

Shaping an atomic electron wave packet

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Abstract: We consider coherent control of the shape of an atomic electron's wavefunction using a train of short transform-limited laser pulses. This type of control is experimentally demonstrated by exciting with a train of three pulses and measuring the resulting quantum state distribution. We also present a general theory for control with a train of N pulses in the weak field limit and discuss the extension of this theory to the strong field limit.

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1. Introduction

The coherent excitation of a superposition of quantum eigenstates results in the formation of a wave packet whose shape and evolution can be controlled by adjusting the relative amplitude and phase of the states in the superposition. With such shaped wave packets the optical, mechanical, chemical and electrical properties of matter can be controlled. This leaves coherent control researchers with the task of determining the shape and evolution of the wave packet necessary to produce the desired effect as well as a method of exciting the superposition state that makes up this wave packet. In this paper we focus on the second aspect of this problem by developing a scheme for coherently exciting a wave packet of any desired shape.

In one coherent control approach the fundamental and second harmonic of a laser field are mixed and used to excite a superposition of states with different parity, but the same energy.¹ In an atomic system this scheme was used to control the angular distribution of ionized electrons by adjusting the relative phase between the two fields,² and in an unbiased semiconductor superlattice, the direction of current flow was controlled in a similar way.³

Another control approach is to shape the spectrum of the exciting field by using an appropriate filter in the frequency domain.^{4,5} Recent advances in shaping techniques for femtosecond pulses have made possible the production of very complicated exciting fields.^{6,7,8} Using tailored optical fields, shaped nuclear vibrational wave packets have been excited in diatomic molecules,^{9,10} as well as shaped radial electron wave packets in atoms.¹¹ These pulse shaping techniques have also been used to control charge oscillations in coupled quantum wells.¹²

The method of control that we have developed also uses a shaped exciting field, however, the shaping is done in the time domain. It comes out of an intuition gained from careful study of the free evolution of atomic electron wave packets, and is particularly well suited to the discrete nature of the quantum mechanical system. This method consists of exciting the atom with a train of transform-limited laser pulses to form a shaped radial electron wave packet. The pulses in the train are spread out equally in time over a single Kepler period (the natural time scale for this system) to ideally span the accessible space. By careful control of the relative amplitude and phase of each pulse in the train, a wave packet of any desired shape can be excited.

This paper is organized as follows. In Section 2 we describe the excitation of a radial electron wave packet with a single short laser pulse. We also review the free time evolution of this wave packet, in particular noting the phase evolution of the complex wavefunction. Section 3 begins with an outline of the general experimental setup used to excite shaped wave packets, and the detection method used to verify their shape. Next, we discuss the excitation with a train of three pulses, demonstrating the nature of this type of control. A simple theory is presented in Section 4, which generalizes the excitation to a train of N pulses, allowing for a wave packet of any shape to be created. Here we also comment on the extension of this simple weak field theory to the strong field limit. Finally, in Section 5, we draw some conclusions.

2. Radial wave packet

The interaction with a short laser pulse to produce an atomic electron wave packet was introduced by Parker and Stroud in 1986.^{13,14} They considered a laser pulse tuned to excite the atomic electron from its ground state to the Rydberg series, where the energy eigenstates are closely spaced. In this region the laser pulse can have enough coherent bandwidth to excite several Rydberg states simultaneously. The result of exciting such a superposition is the localization of the electron in the radial coordinate to a shell of probability that oscillates in and out in a breathing motion.

Initially, these radial oscillations of the electron have a period that correspond to that of a classical particle traveling in an elliptical orbit of the same energy. However, this classical oscillation of the electron does not persist forever. Since the atomic potential is anharmonic, the electron wave packet spreads after making several orbits. Once the electron is spread around the entire orbit, its wave nature becomes evident as it begins to interfere with itself producing a series of fringes. This interference eventually leads to a nearly complete reformation of the wave packet, which is referred to as a full revival. At fractions of the full revival time, the wave packet splits into several miniature replicas of the original wave packet spaced evenly around the Kepler orbit. For instance, at the one-half fractional revival, the wave packet is split into two wave packets positioned on opposite sides of the Kepler orbit, each moving at the classical velocity.

