 &= \left| \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega_c \exp\left[-\left(\frac{t}{\tau}\right)^2 \right] \exp\left[i(\Delta \tau - \delta_c \tau) \frac{t}{\tau} + i\beta \tau^2 \left(\frac{t}{\tau}\right)^2 \right) \right] \right|^2 \end{aligned} \tag{5.6} \\ &= \left(\frac{\Omega_c \tau}{2}\right)^2 \left| \int_{-\infty}^{\infty} dX \exp\left[-X^2 + iAX + iBX^2 \right] \right|^2 \\ &= \left(\frac{\Omega_c \tau}{2}\right)^2 \frac{\pi}{\sqrt{1+B^2}} \exp\left(-\frac{A^2}{2(1+B^2)} \right), \end{aligned}$$

where $A = (\Delta - \delta_c) \tau$ and $B = \beta \tau^2 = 2a_2/\tau_0^2$.

 $|\theta_f|^2$ is given as a product of two competing functions of linear chirp rate $a_2 = 0$: the first term $(\Omega_c \tau/2)^2 \pi/\sqrt{1+B^2}$ decreases as a function of $|a_2|$, and the second term $\exp\left(-A^2/(2(1+B^2))\right)$ is an increasing function of $|a_2|$. We first consider Δ is zero. Then, unless the dynamic Stark shift is sufficiently small, or if the laser intensity is strong enough, θ_f has double maxima, symmetrically located around $a_2 = 0$. In this case, the pulse area is given as

$$|\theta_f|^2 = \pi (\frac{\Omega_c \tau}{2})^2 \frac{1}{\sqrt{1+B^2}}.$$
(5.7)

In comparison, the experimental result is shown in Fig. 5.3. The fluorescence measured as a function of linear chirp rate at various scaled intensity (I_0/I_c) , of laser pulse, is presented in Fig. 5.3 (a). It shows that a single peak is transformed to the double peaks with growing pulse intensities. The experimental fluorescence is well fitted with Eq. (5.6). To estimate the onset intensity between the weak and strong fields, we approximate Eq. (5.6) around the zero chirp rate,

$$|\theta_f|^2 = \pi (\frac{\Omega_c \tau}{2})^2 \left[1 + \frac{1}{2} (A^2 - 1) B^2 \right] \exp\left(-\frac{A^2}{2}\right).$$
(5.8)

The term of $O(B^4)$ can be neglected because B is very small. A = 1 is the boundary between the single and double peaks. The onset intensity, the pulse peak intensity at the boundary, is given by

$$I_c = \frac{1}{\eta \tau},\tag{5.9}$$

where $\eta = -50.4$ Trad/s at the $I_0 = 1 \times 10^{11} W/cm^2$. The onset intensity is calculated as $I_c = 0.25 \times 10^{11} W/cm^2$. Over the onset intensity, double peaks appear as shown in Fig. 5.3 (b), while below, a single peak as in Fig. 5.3 (c).

The time-evolution of the excited state in various chirp rates is given in Fig. 5.4. For $I/I_c = 4$, the Schrödinger equation is solved numerically, and shows that, for the maximal two-photon excitation, a pulse with zero chirp rate, or the transform-limited pulse, is not an optimal solution.

Figure 5.5 compares the measured fluorescence and calculation, both given as a function of linear chirp rate at various pulse intensities. The numerical simulation is obtained by solving the Schrödinger equation. Figures 5.5 (a) and (b) are results in the weak-field regime (level shifts are negligible) and (c)-(f) are in the strong-field regime.

In this section, we found out that a transform-limited pulse is not optimal for strong-field twophoton absorption. Because a transform-limited pulse induces the highest dynamic Stark shift, it causes the largest off-resonance. Hence the two-photon excitation is reduced. The experimental results show a good agreement with the theoretical prediction.

5.2 Two-Photon excitation induced by frequency-detuned pulses

The two-photon excitation by frequency-detuned pulses is also described by Eq. (5.6), where the parameter A contains the detuning term Δ and the dynamic Stark shift δ_c . If A is nonzero, the pulse area has double peaks as verified experimentally in Fig. 5.6. At a proper detuning $\Delta = \delta_c$, A goes to zero. Then, a single peak is recovered. Figure 5.6 (a) shows the calculated two-photon absorption of Cesium at the pulse intensity $I = 1.43I_c$. Around the maximum point $(0, \delta_c)$ (i.e., A = 0, B = 0) in the chirp rate-detuning space, the equi-signal line in Fig. 5.6 follows the equation of a looped curve (a heart shape) given as

$$C = \frac{1}{2}X^2 - \frac{1}{2}\delta_c \tau X^2 Y + \frac{1}{2}Y^2, \qquad (5.10)$$

where C is a constant, $X = 2a_2/\tau_0^2$, and $Y = (\Delta - \delta_c)\tau_0$. In the weak field regime, the level shift δ_c goes to nearly zero. Then the looped curve becomes the circle centered at the origin. If the pulse intensity is enough to induce the sufficiently large dynamic Stark shift, the circle is distorted and becomes a heart shape centered $(0, \delta_c)$. Around δ_c , the single peak is recovered and the maximum fluorescence is obtained at zero chirp rate even in the strong field regime. Because the overall dynamic Stark shift is negative in Cesium, the detuning to compensate the level shift is also negative. When the linear chirp rate a_2 is fixed at zero, Eq. (5.6) is simplified as

$$|\theta_f|^2 = \pi (\frac{1}{2}\Omega_c \tau)^2 \exp\left(-\frac{1}{2}(\delta_c - \Delta)^2 \tau_0^2\right).$$
 (5.11)

The detunings Δ to compensate the level shifts δ_c are 2.8 nm, 4.3 nm, and 6.2 nm for $I_0 = 0.51I_c$, $0.77I_c$, and $1.03I_c$, respectively. Note that δ_c and Ω_c are proportional to the pulse peak intensity. As shown in Fig. 5.8, the detuning for compensation of the dynamic Stark shift is proportional to I_0 , showing good agreement with Eq. (5.11).

5.3 Two-photon excitation induced by quadratically-chirped pulses

In the previous section, we have considered the two-photon excitation with linearly-chirped pulses and detuned pulses. We found out that linearly-chirped pulses with a certain linear chirp rate leads to



Figure 5.4: Calculated time evolution of the excited population by solving the Schrödinger equation when linearly chirped pulses are applied. (a) Experimental fluorescence and calculation; (b) Time evolution of Cesium 6s and 8s states at zero chirp rate. (c) At linear chirp rate of $a_2 = -8000 f s^2$. (d) $a_2 = 8000 f s^2$. (e) $a_2 = -20000 f s^2$. (f) $a_2 = 20000 f s^2$. The pulse intensity is fixed at $I_0/I_c = 4$, and the centerwavelength $\lambda = 818$ nm.



