

Interaction of a finite train of short pulses with an atomic ladder system

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When a train of optical pulses interacts with a medium, the time between pulses and the pulse-to-pulse phase jump have a profound effect on the outcome of the interaction. For a near-infinite train of regularly separated pulses having a constant pulse-to-pulse phase jump, as in the output of a frequency comb laser interacting with a ladder system, excitation of that system has been previously calculated and compared with experiment. In that case, the number of pulses in the train is very large, and the energy per pulse is very small. In the present work, the other extreme is experimentally examined: A small number of regularly spaced optical pulses are made to interact with a three-level ladder system in atomic rubidium. The pulses have been amplified, possibly placing the interactions into a nonperturbative regime. The experimental results are compared to a simple intuitive model and found to be in good general agreement. However, some aspects of the experimental results seem to be at odds with the simple model.

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The interaction of an infinite series of identical short optical pulses with two-level [1] and multilevel [2,3] ladder systems has been the subject of several recent articles, partly because of its application toward direct frequency comb spectroscopy [4–6]. Although the interaction of frequency combs with atomic or molecular systems is generally described in the frequency domain, it has also been modeled [1,3] in the time domain. In another interesting class of experiments, a pair of pulses interacts with an atomic or molecular system. In the one-color version of this pump-probe arrangement, a single fast optical pulse is split into two pulses, which are then recombined after first introducing a variable delay between them. One application of the one-color pump-probe interaction is coherent control [7], and the interaction of the laser field with the atomic or molecular systems is generally described in the time domain. In the work we describe here, we bridge the gap between the very large number of optical pulses typical of frequency combs and the two pulses employed in one-color pump-probe experiments. Specifically, we study the interaction of a ladder system with a train of 3–11 optical pulses, separated by a well-defined time interval and having a well-defined pulse-to-pulse temporal phase change. The purpose of these experiments is to provide data that can be compared to theoretical models that should be generally applicable to any ladder system interacting with a regular train of short optical pulses, whether the number of pulses in the train approaches infinity, as in the case of a frequency comb, or is limited to two, as in pump-probe experiments.

To get a simple, intuitive picture of the interaction of a finite train of pulses with a ladder system, first consider a two-level system in which the ground and excited states are labeled $|g\rangle$ and $|e\rangle$, respectively. We assume that prior to the interaction, all of the population is in $|g\rangle$. At time $t = 0$, one of the pulses from the pulse train excites some of the atoms to $|e\rangle$. There the phase of the wave function, $\Psi_e(t)$, evolves as $\exp(i\omega t)$, where $\hbar\omega$ is the energy difference between $|g\rangle$ and $|e\rangle$. The next pulse in the sequence, the phase of which differs from the previous

one by ϕ_0 , can excite more of the population from $|g\rangle$ to $|e\rangle$. Excitation to $|e\rangle$ is maximized when the total phase, that is, the accumulated phase of Ψ_e after time T and the additional relative phase ϕ_0 of the second pulse, is equal to some integer times 2π ; that is, there will be constructive interference when

$$\omega T + \phi_0 = 2\pi m, \quad (1)$$

whereas the interference will be destructive when

$$\omega T + \phi_0 = \pi(2m + 1) \quad (2)$$

for all integers m . This model is not new and has been the basis for pump-probe experiments [7] as well as descriptions of direct frequency comb spectroscopy experiments [1,4]. In this simple model, stimulated emission is not considered but should not substantively change the general predictions of Eqs. (1) and (2) because this mechanism for redistributing the population is coherent. Spontaneous emission, on the other hand, is a stochastic process and can significantly affect the result. For example, if the spontaneous lifetime is comparable to or shorter than the pulse repetition time, one would expect to see a uniform background of incoherent excitation that is independent of T and ϕ_0 .