It is these fractional revivals that we are most interested in for the purpose of this paper. In the above description, the wave packet initially acts as a classical particle, however, it still retains its wave like nature. In particular, this well localized wave packet also has a well defined phase, and at the fractional revivals, each of the wave packets have a well defined phase relationship. For instance, at the one-half fractional revival, one wave packet's phase is shifted by $+\pi/4$ relative to the phase of the initial wave packet and the other is shifted by $-\pi/4$. So, the two wave packets of the half revival have a relative phase difference of $\pi/2$. At the one-third fractional revival, two of the wave packets have zero relative phase difference while the third is shifted by $2\pi/3$.

The phase evolution of the wave packet can be seen very clearly if we look at the evolution of a circular orbit wave packet. This wave packet is made up of a superposition of Rydberg states with maximum angular momentum rather than the low angular momentum states of the radial wave packet. A circular orbit wave packet is localized in all three spatial coordinates and moves along a circular trajectory about the nucleus. This allows us easily to represent the phase of the wave packet using color. In Fig. 1 we show an animation of the evolution of a circular orbit wave packet. The probability distribution in the $x - y$ plane is plotted as the height out of this plane and the phase distribution is represented using different colors for the different phases of the wavefunction. Although the phase evolution of a radial wave packet is identical to the circular orbit wave packet, it is more difficult to represent visually since the ingoing and outgoing waves in the radial direction cannot be separated.

The control scheme that we have developed in some ways mimics the fractional revivals seen in the free evolution of a wave packet by directly exciting them. In this direct excitation, not only is the probability distribution important, but so is the phase distribution, both of which we can control.

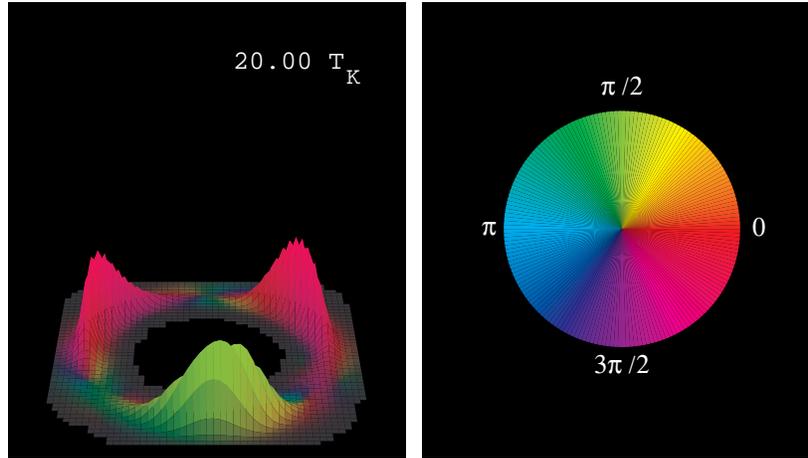


Fig. 1: (QuickTime animation 2.2 Mb) Phase evolution of a circular orbit wave packet. The probability distribution in the $x - y$ plane is plotted as the height out of this plane and the phase distribution is represented using different colors for the different phases of the wavefunction. The color wheel to the right of the figure shows the continuous mapping of the phase onto color. This wave packet is made up of a Gaussian distribution (full width at half maximum of 8) of circular states centered near principle quantum number $n_0 = 180$. Time is in units of the Kepler period, T_K , and the frame shown above is at the time where the one-third fractional revival appears. The diameter of the orbit is slightly over $3 \mu\text{m}$.

3. Experiment

Our control scheme is based on the excitation of a wave packet, not with a single transform limited laser pulse as was described above, but with a train of transform-limited pulses. These pulses are spaced equally in time over a total duration of one Kepler period. With such a train of pulses we can directly mimic the fractional revival structures that result from free evolution of a single wave packet. We can also vary the relative amplitude and phase of the different wave packets that make up a fractional revival to excite more generally shaped wave packets.

A portion of the experimental setup used to generate trains of up to three pulses is shown in Fig. 2. We used a mode-locked, Q-switched, and frequency-doubled Nd:YAG laser to synchronously pump a dye laser, which generated the short, tunable, optical pulses. A single pulse was switched out of the dye laser at the Q-switch repetition rate of 800 Hz and frequency doubled in a β -barium borate crystal. This pulse was nearly transform limited with a width of about 20 ps. The pulse was then sent into a series of beam splitters and delay lines to generate the pulse train. The relative delay between different pulses in the train was actively and independently controlled with a simple servo system so that each pulse had a well defined and controllable phase relationship. Finally, the pulse train was sent into a vacuum chamber to excite the desired wave packet in an atomic beam of potassium.