Figure 5.5: Fluorescence as a function of linear chirp rate at various pulse intensities. The solid-black line in the experimental data represents the pulse area calculation. The center-wavelength λ is 822 nm



Figure 5.6: (a) Calculated 7p-6s fluorescence signals of atomic cesium in color (white is the biggest), plotted as a function of linear chirp rate and detuning. The transform-limited intensity is $I_0 = 1.43I_c$. (b) Measured 7p-6s fluorescence as a function of chirp rate

and enhancement of the two-photon absorption above the result with zero chirp rate. In this section, we consider quadratically-chirped pulses. In particular, quadratically-chirped pulses maintain the resonance condition in a longer period of time than what was possible by linearly-chirped pulses. An electric field which has a temporal cubic phase is given by

$$E(t) = \frac{1}{2} E_0 \sqrt{g(t)} e^{i\nu t} e^{i\phi(t)} + c.c., \qquad (5.12)$$

where $\phi(t) = a_1 t + a_3 t^3$ and $g(t) = \exp(-t^2/\tau^2)$. To estimation the probability of the two-photon excitation, we calculate the square of the pulse area (excitation probability in perturbative regime):

$$P_e = |\theta_f|^2 = \left| \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega_c g(t) \exp\left[i \left(-\int_{-\infty}^t \delta_c g(t') dt' + \Delta t + 2\phi(t) \right) \right] \right|^2, \tag{5.13}$$

where δ_c is the amplitude of the level shift, and Ω_c is the amplitude of the two-photon Rabi frequency, respectively, -50.4 Trad/s (effective level shift) and -14 Trad/s (effective two-photon Rabi frequency) at $I_0 = 1.0 \times 10^{11} \text{W/cm}^2$. We keep the terms of an order of $(t/\tau)^3$. Then, the pulse area can be written as

$$P_{e} = |\theta_{f}|^{2}$$

$$= \left| \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega_{c} e^{-(t/\tau)^{2}} \exp \left[i \left(2\phi(t) + (\Delta - \delta_{c})t + \frac{1}{3} \frac{\delta_{c}}{\tau^{2}} t^{3} \right) \right] \right|^{2}$$

$$= \left| \int_{-\infty}^{\infty} dt \frac{1}{2} \Omega_{c} e^{-(t/\tau)^{2}} \exp \left[i (-\delta_{c}\tau + \Delta\tau + 2a_{1}\tau) \frac{t}{\tau} + i \left(2a_{3}\tau^{3} + \frac{\delta_{c}\tau}{3} \right) \left(\frac{t}{\tau} \right)^{3} \right) \right] \right|^{2}$$

$$= \left(\Omega_{c}\tau/2 \right)^{2} \left| \int_{-\infty}^{\infty} \exp \left[-x^{2} + i\eta x + i\xi x^{3} \right] dx \right|^{2},$$
(5.14)





Figure 5.7: Comparison between the result of the numerically solved Schrödigner equation and the experimental data with the pulse-area calculation at $I_0/I_c = 1.43$.



Figure 5.8: Measured 7p-6s fluorescence signals in symbols as a function of detuning at various fixed peak intensities, compared with numerical fits in lines using Eq. (5.11).

where $x = t/\tau$, $\eta = (-\delta_c + \Delta + 2a_1)\tau$, and $\xi = (2a_3\tau^2 + \delta_c/3)\tau$. Both Eq. (5.14) and Eq. (5.13) show a good agreement with the experimental data.

The phase matching condition for the two-photon absorption is given by

$$2\phi(t) = (-\Delta + \delta_c)t - \frac{1}{3}\frac{\delta_c}{\tau^2}t^3.$$
 (5.15)

This phase matching condition is consistent with the conditions of $\eta = 0$ and $\xi = 0$ which maximize P_e . For the laser detuning $\Delta = 0$, the first and third order terms of the dynamic Stark shift can be compensated by the conditions of $a_1 = \delta_c/2$ and $a_3\tau^2 = -\delta_c/6$. Figure 5.9 shows the probability of strong-field two-photon excitation from Eq. (5.14). The point O in Fig. 5.9(a) corresponds to the unshaped transform-limited pulse, and O_p corresponds to the optimal pulse shape($\eta = \xi = 0$). The spectro-temporal shape at O_p can be illustrated as in Fig. 5.9(e). The control parameter a_1 in η and a_3 in ξ denote the frequency offset(detuning) and the frequency curvature in a spectrogram. Therefore, the change of η along the path \overline{OA} in Fig. 5.9(a) is the frequency detuning as illustrated in Fig. 5.9(c). Also, the change of ξ along \overline{AB} is the frequency curvature control as shown in Fig. 5.9(d).

The verification of the control schemes illustrated in Fig. 5.9 is carried out by measuring the 7p-6s fluorescence as a function of a_1 and $a_3\tau^2$. Fig. 5.11 (a) shows the line shapes of the signal, measured for three different laser intensities, as a function of a_1 at $a_3 = 0$ and the linear chirp rate $a_2 = 0$. Also the fluorescence is measured as a function of a_3 at $a_1 = 0$. As shown in Fig. 5.11(b), the line shape is symmetric at a low intensity (the lowest red line) but becomes gradually asymmetric at higher intensities (the black (middle) and blue (upper) lines). As the peak intensity increases, the overlap between the shifted energy level and the laser spectral distribution gradually decreases. As a result, the two-photon excitation(TPE) in Cesium at zero-frequency offset is better achieved by a negative cubic phase term. This seems counterintuitive because the curvature of the laser spectral distribution is opposite that of the shifted energy level. However, as illustrated in the top panel of Fig. 5.11(b), the pulse at E' with



Figure 5.9: (a) Strong-field two-photon excitation probability, $P_e(\eta, \xi)$, calculated as a function of dimensionless variables: the frequency detuning η and the spectral curvature ξ . (b) The two-photon spectrogram (blue) of the unshaped pulse at the point $O(-\delta_c \tau + \Delta \tau, \delta_c \tau/3)$ where Δ and δ_c represent the static and dynamic level shifts, respectively. The dynamic level shift is drawn in black. (c) Control of frequency detuning along \overline{OA} . (d) The spectral curvature control along \overline{AB} . (e) The two-photon spectrogram of the optimally shaped pulse at the point $O_p(0,0)$.



Figure 5.10: Fluorescence 2D maps measured at laser peak intensities, $I_{\text{peak}}/I_0 = 0.06$, 0.14, 0.17 and 0.21, as a function of ' a_1 ' and ' $a_3\tau^2$ ' parameters. $I_0 = 1.0 \times 10^{11} W/cm^2$. Contour lines are calculated using Eq. (5.14).



Figure 5.11: Strong-field two-photon excitation of cesium studied at three different laser intensities, $I_{\text{peak}}/I_0 = 0.21$ (blue), 0.14 (black), 0.10 (red). The theoretical lines from Eq. (5.14) are compared with $7P_{1/2}-6S_{1/2}$ fluorescence signal measured (a) as a function of frequency offset 'a₁' at zero frequency curvature $a_3 = 0$; and (b) as a function of frequency curvature 'a₃' at zero frequency offset $a_1=0$. $I_0 = 1.0 \times 10^{11} W/cm^2$.

a negative cubic phase makes a better overlap with the detuned energy level than the pulse at E with a positive cubic phase. Therefore, the TPE rate in Cesium at zero-frequency offset is higher with a negative quadratic frequency chirp (cubic phase). The numerical calculation in Fig. 5.12 also shows a good agreement.