We used state-selective field ionization to measure the population distribution of the excited Rydberg states. Although this measurement does not reveal the phase relationship between excited Rydberg states, it does provide a simple demonstration of our method of coherent control.

To state-selectively field ionize we ramped on an electric field after the laser excitation to the Rydberg states. This electric field ramp ionized different Rydberg

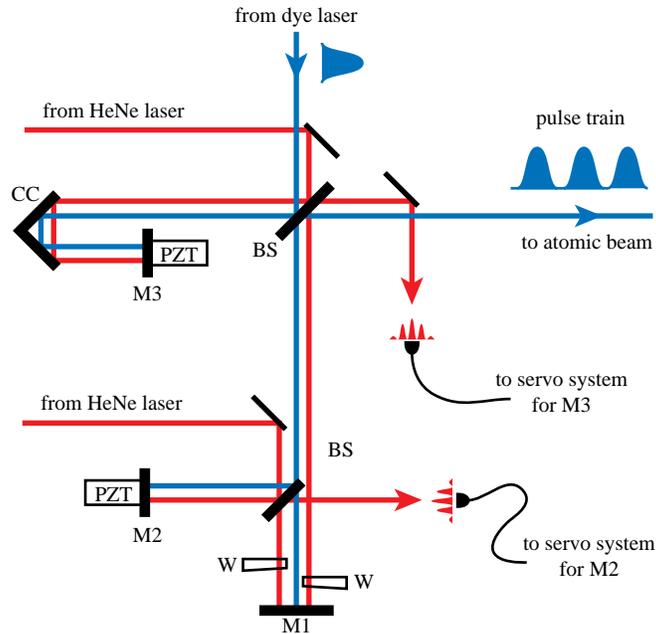


Fig. 2: Experimental setup used to produce a train of phase locked pulses. The beam splitters (BS) split a single laser pulse into three. The mirrors (M1, M2, and M3) are adjusted to recombine these three pulses so that they exactly spatially overlap, but are delayed in time. Two HeNe laser beams are used to actively stabilize the relative phase between pairs of pulses. This is done interferometrically by sending the HeNe beam in one of the delay arms through a glass wedge (W) to produce tilt fringes in the recombined HeNe beams. The position of these fringes and thus the delay between pulses can be locked with a simple servo system through feedback to the piezo electric translator (PZT) on which the associated mirror is mounted. By translating the fringe detector in the servo system the delay between pulses can be varied with an accuracy of 1/100th of the optical period.

states at different times as each states ionization potential was reached during the ramp. By collecting the resulting ions as a function of their arrival time we obtained an ion signal with a series of peaks, which corresponded directly to the different states in the Rydberg series. The integrated signal under each of these peaks is a direct measure of the population in each Rydberg state.

We began by exciting with a pair of pulses whose amplitudes were the same and separation was one-third the Kepler period. We adjusted the relative phase of the two pulses in small steps, measuring the population distribution at each step. The three most dramatic population distributions are shown in Fig. 3. For each of these phases a different set of states is eliminated from the superposition. In each case the states that are missing are separated by three, but with a different offset. To the right of each experimental data set we show a cartoon representation of the pulse train used in each case. The phase written above each pulse was not experimentally measured, but is the phase that would result in the complete elimination of the states highlighted in the data.

In Fig. 3(a) we show the state distribution resulting from excitation with a single pulse. This trace is used as a reference to identify the locations of the different states in the superposition. We also see here that the different peaks are not completely resolved. The appearance of some signal in the region of blue highlighted states of Figs. 3(b)-(d) is