Figure 5.13 (a) shows the fluorescence as a function of detuning a_1 at various fixed curvature a_3 . At $a_3 = 0$, the maximum fluorescence is obtained at the negative a_1 because δ_c is negative in Cesium. Note that the optimal point for $I = 1.7 \times 10^{10}$ W/cm² is located at $a_1 = -4.28$ Trad/s and $a_3\tau^2 = 1.43$ Trad/s. The curvature control experiments shown in Fig. 5.13(b) are along the horizontal lines in Fig. 5.9(a). The measured signals are of more complex line shapes: Near the optimal detuning at $a_1 = 0$ [black line (with open squares)], as the curvature a_3 increases, the signal gradually grows and rapidly increases near $a_3 = 0$ (near O_p). For a more (less) detuned case with the positive (negative) a1 in the blue (with open circles) [red (with open triangles)] line, the signal rapidly decreases (increases) near $a_3 = 0$. Finally, from Eq. (5.14), the intensity invariant forms of excitation probability can be calculated as a function of each



Figure 5.12: Time evolution of cubic phases at fixed $a_1 = 0$; (a) $a_3\tau^2 = 10$ Trad/s; (b) $a_3\tau^2 = -10$ Trad/s; the pulse peak intensity is fixed at $0.5 \times 10^{11} W/cm^2$; insets represent corresponding control scheme.



Figure 5.13: Pulse-shape dependence of two-photon excitation in cesium: The excitation is measured (a) as a function of frequency curvature ' a_3 ' at fixed frequency offsets ' a_1 ' = 0 (black), 10 (blue), and -10 Trad/s (red), respectively; and (b) as a function of frequency offset ' a_1 ' at fixed frequency curvatures ' a_3 ' = 0 (black), 6 (blue), and -6 Trad/s (red), respectively. The peak intensity of the laser pulse is maintained at $I_{\text{peak}} = 1.7 \times 10^{10} \text{ W/cm}^2$.



Figure 5.14: Scaled strong-field two-photon absorption profile $P^{2\omega}/\Omega_c^2 \tau^2$ in Eq.(5.14) is plotted as a function of (a) $\xi = (\delta_c/3 + 2a_2\tau^2)\tau$ and (b) $\eta = (-\delta_c + 2a_1)\tau$.

single parameter η and ξ ,

$$P_e(\eta, 0) / (\Omega_c \tau/2)^2 = \sqrt{\pi} e^{-\eta^2/2}$$

$$P_e(0, \xi) / (\Omega_c \tau/2)^2 = \sum_{k=0}^{\infty} (-1)^k \eta^{2k} \frac{\Gamma(3k+1/2)}{(2k)!}$$
(5.16)

They are drawn in Fig. 5.14 overlaid with the measured data points from Fig. 5.11. We note that the overall probabilities of strong-field two-photon transition $P_e/I^2/\tau^2$ follow the theoretically obtained intensity invariant forms from Eq. (5.14). But this invariant form breaks in non-perturbative regime due to higher oder transition which makes the higher order dependence I^n in the excited population.

5.4 Further optimization of the phase-matching condition

One can further improve the approximate condition Eq. (5.15) by determining conditions that allow one to recover the population transfer that would be obtained without Stark shifts. To that end, we determine the population transfer to the excited state at the end of the process, from the numerical integration of the Schrödinger equation, for various (strong or not) peak-field amplitudes and Stark shifts by using a phase of the form $\phi(t) = a_1t + a_3t^3$. Here, we do not consider the spatial averaging. We first make the analysis by using the two-state model. Figure 5.15 shows two typical contour plots of the deviation from the population transfer to the excited state achieved without Stark shifts, with a pulse area of (a) π and (b) $\pi/2$, which correspond to a population transfer without Stark shifts of 1 and 0.5, respectively. We obtain (by taking $\Delta = 0$) the approximate optimal function that allows one to accurately recover the population transfer without Stark shifts:

$$2\phi(t) = \delta_c \left(0.89t - 0.12 \frac{t^3}{\tau^2} \right)$$
(5.17)

This has been obtained for a field intensity not larger than the one that leads to a complete population transfer in the absence of Stark shifts.



Figure 5.15: Contour plot (in the logarithmic scale to base 10) at the end of the pulse of the deviation from the population transfer in the absence of Stark shifts as a function of the dimensionless quantities $2a_1/\delta_c$ and $2a_3\tau^2/\delta_c$ for $\Delta = 0$, (a) $\tau\Omega_c = \sqrt{\pi}$ and (b) $\tau\Omega_c = \sqrt{\pi}/2$. They correspond to complete and half population transfers, respectively, in the absence of Stark shifts.

The demonstration of the exact optimal values of the linear and cubic terms in Eq. (5.17) is found to be beyond the scope of the accuracy of the present experiments. However, it is remarkable that this optimized function is a simple linear function of the peak Stark shift and, thus, of the peak-field amplitude, as anticipated in the preceding analysis. The value obtained for the linear term is close to the one determined with the truncated expansion Eq. (5.15). We have checked that the perturbation theory Eq. (5.13) gives a good approximation for the population transfer until the transfer of approximately 0.15 (error of 5%), which corresponds to a pulse area approximately of 0.25π (consistent with the estimated error of the perturbative expansion). Despite this limitation, we have obtained the interesting result that the line shapes can be approximately well described, up to a scaling factor (which depends on the intensity and the Stark shifts and that has to be determined with the numerical simulation), by the perturbation theory, even for stronger-field intensities.

5.5 Beyond two-level approximation

In previous section, we considered the two-photon transition in Cesium between the state $6S_{1/2}$ and $8S_{1/2}$. In this section, by introducing a four-level model for the strong-field transition between 6s-8s states, we verify the validity of the two-level approximation.

The mean frequency ν of the laser (corresponding to the laser frequency of the Fourier transform pulse, i.e. before its shaping) is exactly two-photon resonant: $\nu = \omega_{eg}/2$. The level scheme of Cesium is shown in Fig. 5.1. A single photon allows the ionization of the atom from the 8s excited state but the ionization rate Γ_e is smaller than the two-photon Rabi frequency Ω , $|\Gamma_e/\Omega| = 6.44 \times 10^{-2}$ [28]. It means that only a negligible portion of the excited population is depleted by ionization. Because two intermediate states $6P_{1/2}$ (state g1) and $6P_{3/2}$ (state g2) are close to a single photon resonance, we consider a four-level approximation. The one-photon detunings are $\Delta_{g1}^0 = -4.47 \times 10^{-3}a.u$. and $\Delta_{g2}^0 = -1.94 \times 10^{-3}a.u$. At the pulse intensity of $0.1 \times 10^{11} W/cm^2$, we get for the single-photon Rabi frequencies $\Omega_{g1} = 2.4 \times 10^{-3}a.u$. $< |\Delta_{g1}|$ and $\Omega_{g2} = 3.38 \times 10^{-3}a.u$. $\sim |\Delta_{g2}|$. Because orders of the onephoton Rabi frequencies are same to those of the detuning, the intermediate states can be populated. Thus the two-level approximation is insufficient to describe Cesium 6s-8s transitions. The four-level Hamiltonian can be expressed as:

$$\mathbf{H}_{4}(\mathbf{t}) = (5.18) \\
\begin{pmatrix}
S_{g}(t) & \Omega_{g1}(t)/2 & \Omega_{g2}(t)/2 & \Omega_{ge}(t)/2 \\
\Omega_{g1}(t)/2 & \Delta_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) \\
\Omega_{g1}(t)/2 & \Omega_{g1} - \dot{\phi}(t) + S_{g1}(t) & 0 & \Omega_{e1}/2 \\
\Omega_{g1}(t)/2 & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) \\
\Omega_{g1}(t)/2 & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) \\
\Omega_{g1}(t)/2 & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) & \Omega_{g1}(t) \\
\Omega_{g1}(t) & \Omega_{$$

$$\begin{pmatrix} \Omega_{g2}(t)/2 & 0 & \Delta_{g2} - \dot{\phi}(t) + S_{g2}(t) & \Omega_{e2}/2 \\ \Omega_{ge}(t) & \Omega_{e1}(t)/2 & \Omega_{e2}(t)/2 & -2\dot{\phi}(t) + S_{e}(t) - i\Gamma_{e}(t)/2 \end{pmatrix}$$

However, we check numerically that two-level approximation is still a good approximation for peak intensities up to $I_0 = 0.1 \times 10^{11} \text{W/cm}^2$, due to the additional Stark shift S_2 that allows state g2 to be sufficiently shifted from the resonance. The optimal condition in four-state approximation can be obtained by using numerical calculation at $I_0 = 0.47 \times 10^{11} \text{W/cm}^2$,

$$2\phi(t) = \delta_c \left(0.75t - \frac{0.10}{\tau^2} t^3 \right).$$
(5.19)

We get a population transfer of 99 %, and the rest is ionized. We obtain numerically that the coefficients after the spatial averaging saturates to $a_1 \simeq -4.5$ Trad/s and $a_3\tau^2 \simeq 0.5$ Trad/s for fields intensities beyond $0.2 \times 10^{11} W/cm^2$ in consistency with the experimental results.

Chapter 6. Coherent quantum control of multi-photon absorptions in Sodium

Energy ladder climbing systems are one of popular examples which are investigated by linear chirp pulses [48, 49, 50]. In this chapter, we describe coherent control of 2+1 multi-photon absorption in a Sodium 3s-4s-7p ladder system. '2+1' means that the system consists of a two-photon transition and a one-photon transition [51]. We measure shaped-pulse multi-photon absorption in Sodium 3s-4s-7p ladder system, and compare the result with the calculation. when a linearly chirped pulse is used.

The vapor pressure of the Sodium cell in solid phase is given by [45]

$$\log_{10} P_v = 2.881 + 5.298 - \frac{5603}{T},\tag{6.1}$$

where the temperature T is in K and the vapor pressure P_v in Torr. The melting point of Sodium is 371 K. At the room temperature 293 K, the vapor pressure is 1.14×10^{-11} Torr. The corresponding atom density is calculated as $3.76 \times 10^{11}/m^3$. The density of Sodium is much lower than that of Cesium at the room temperature. Thus the fluorescence signal of Sodium is about 10^5 times smaller than that of Cesium. In the experiment, the temperature of the vapor cell is maintained at 423 K, and the atom density is $1.92 \times 10^{17}/m^3$. The atomic number of Sodium is 11, and the ground state is 3s state. After absorbing two photons of 777 nm wavelength, Sodium atoms are excited to the 4s state. The lifetime of 4s state is 38 ns. The specific energy level diagram is given in Fig. 6.1. In Sodium, an ultrafast pulse of center-wavelength 777 nm induces the 3s-4s non-resonant TPA process and, simultaneously, the 4s-7p one-photon transition as well. The excited population of 7p state decays radiatively to the 3p state through the 6d or 7s state. The resonant energy of 7s-3p (6d-3p) transition is 474.8 nm (466.5 nm) in wavelength scale. The excited 4s-state atoms decay to the 3s state through the 3p state, generating 590 nm photons. The fluorescence is measured through either 476.5 nm or 590 nm bandpass filters, to estimate the 7p and 4s excited populations, respectively. The dynamic Stark shift of 3s state is determined by couplings with p states. The 3s state is shifted by -32.8 Trad/s at 1.0×10^{11} W/cm², and the shift of 4s state is 28.9 Trad/s at 1.0×10^{11} W/cm². Thus the total shift of the two-photon transition is positive, 61.7 Trad/s. The 7p state is also shifted by 8.7 Trad/s due to the Ponderomotive shift. The two-photon Rabi coupling is -25.0 Trad/s and the Rabi coupling between 4s and 7p states is 5.5 Trad/s. Because the Rabi frequency of TPA is larger than that of the 4s-7p transition, the atoms are mainly excited to the 4s state.

6.1 Control of Sodium $|7p\rangle$ -state excitation

Figure 6.2 shows the dressed-state picture of the given 2+1 photon absorption processes in a sodium three-state model system. First, we consider the transition paths from $|3s\rangle$ to $|f\rangle = |7p\rangle$ state. There are three possible paths indicated with in Fig. 6.2(a). For a positively-chirped pulse, the photon frequency increases as a function of time, and, therefore, the $|3s\rangle - |7p\rangle$ transition is possible along the path (III), which is a direct transition path from $|3s\rangle$ to $|7p\rangle$, not passing by the $|4s\rangle$ state. On the other hand, for a negatively-chirped pulse, the photon frequency decreases, and two paths are available: a sequential path



Figure 6.1: Energy levels and transitions of atomic sodium. [46]

transition	dipole moment	energy	DSS
_	$(\times 10^{-29} \text{Cm})$	$(\times 10^{15} \text{ rad/s})$	$(\mathrm{Trad/s})$
$3s_{1/2} - 3p_{1/2}$	2.11	3.20	-11.15
$3s_{1/2} - 3p_{3/2}$	2.99	3.20	-22.21
$3s_{1/2} - 4p_{1/2}$	0.19	5.71	-0.03
$3s_{1/2} - 4p_{3/2}$	0.27	5.71	-0.05
$3s_{1/2} - 5p_{1/2}$	0.07	4.85	0.00
$3s_{1/2} - 5p_{3/2}$	0.09	4.86	-0.01
$4s_{1/2} - 3p_{1/2}$	2.15	-1.66	-8.21
$4s_{1/2} - 3p_{3/2}$	2.15	-1.65	-8.22
$4s_{1/2} - 4p_{1/2}$	5.03	0.85	14.18
$4s_{1/2} - 4p_{3/2}$	7.11	0.85	28.39
$4s_{1/2} - 5p_{1/2}$	0.56	1.75	0.67
$4s_{1/2} - 5p_{3/2}$	0.80	1.75	1.36
$4s_{1/2} - 6p_{1/2}$	0.23	2.17	0.33
$4s_{1/2} - 6p_{3/2}$	0.32	2.17	0.67
$4s_{1/2} - 7p_{1/2}$	0.13	2.41	-
$4s_{1/2} - 7p_{3/2}$	0.18	2.41	-
$4s_{1/2} - 8p_{1/2}$	0.09	2.56	-0.10
$4s_{1/2} - 8p_{3/2}$	0.12	2.56	-0.21

Table 6.1: Dipole moments and transition energies of atomic Sodium. The dynamic Stark shifts (DSS) are calculated for the laser intensity at $1 \times 10^{15} W/m^2$. The line strengths are from NIST atomic database [46].



Figure 6.2: (a) The dressed-state picture for a 2+1 photon absorption processes in a sodium three-state model system. Three arrows indicate possible transition paths from $|g\rangle$ to $|r\rangle$, when a negatively-chirped pulse interacts. (b) Schematic illustrations of dynamically-Stark-shifted resonant frequencies of $|3s\rangle - |4s\rangle$ and $|4s\rangle - |7p\rangle$ transitions (dashed lines) of sodium atoms, overlaid with two-photon (blue) and one-photon (red) time-frequency spectrograms for the negatively-chirp pulse of $a_2 = -5000$ fs². (c) The similar illustration for a positively-chirped pulse with $a_2 = 5000$ fs². The spectrograms are vertically shifted to match the corresponding transitions.