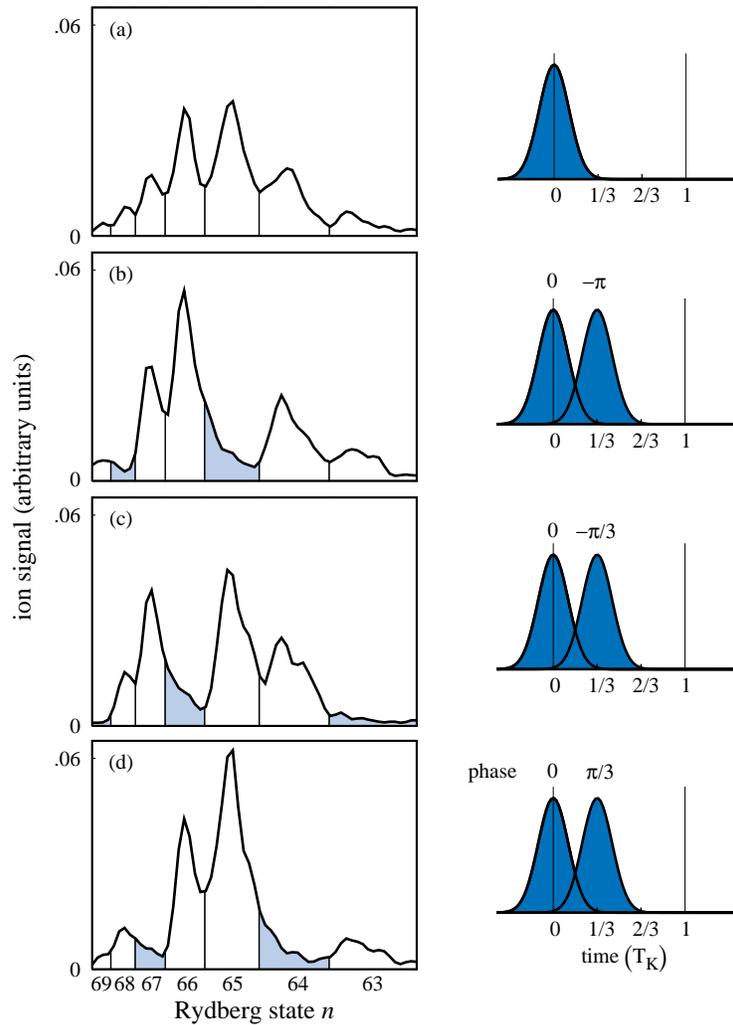


Fig. 3: Control with a pair of pulses separated by one-third the Kepler period, T_K . The top state distribution, (a), was taken with only one pulse present and is used as a reference to identify the locations of the different states in the superposition. The next three state distributions, (b)-(d), are for three choices of phase between the two pulses. The phases were chosen to have the effect of nearly eliminating population from one of the three central states in the superposition.

in fact mostly due to the signal from the wings of neighboring states.

Although we have not directly measured the relative phase between the states in the superposition, these oscillatory state distributions are an indication of the relative phase between the separated wave packets that were excited with the pulse pair. This correspondence has been discussed in great detail in the context of Schrödinger cat states^{15,16} and in fact the recent experimental demonstration of a Schrödinger cat state¹⁷ is an excellent example of a simple form of this method of coherent control.

Next we investigated the effects of a train of three pulses, again of equal amplitudes and separated by one-third the Kepler period. Using the first data set as a calibration of the phase we adjusted the phase between the first two pulses to $2\pi/3$

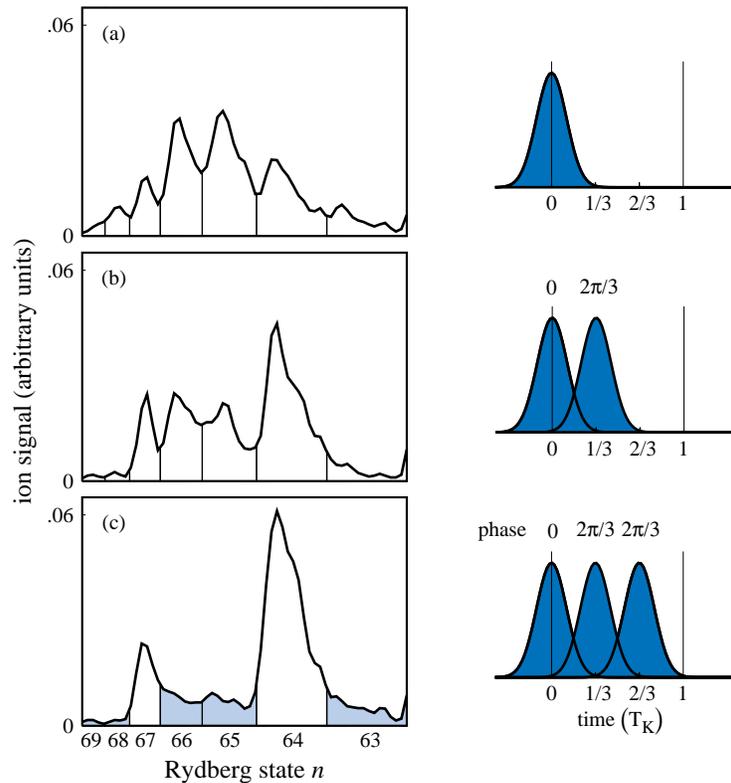


Fig. 4: Here the effect of using three pulses in the control train was investigated. The top state distribution, (a), is again a reference trace taken with only one pulse present. Next, in (b), the second pulse was turned on and its phase adjusted to equalize the population in $n = 65$ and 66 . Finally, the third pulse was turned on, and its phase adjusted to form the distribution shown in (c), which is dominated by every third state.