(I) and a direct path (II). It is expected, considering the fact that the path (III) is an inefficient excitation, that the $|7p\rangle$ atoms are better created by a negatively-chirped pulse than by a positively-chirped one.

This asymmetric excitation to $|7p\rangle$ state, obtained as a function of the chirp parameter, can be schematically understood in time-frequency picture as well. Figures 6.2(b) and (c) show one-photon (red) and two-photon (blue) spectrograms of chirped laser pulses plotted in a two-dimensional space of time and frequency. They are overlaid with the resonant frequency shifts of $|4s\rangle - |7p\rangle$ and $|3s\rangle - |4s\rangle$ transitions (dashed lines). In Fig. 6.2(b), the $|3s\rangle - |4s\rangle$ transition occurs first and the $|4s\rangle - |7p\rangle$ later, so a sequential excitation along the $|3s\rangle \rightarrow |4s\rangle \rightarrow |7p\rangle$ path is satisfied. On the other hand, for a positivelychirped pulse case shown in Fig. 6.2(c), because the $|3s\rangle - |4s\rangle$ resonance frequency is up-shifted during the optical interaction, the two-photon spectrum overlaps with the resonance line after the temporal center of the pulse, and as a result, the $|3s\rangle - |4s\rangle$ and $|4s\rangle - |7p\rangle$ transitions occur in time-reversal sequence, causing the sequential excitation not possible. Therefore, sequential excitation is not possible for a positively-chirped pulse, and only non-sequential excitations, e.g. along path (III) in Fig. 6.2(a), are possible.

In the following section, we use the theoretical model to calculate the sequential and non-sequential excitations to $|7p\rangle$ state, and verify the results with corresponding experiments.

6.2 Calculation of the sequential and direct $|7p\rangle$ excitations

The probability amplitude of the $|7p\rangle$ state is obtained in Eq. (3.55). In order to calculate the transition probability amplitudes for the sequential and direct excitation paths, we separate Eq. (3.55) into two parts: the resonant and non-resonant parts. In the perturbative interaction regime, the probability amplitudes corresponding to these two parts can be written, respectively, as [47]:

$$a_{\rm f,res} = i\pi E(\omega_{re} + \delta_{fe}) \int_{-\infty}^{\infty} d\omega E(\omega) E(\omega_{eg} + \delta_{eg} - \omega), \qquad (6.2)$$

$$a_{f,\text{nonres}} = \wp \int_{-\infty}^{\infty} d\omega \frac{E(\omega)}{\omega - \omega_{fe} - \delta_{fe}} \int_{-\infty}^{\infty} d\omega' E(\omega') E(\omega_{fg} + \delta_{fg} - \omega - \omega'), \tag{6.3}$$

where \wp is the Cauchy principal value. The subscripts 'res' and 'nonres' denote the resonant and nonresonant excitations, and δ_{eg} and $(\delta_{fe}$ are the maximum amplitudes of the total level shifts of the $|3s\rangle - |4s\rangle$ and $|7p\rangle - |4s\rangle$ transitions, respectively, which are calculated at the temporal peak of a transform-limited pulse. $E(\omega)$ is the Fourier transform of the electric field profile, a spectrally chirped gaussian pulse, given by

$$E(\omega) = \sqrt{\pi} E_o T_o \exp\left(-\frac{1}{4}(\omega - \nu)^2 T_o^2 + ia_2(\omega - \nu)^2\right),$$
(6.4)

where T_o is the gaussian width of a transform-limited pulse and a_2 is the linear chirp parameter in the spectral domain.

The resonant $|3s\rangle \rightarrow |7p\rangle$ excitation is obtained by substituting Eq. (6.4) into Eq. (6.2) as

$$a_{\rm f,res} = \sqrt{\frac{2\pi^3}{T_o T_p}} E^3(\nu) \exp\left[-\left(\frac{\delta_{\rm 1ph}^2(\nu)}{4} + \frac{\delta_{\rm 2ph}^2(\nu)}{8}\right) (T_o^2 + 2ia_2)\right] e^{i(\theta + \pi)/2},\tag{6.5}$$

where $\tan \theta = -2a_2/T_o^2$, the two-photon detuning is defined as $\delta_{2ph}(\nu) = \omega_{eg} + \delta_{eg} - 2\nu$, and the one-photon detuning $\delta_{1ph}(\nu) = \omega_{fe} + \delta_{fe} - \nu$. Alternatively Eq. (6.5) can be written as

$$a_{\rm f,res} = \sqrt{\frac{2\pi^3}{T_o T_p}} E^3(\nu) \exp\left[-\left(\frac{\delta_{\rm 3ph}^2(\nu)}{12} + \frac{3\delta_s^2}{8}\right) (T_o^2 + 2ia_2)\right] e^{i(\theta + \pi)/2},\tag{6.6}$$

where three-photon detuning is defined as $\delta_{3ph}(\nu) = \omega_{fg} + \delta_{fg} - 3\nu$, and δ_s is the structure factor, $\delta_s = \omega_{fe} + \delta_{fe} - (\omega_{fg} + \delta_{fg})/3$ that is ν independent.

The transition probability for the resonant excitation to $|7p\rangle$ is the absolute square of Eq. (6.6). The result is a symmetric function of chirp a_2 , and does not explain the prediction in Sec. 6.1 saying that the sequential excitation should be an asymmetric function of the chirp. However, there is indeed another sequential path in the non-resonant excitation path in Eq. (6.3). While the resonant excitation $(a_{\rm f,res})$ contributes only to the sequential transition path $(|3s\rangle \rightarrow |4s\rangle \rightarrow |7p\rangle)$, the non-resonant part $(a_{\rm f,nonres})$ contributes to both the sequential and direct paths, i. e.,

$$a_{\rm f} = a_{\rm f,res}^{\rm seq} + a_{\rm f,nonres}^{\rm seq} + a_{\rm f,nonres}^{\rm direct}, \tag{6.7}$$

where $a_{\rm f,res}^{\rm seq} = a_{\rm f,res}$ because there is no resonant non-sequential transitions.

For the calculation of the non-resonant direct excitation $a_{r,nonres}^{direct}$, we consider Eq. (6.3) near the three-photon resonant condition, i.e., $\omega \approx (\omega_{fg} + \delta_{fg})/3$. Then, the denominator of the integrand in Eq. (6.3) can be treated as a constant, and it is simple to show that

$$a_{\rm f,nonres}^{\rm direct} = -\frac{4\pi}{\sqrt{3}\delta_s T_o T_p} E^3(\nu) \exp\left[-\frac{\delta_{\rm 3ph}^2}{12}(T_o^2 + 2ia_2)\right] e^{i\theta}.$$
 (6.8)

Lastly, for the calculation of $a_{r,nonres}^{seq}$, we consider the small frequency range around the pole, i.e. $\omega \approx \omega_{fe} + \delta_{fe}$, in Eq. (6.3). Equation (6.3) is written as

$$a_{\rm f,nonres}^{\rm seq} = \sqrt{\frac{2\pi}{T_o T_p}} E^3(\nu) \exp\left[-\frac{\delta_{3ph}^2}{12}(T_o^2 + 2ia_2)\right] e^{i\theta/2} \\ \times \wp \int_{-\infty}^{\infty} \frac{d\omega}{\omega - \omega_{fe} - \delta_{fe}} \exp\left[-\frac{3}{8}\left(\omega - \frac{\omega_{fg} + \delta_{rg}}{3}\right)^2(T_o^2 + 2ia_2)\right], \quad (6.9)$$
and, by neglecting the term of the order of $O(\omega - \omega_{fe} + \delta_{fe})^2$, the non-resonant sequential excitation is given by

$$a_{\rm f,nonres}^{\rm seq} = \sqrt{\frac{2\pi^3}{T_o T_p}} E^3(\nu) \exp\left[-\left(\frac{\delta_{\rm 3ph}^2(\nu)}{12} + \frac{3\delta_s^2}{8}\right) (T_o^2 + 2ia_2)\right] e^{i(\theta + \pi)/2} \mathrm{sgn}(a_2\delta_s) = a_{\rm f,res}^{\rm seq} \mathrm{sgn}(a_2\delta_s),$$
(6.10)

where the sign function, defined by $\operatorname{sgn}(x) = +1$ (-1) for x > 0 (x < 0), is due to the contour integral given as a function of the sign of $\delta_s a_2$. The result is valid in the range of chirp in $|a_2|/T_o^2 \gg 1$. In the experiment, T_o is 37 fs and the approximation $|a_2|/T_o^2 \gg 1$ is valid for $|a_2| > 1369$ fs². In Sodium, $\delta_s =$ -8.3 Trad/s, and, as a result, $a_{f,nonres}^{seq} + a_{f,res}^{seq} = 0$ for $a_2 > 0$. Therefore, the net sequential excitation path to $|7p\rangle$, in Eq. (6.7), vanishes for a positively-chirped pulse by canceling the resonant and non-resonant contributions with each other.

6.3 Verification of shaped-pulse $|7p\rangle$ -state excitation

The sodium $|7p\rangle$ excitation was experimentally tested as a function of the chirp rate a_2 . Figure 6.3 shows the calculation of the net transition probability (in solid line) of $|3s\rangle - |7p\rangle$ given in Eq. (6.7), the components of which are obtained in Eq. (6.6), Eq. (6.8), and Eq. (6.10), as a function of linear chirp rate. The laser (transform-limit) peak intensity was kept at $I = 3.0 \times 10^{11} \text{ W/cm}^2$, and the chirp rate a_2 was varied in the range $[-1.0, 1.0] \times 10^4$ fs². As predicted in the schematic picture in Sec. 6.1, the excitation is significantly enhanced by negatively chirped pulses, and the reason is because the sequential excitation path along $|3s\rangle - |4s\rangle - |7p\rangle$ is zero for positively chirped pulses. The direct transition from $|3s\rangle - |7p\rangle$ which is a symmetric function found in Eq. (6.8) is about 10 times smaller than the sequential one for the tested laser peak intensity. For comparison, the sequential and direct excitation probabilities are plotted in dashed and dot-dash lines, respectively. For the numerical calculation, the dynamic Stark shift of $|3s\rangle$ state is $S_g = -32.8(I/I_o)$ Trad/s, determined by couplings with $|p\rangle$ states, where I_o is the reference laser intensity, $I_o = 1.0 \times 10^{11} \text{ W/cm}^2$. The shift of $|4s\rangle$ state is $S_e = 28.9(I/I_o)$ Trad/s at the same intensity. Thus, the net frequency shift of the two-photon transition is positive. The $|7p\rangle$ state is shifted by couplings with not only $|s\rangle$ and $|d\rangle$ states but also continuum states. It is known that the presence of continuum increases the energy of the excited state by the ponderomotive energy given by $S_r(t) = e^2 E^2(t)/4m\nu^2$, where ν depicts the laser frequency and m is electron mass. As a result, the $|7p\rangle$ state is up-shifted $S_r = 8.7(I/I_o)$ Trad/s, so the net frequency shift of $|4s\rangle - |7p\rangle$ transition is negative $S_{re} = -20.2(I/I_o)$ Trad/s. Also, because the laser beam has a spatial intensity distribution of a gaussian shape, the total excitation probability was calculated as the sum of local excitations, i.e., $P_{\text{total}} = \int_0^\infty P(I(r)) d^3 r.$

The excitation probability calculated as a function of a_2 and laser peak intensity I is compared with the experimentally tested results in Fig. 6.4. The $|7p\rangle$ excited atoms are measured, by monitoring the $|7s\rangle - |3p\rangle$ fluorescence, as a function of laser (transform-limited) peak intensity and chirp parameter (a_2) . The laser peak intensity I was changed from zero to $I = 6.0 \times 10^{11} \text{ W/cm}^2$, and a_2 was varied in the range of $[-1.0, 1.0] \times 10^4 \text{ fs}^2$. The net transition probability which is caused as the sum of sequential and direct transitions calculated in Fig. 6.4(a) shows an excellent agreement with the experimental results in Fig. 6.4(b).



Figure 6.3: Three-photon sodium excitation to $|7p\rangle$ state with a chirped optical pulse. The curves show the calculation of total (solid line), sequential (blue dashed line), and direct (green dot-dash line) transitions. The experimental measurements are shown in circles. The inset shows the sequential and direct transition paths from $|3s\rangle$ to $|7p\rangle$.



Figure 6.4: (a) Theoretical and (b) experimental results of chirped-pulse three-photon excitation probability of sodium atoms as a function of linear chirp rate and laser peak intensity. For the relative measurement of $|7p\rangle$ sodium atoms, the $|7s\rangle - |3p\rangle$ fluorescence signal was recoded.

6.4 Chirped-pulse excitation of $|4s\rangle$ -state atoms

In this section, we consider the excitation of $|4s\rangle$ atoms. It is clear from Eq. (3.61) that the dominant excitation to $|4s\rangle$ -state is the direct two-photon absorption from $|3s\rangle$ -state. The presence of the $|7p\rangle$ state affects this excitation in terms of the third order Dyson series, and the new excitation path is a four-photon sequential excitation along $|3s\rangle \rightarrow |4s\rangle \rightarrow |7p\rangle \rightarrow |4s\rangle$. Therefore the excitation probability amplitude of $|4s\rangle$ atoms is given by

$$A_e = A_e^{\text{direct}} + A_e^{\text{seq}},\tag{6.11}$$

where the direct two-photon transition $A_e^{\rm direct}$ is

$$A_e^{\text{direct}} = -i \int_{-\infty}^{\infty} dt \frac{\Omega(t)}{2} e^{-iQ_1(t)}, \qquad (6.12)$$

and the sequential two-photon transition a_e^{seq}

$$A_{e}^{\text{seq}} = i \int_{-\infty}^{\infty} dt \frac{\Omega_{er}(t)}{2} e^{-iQ_{2}(t)} \int_{-\infty}^{t} dt' \frac{\Omega_{er}(t')}{2} e^{iQ_{2}(t')} \int_{-\infty}^{t''} dt'' \frac{\Omega(t'')}{2} e^{-iQ_{1}(t'')}.$$
(6.13)