with the third pulse off. This resulted in the state distribution shown in Fig. 4(b). Now, when the third pulse was turned on, its phase could be adjusted to leave a distribution dominated by every third state, in this case $n = 64$ and 67 .

By spreading the train of pulses evenly over one Kepler period we can ideally span the space over which we can excite a wave packet. If the wave packet is made up of a superposition of three states, each with a complex amplitude, then the amplitudes and phases of the three laser pulses map one-to-one to uniquely determine the three state amplitudes. Our experiment did not quite meet this ideal because the pulses were sufficiently short so that they produced a superposition of approximately five states. The three complex control parameters were then able to determine the state amplitudes modulo three, *i.e.*, in sets of three. Had we used slightly longer pulses, or tuned to lower levels we might have approached the ideal even more closely.

4. General control

In this section we develop the theory for excitation with a train of N pulses. We begin by assuming that the excitation is in the perturbative regime in which the experiment

was done. A train of N pulses can be written,

$$\vec{E}(t) = \sum_{j=1}^N \mathcal{E}_j f(t - T_j) \left[e^{i\omega(t-T_j)} + e^{-i\omega(t-T_j)} \right] \hat{z}, \quad (1)$$

where \mathcal{E}_j and T_j are the amplitude and time delay of each pulse, and ω is the laser carrier frequency. The slowly varying envelopes of the pulses, $f(t - T_j)$, are assumed to be identical in shape, but offset in time. We tune the carrier frequency of our laser to couple the ground state of the atom to the Rydberg series. In the interaction picture state basis, $\psi_g|g\rangle + \sum_n \psi_n e^{-i\omega_n t}|n\rangle$, the time evolution of the excited state amplitudes are,

$$\dot{\psi}_n = i \sum_j \frac{d_n}{\hbar} \mathcal{E}_j f(t - T_j) e^{-i\Delta_n t} e^{i\omega T_j} \psi_g. \quad (2)$$

Here we have made the rotating-wave approximation and defined the dipole moment $d_n = \langle n|\vec{d}|g\rangle \cdot \hat{z}$ and the detuning $\Delta_n = \omega - \omega_n$. In the weak-field limit the ground state population remains unchanged, $\psi_g \approx 1$, so we can formally integrate Eq. 2 to get,

$$\psi_n = i \frac{d_n}{\hbar} \sum_{j=1}^N F_j \int_{-\infty}^t f(t' - T_j) e^{-i\Delta_n t'} dt', \quad (3)$$

where $F_j = \mathcal{E}_j e^{i\omega T_j}$.

Within the slowly varying envelope we can approximate the time delay of each pulse to be uniformly spread out over one Kepler period, $T_j = (j - 1)T_K/N$. Now if we extend the upper limit of integration to $+\infty$ we get the equation for the excited state amplitudes after the interaction with the pulse train:

$$\psi_n = i \frac{d_n}{\hbar} \sum_{j=1}^N F_j e^{-i\Delta_n j T_K/N} \mathcal{F}(\Delta_n), \quad (4)$$

where $\mathcal{F}(\Delta_n)$ is the Fourier transform of the pulse envelope,

$$\mathcal{F}(\Delta_n) = \int_{-\infty}^{\infty} f(X) e^{-i\Delta_n X} dX. \quad (5)$$

Finally, we define a scaled state amplitude as $\Psi_n = \psi_n / i d_n \mathcal{F}(\Delta_n)$ which reduces Eq. 4 to:

$$\Psi_n = \sum_{j=1}^N F_j e^{-i\Delta_n j T_K/N}. \quad (6)$$

When written in matrix notation, this equation looks like:

$$\mathbf{\Psi} = \mathbf{M}\mathbf{F}, \quad (7)$$

where the elements of the matrix \mathbf{M} are $M_{nj} = e^{-i\Delta_n j T_K/N}$ and the elements of the column vectors $\mathbf{\Psi}$ and \mathbf{F} are Ψ_n and F_j respectively.

Now, by simply inverting the matrix \mathbf{M} , we can find the amplitude and phase of each pulse in the train necessary to excite a given state distribution:

$$\mathbf{F} = \mathbf{M}^{-1}\mathbf{\Psi}. \quad (8)$$

Note that, in general, to control the amplitude and phase of N different states we need a train of N pulses.

In both the experimental results presented earlier and the theoretical results presented here we have concentrated on the control of a complex state distribution. A given state distribution is directly correlated with a given shaped wave packet. If one would rather specify the complex radial distribution of the electron, it is a simple task to find the corresponding state distribution by performing overlap integrals between the desired wave packet and the radial eigenfunctions.

The use of a discrete excitation is one of the strengths of our control scheme since it takes advantage of the fact that the superposition we are trying to excite is discrete. This makes the problem we are solving Eq. 7 well specified and its solution unique because we have exactly the right number of parameters to vary for the system we are trying to control.

Spreading the pulses out equally over one Kepler period allows us to optimally span the available space and excite population anywhere around the orbit. When we have as many pulses as there are states in the distribution these pulses will overlap significantly. Coherently interfering all of these pulses is certainly a method of pulse shaping, however, this method of pulse shaping has exactly the proper resolution to match the features of the wave packet we are trying to excite.

One important aspect of coherent control is its extension to the strong field limit. In this limit the ground state population does not remain approximately constant, and in fact one would like to be able to invert the atom completely, that is, to excite all of the population to the wave packet state. We conclude this section with some discussion of the extension of our control techniques to this strong field limit.

One intuitive way of approaching this limit is to consider a train of many pulses, which individually interact perturbatively, but in combination result in a large population transfer. Let's refer back to Eq. 3 but now include the changing ground state amplitude,

$$\psi_n = i \frac{d_n}{\hbar} \sum_{j=1}^N F_j \int_{-\infty}^t f(t' - T_j) e^{-i\Delta_n t'} \psi_g(t') dt'. \quad (9)$$

Since the population excited to the Rydberg series by the first pulse is in the form of a wave packet, it will have moved away from the core by the time the second pulse arrives, and the population excited by the second pulse will have moved out of the way in time for the third pulse, and so on. So, we only need to take into account the small (perturbative) change in the ground state population after one pulse before considering the next pulse. Once again, we need to be careful to limit the total duration of the pulse train to one Kepler period so that the wave packet excited by the first pulse cannot return to the core in time to interfere with a later pulse in the train.

The flaw in the above logic is the assumption that the pulses do not overlap. As we saw in the weak field analysis, in order to control N states we need N pulses, and if a single pulse has only enough bandwidth to excite a total of N states then these N pulses will overlap significantly in time. So, the ground state amplitude is really going to change according to some complicated function of the pulsed field rather than in discrete steps as described above. The ground state amplitude can be written,

$$\psi_g = \exp \left[-\gamma \sum_j \sum_{j'} F_j^* F_{j'} \int_0^t f(t' - T_j) f(t' - T_{j'}) dt' \right]. \quad (10)$$

Here, γ is the decay rate, proportional to the dipole moment (assumed to be constant here) and inversely proportional to the level spacing in the Rydberg series (also assumed to be constant).

Beginning with the weak field solution of Eq. 8 for the pulsed field and using an iterative technique with Eq. 10 should lead to the strong field solution for the pulse train. Over the time scale of one Kepler period, the total duration of the pulse train, the Rydberg series effectively looks like a continuum. So, we expect the ground state population to decrease much like a *Fermi's golden rule* type of decay in a photoionization process. As we iterate on the weak field solution we expect the pulses at the end of the train to become stronger since there is less total population available for them to work with.

5. Conclusion

We have designed a method of coherently controlling the shape of a wave packet using a train of transform-limited pulses. Although this control technique was demonstrated in an atomic system it should be useful in any system in which one would like to excite a tailored superposition of quantum states using a short laser pulse. Such systems include molecules, semiconductor superlattices, and excitons in addition to the simple atomic system.

This control scheme may be particularly useful when there are relative few states in the wave packet superposition. Its experimental setup was straightforward for three pulses and could have easily been extended to five pulses. With more overlapping pulses beam splitters would not be the best way of generating the needed field, but the technique of limiting the exciting field duration to one Kepler period, and matching the number of field parameters to the number of levels forming the wave packet remains optimal.

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