Figure 6.5 shows the calculation and experiment of the excitation probability of $|4s\rangle$ -state atoms, obtained as a function of linear chirp rate. We apply the Fourier transform method ,which is used to obtain the 7p probability, to A_e^{direct} . The probability amplitude of the excitation along the direct transition path is

$$a_e^{direct} = \sqrt{\frac{2\pi}{T_o T_p}} E^2(\nu) \exp\left[-\frac{\delta_{2ph}^2(\nu)}{8} (T_o^2 + 2ia_2)\right] e^{i(\theta + \pi)/2},\tag{6.14}$$

which is the dominant contribution to the net excitation, and the sequential transition via 7p state is negligible. We note that the factor $\sum_{j} \mu_{ej} \mu_{jg} / (\hbar^2(\omega_{jg} - \nu))$ is omitted. Therefore, the excitation probability is given by

$$P_e \simeq |a_e^{direct}|^2 \propto \frac{1}{\sqrt{1+B^2}} \exp\left[-\frac{A^2}{2(1+B^2)}\right],$$
 (6.15)

where $A = \delta_{2ph}(\nu)\tau$ and $B = 2a_2/\tau_0^2$. The net excitation probability shows a symmetric function of linear chirp rate, and nearly vanishes at zero chirp. The dynamic Stark shift of $|3s\rangle - |4s\rangle$ transition causes the off-resonance condition to the two-photon excitation. As the dynamic Stark shift, which is stored in the parameter A, increases, the term exp $\left(-A^2/(2(1+B^2))\right)$ becomes important, and the net probability makes a local minima at zero chirp rate.

The $|4s\rangle$ excited atoms are measured, by monitoring the $|3p\rangle - |3s\rangle$ fluorescence, as a function of laser (transform-limited) peak intensity and chirp parameter (a_2) . The transition probability calculated in Fig. 6.6(a) are compared with the experimental results in Fig. 6.6(b), showing an excellent agreement.



Figure 6.5: Sodium excitation to $|4s\rangle$ state with a chirped optical pulse. The curves show the calculation of total (solid line), sequential (blue dashed line), and direct (green dot-dash line) transitions. The curve of the sequential transition is magnified by 1000 times. The experimental measurements are shown in circles. The sequential and direct transition paths from $|3s\rangle$ to $|4s\rangle$ are shown in the inset.



Figure 6.6: (a) Theoretical and (b) experimental results of chirped-pulse two-photon excitation probability of sodium atoms as a function of linear chirp rate and laser peak intensity. For the relative measurement of $|4s\rangle$ sodium atoms, the $|3p\rangle - |3s\rangle$ fluorescence signal was recoded.

Chapter 7. Conclusion

In modern laser optics, light controlled in its spectral and temporal shape is used not only as a tool for observation but as a tool for controlling quantum objects and light-induced phenomena. In recent decades, the concept of coherent control has been combined with ultrafast lasers. Coherent control with ultrafast lights has been applied to selective chemical processes, multi-photon microscopy, spectroscopy, and the optimization of nonlinear light-matter interaction. In particular, ultrafast coherent control of multi-photon absorption has been widely studied in conjunction with quantum interference engineering.

In this thesis, we have devised a new coherent control method for a multi-photon excitation of Alkali atoms in the strong-field interaction regime. The energy levels change in the strong-field regime and, for example, the dynamic Stark energy level shift prevents the enhancement of multi-photon absorptions by a transform-limited pulse. In the newly developed analytical coherent control, laser pulses are shaped as a polynomial sum of frequency and/or time in an acousto-optic programmable dispersive filter. For the given light-matter interactions, the model Hamiltonians developed in the strong-field regime are analytically studied and the derived excitation probability formulas given as analytical solutions are compared with the experimental results.

We have demonstrated in experiments with atomic Cesium the analytical coherent control of the twophoton absorption in a dynamically shifted energy level structure. By solving the two-photon two-level model Hamiltonian, the transition probability of the two-photon broadband excitation has been obtained as a function of pulse shape parameters, frequency detuning, and laser intensity. We have found that transform-limited pulses are not optimal in the strong-field regime, and certain linearly chirped pulses with a lower peak intensity enhance the given non-linear process. Experiments carried out in the strongfield regime of two-photon absorption reveal that the analytically obtained offset and curvature of a laser spectrum compensate the effect of both static and dynamic energy shifts of the given light-atom interaction.

In addition, we have applied the analytical coherent control method to the 2+1 multi-photon energy ladder system of atomic Sodium. In experiments with atomic Sodium, we have coherently controlled the interference among sequential and non-sequential excitation paths from the ground 3s state to the 7pstate in the strong-field regime. Both analytic formula and experimental results show that a negatively chirped pulse enhances the 7p population because the sequential path is opened by a negatively chirped pulse.

Our final goal is the demonstration of quantum logic gates and quantum algorithm by using coherent quantum control in a classical fields - atom system. Electronic states are used as qubits and controlled by coherent quantum control method. We expect that the suitable pulse shape enables operations of certain quantum algorithm and quantum gate. For example, Glover's search algorithm was demonstrated in the Rydberg atom by impulse terahertz pulses [52]. Also the idea that the sequence of chirped pulses perform a series of quantum logic gates was proposed [53]. To demonstrate quantum logic and quantum algorithms in real atomic systems, there are various obstacles. One of the obstacles is unwanted phase due to level shifts and power broadening. For a more accurate pulse design, the level shifts have to be considered. We experimentally demonstrated coherent control method to maintain the resonance condition by chirped pulses in the presence of level shifts. We expect that our studies give the possibility to obtain pulse shapes for more reliable operation of quantum logic gates.

Chapter 8. Appendix A: adiabatic passage

Generally, the coherent control involves the requirement of the complex pulse shape. The adiabatic passages use the simple pulse shape such as a linearly chirped pulse. The popular example of the adiabatic passage, STIRAP(Stimulated Raman Adiabatic Passage) needs the coherent two laser beams that delay between two pulses are controlled. The controlled coherent two laser beams creates the coherence between states and the STIRAP makes it possible to transfer the population utilizing the coherence between states. In this aspect, we contain the adiabatic passages to coherent control. Typically, the adiabatic passages has been represented using the dressed state picture.

8.1 Introduction of adiabatic passage

The first laser-induced adiabatic rapid passage was demonstrated in 1974 with a monochromatic microsecond laser and a sweeped dc Stark field [54]. The adiabatic passage enables to transfer the population to the desired state completely. It is based on keeping the time-dependent state at the instant eigenstate of the system. Assume that the time-dependent Hamiltonian is written as:

$$H = \frac{\hbar}{2} \begin{bmatrix} -\Delta(t) & \Omega(t) \\ \Omega(t) & \Delta(t) \end{bmatrix}$$
(8.1)

where the Rabi frequency $\Omega(t)$ is given as $\Omega(t) = -\vec{\mu}_{eg} \cdot \vec{E}(t)$ and the detuning $\Delta(t)$ is the difference between the transition frequency ω_{tr} and the laser frequency ω_0 , $\Delta = \omega_{tr} - \omega_0$. The eigenstates of the system is represented as:

$$|+\rangle = \cos\theta |1\rangle + \sin\theta |2\rangle \tag{8.2}$$

$$|-\rangle = -\sin\theta |1\rangle + \cos\theta |2\rangle \tag{8.3}$$

here the mixing angle θ depends on the Rabi frequency and the detuning:

$$\tan 2\theta = -\frac{\Omega(t)}{\Delta(t)} \tag{8.4}$$

We transform the Hamiltonian to the adiabatic one using the adiabatic basis:

$$H_{ad} = \frac{\hbar}{2} \begin{bmatrix} \lambda_+ & 2i\dot{\theta} \\ -2i\dot{\theta} & \lambda_- \end{bmatrix}$$
(8.5)

The eigenvalue of the system λ_{\pm} is written as $\lambda_{\pm} = \pm \sqrt{\Delta^2 + \Omega^2}$. To keep the adiabaticity, the offdiagonal terms have to be negligible relative to the diagonal terms. Thus the following adiabatic condition are induced as:

$$|\lambda_{+} - \lambda_{-}| >> 2 \left| \dot{\theta} \right| \tag{8.6}$$

After some calculation, we obtain the adiabatic condition as

$$\left|\dot{\Delta}\Omega - \dot{\Omega}\Delta\right| << 2(\Delta^2 + \Omega^2)^{3/2} \tag{8.7}$$

When the adiabatic condition is satisfied, the perfect population transfer is possible. Initially, before the electric field $(t \to -\infty)$, the Rabi frequency satisfies that $\Omega(t) \simeq 0$ and the detuning $\Delta(t)$ has the finite positive value. Thus the mixing angle goes to 0. After interaction with the electric field $(t \to \infty)$, the Rabi frequency goes to 0 again and the detuning has the negative value. Therefore the mixing angle becomes $\pi/2$. The system in the |1 > (or |2 >) state evolves into the |2 > (or |1 >) state. For this, a linearly chirped pulse is required and also the sufficient fluence of the laser required.

8.2 Stimulated Raman Adiabatic Passage

Many studies have demonstrated adiabatic passages. The one of typical examples is stimulated Raman adiabatic passage (STIRAP). This scheme was demonstrated in 1988 [55]. For STIRAP, two colored laser pulses are applied to the Λ -type three level system. There has been two types for realization of two pulses: spatially partially overlapped two CW laser beams and a laser pulse sequence [56]. If the adiabatic condition are satisfied, the complete population transfer are possible. The Hamiltonian for the simplest implementation of STIRAP, which describes the couplings of the three levels by two coherent electric field, is represented as

$$H_{ad} = \frac{\hbar}{2} \begin{bmatrix} 0 & \Omega_p & 0\\ \Omega_p & 2\Delta_p & \Omega_s\\ 0 & \Omega_s & 2(\Delta_p - \Delta_s) \end{bmatrix}$$
(8.8)

The Rabi frequencies Ω_p and Ω_s determine the coupling strength between the states. The detuning of pump beam relative to the the transition between $|1\rangle$ and $|2\rangle$ can be written $\hbar\Delta_p = (E_2 - E_1) - \hbar\omega_p$. The detuning of the Stokes beam Δ_s is given as $\hbar\Delta_p = (E_2 - E_3) - \hbar\omega_s$. The Hamiltonian describes the three level system that the $|1\rangle$ and $|2\rangle$ states are coupled with each other and the $|2\rangle$ and $|3\rangle$ states coupled but the $|1\rangle$ and $|3\rangle$ state are not. For simplicity, $\Delta_p = 0$ and $\Delta_s = 0$. Eigenstates of the system are linear combinations of diabatic state $|1\rangle$, $|2\rangle$, and $|3\rangle$:

$$|a^{+}\rangle = \sin\theta \sin\phi |1\rangle + \cos\phi |2\rangle + \cos\theta \sin\phi |3\rangle$$
(8.9)

$$|a^0\rangle = \cos\theta|1\rangle - \sin\theta|3\rangle \tag{8.10}$$

$$|a^-\rangle = \sin\theta \cos\phi |1\rangle - \sin\phi |2\rangle + \cos\theta \cos\phi |3\rangle$$
(8.11)

(8.12)

where the mixing angle θ is defined by

$$\tan\theta = \frac{\Omega_p}{\Omega_s} \tag{8.13}$$

Intuitively, it might be expected to require a sequence of the population transfers $|1\rangle \rightarrow |2\rangle$ and $|2\rangle \rightarrow |3\rangle$ to populate the $|3\rangle$ state. But there are loss of the population when the $|2\rangle$ state is lossy due to the radiative decay to other states. To prevent loss of the population, the coherence between the $|2\rangle$ and $|3\rangle$ states are generated by the Stoke pulse at first. After then, the pump pulse make the coupling between the $|1\rangle$ state and the prepared coherence. Because the $|2\rangle$ state is never populated, the population can be transferred from the $|1\rangle$ to $|3\rangle$ state perfectly.

Summary

Analytical coherent controls of Alkali atoms in strong field regime

재단된 빛을 이용하여 양자 동역학의 최종 상태를 우리가 원하는 방향으로 나아가게 할 수 있는데, 이러한 개념을 '결맞음 양자 제어'라고 부른다. Silberberg의 이광자 흡수 과정에 대한 결맞음 제어 연구 이래로 다광자 흡수 과정에 대한 많은 연구들이 복잡계에서 시도되었다. 그러나 앞선 연구들은 빛에 의하여 계의 에너지 레벨 구조가 변하지 않는, 약한 전기장 영역을 고려했다. 현재는 강한 전기장 영역에 초점을 맞춰 연구가 이뤄지고 있다. 이 영역에서는 강력장 효과가 발생하여, 강한 펨토초 레이저 장과 물질의 효과적인 상호작용을 방해하는데, 특히 동적 스타크 효과는 다광자 흡수 과정에 결정적인 영향을 끼친다. 우리는 펄스의 주파수 변화를 재단하여 동적 스타크 효과를 보상하는 새로운 해석적 결맞음 제어를 연구했다. 실험에서는 펄스 재단기의 하나인 AOPDF를 이용하여 전기장의 위상이 시 간 및 주파수의 다항식으로 표현되는 레이저 펄스를 재단하고, 이를 이용하여 알칼리 원자의 강력장 다광자 흡수 과정에 적용하였다. 먼저, 세슘 실험에서 우리는 동적 에너지 변화를 보이는 구조에서의 이광자 흡수 과정의 해석적 결맞음 제어를 구현하였다. 우리는 펄스 모양의 함수로 이광자 흡수 과정의 확률을 얻었고, 이를 통하여 초단광 펄스(transform-limited pulse)가 강력장 영역에서는 이광자 흡수의 최적화 펄스가 아님을 확인하였다. 덧붙여 펄스 폭이 길고, 에너지 세기가 낮은 선형 첩 펄스의 경우 이광자 흡수 과정을 더 강화시킬 수 있음을 실험, 이론적으로 보였다. 더 나아가, 레이저-원자 상호 작용동안 공명 조건을 유지시키는 삼차항 첩 펄스를 이용하여 강력장에서 세슘의 이광자 흡수 과정을 최적화 시킬 수 있음을 실험, 이론으로 확인하였다. 마지막으로 소듐의 2+1 다광자 흡수 과정에 해석적 결맞음 제어법을 적용시켰다. 우리는 선형 첩 펄스를 이용하여 소듐의 3s-7p 순차 및 비순차적 경로들의 간섭을 조절할 수 있었다. 특히 음의 선형 첩이 가해졌을 경우, 공명 성분에 의한 순차적 경로와 비공명 성분에 의한 순차적 경로가 서로 보강 간섭을 일이켜 소듐의 7p 여기를 강화시킴을 실험 및 해석적 식을 통하여 알 수 있었다. 우리는 우리의 연구가 다광자 흡수 계에서 양자 논리 게이트의 구현 가능성을 줄 수 있음을 기대한다.

핵심어: 다광자 흡수 과정, 결맞음 제어, 알칼리 원자

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