If one were to create a landscape plot in which the population of $|e\rangle$ is plotted as a function of T and ϕ_0 , one would expect to see parallel ridges of large population and valleys of small population. These ridges and valleys should lie along lines having slopes given by the derivative of Eq. (1) or (2):

$$d\phi_0/dT = -\omega. \quad (3)$$

Thus, if one were to measure such a landscape and determine the slopes of these ridges and valleys, one would obtain the excitation frequency of the transition.

The simple model of multipulse excitation could be extended to multilevel ladder systems as well. In a three-level system, there would be two transitions, one between the ground and intermediate states, $|g\rangle + \hbar\omega_1 \rightarrow |i\rangle$, and another between the intermediate and excited states, $|i\rangle + \hbar\omega_2 \rightarrow |e\rangle$. The simple model then predicts that the maximum production of $|e\rangle$ would occur when both transition frequencies, ω_1 and ω_2 , simultaneously satisfy Eq. (1) (but with different values of m)

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[2]. One might also expect that there would be ridge lines along $d\phi_0/dT = -\omega_1$ and $d\phi_0/dT = -\omega_2$. The simple model, of course, does not show any coherence between the two steps of the excitation. To get these, one would have to solve the optical Bloch equations or directly integrate the time-dependent Schrödinger equation. This is more tedious but has the advantage of making fewer assumptions about cross-terms in the excitation process. In describing the interaction of a multilevel ladder system with a frequency comb, the former approach has been used [1,2], except that in that work, some approximations equivalent to those in perturbation theory were made.

To test the simple model, as well as to provide data for testing more rigorous models such as those of [2,3], we apply a finite train of ultrafast pulses to a three-level ladder system in Rb. To this end, single pulses were selected from an 80 MHz pulse train by a Pockels cell having a repetition rate of 2 kHz. The single pulses had a central wavelength of 795 nm and temporal widths of approximately 35 fs. A sinusoidal spectral phase was applied to the pulses using an acousto-optical programmable dispersive filter pulse shaper [8,9]. The shaped pulses were then amplified and directed onto the target.

The sinusoidal spectral phase is defined here as

$$\phi(\omega) = A \sin(\omega T + \phi_0), \quad (4)$$

where A , T , and ϕ_0 are parameters that are chosen as described later. As is well known [10], when the spectral phase of Eq. (4) is applied to an optical pulse, the result in the time domain is a series of replicas of that input pulse; that is,

$$E_{\text{out}}(t) = \sum_{n=-\infty}^{+\infty} J_n(A) E_{\text{in}}(t - nT) \exp(-in\phi_0), \quad (5)$$

where E_{in} and E_{out} are the electric field envelopes before and after application of the sinusoidal phase, respectively. The replicas are weighted by the Bessel function, $J_n(A)$; they are temporally separated by T ; and they differ in temporal phase by ϕ_0 . Equation (5) implies an infinite series of replicas, but the effective number is determined through the choice of A . A plot of $|E_{\text{out}}|$ from Eq. (5) versus time for values $A = 2.5332$, $T = 250$ fs, and $\phi_0 = \pi/4$ is shown in Fig. 1. In this example, $E_{\text{in}}(t)$ is taken as a Gaussian 35 fs in width. Also shown in the figure is a plot of the relative phase between the pulses in units of π . The phase shifts are asymmetric about the central $n = 0$ peak because of the well-known relationship $J_{-n}(x) = (-1)^n J_n(x)$, giving rise to an additional π phase jump between pulses for odd negative values of n . Except for this (well-defined) phase jump irregularity, the Bessel function amplitudes, and its truncated nature, the train of pulses in Fig. 1 resembles the output from a frequency comb laser that has been plotted in the time domain.

In the target region, the train of shaped pulses has an energy of about $2 \mu\text{J}$ in a 0.2 cm^2 spot. The target consists of a sample of ^{87}Rb that is trapped and cooled to approximately $150 \mu\text{K}$, thus allowing us to ignore Doppler effects. The relevant levels in the ladder are $\text{Rb}(5s_{1/2})$, $\text{Rb}(5p_{3/2})$, and $\text{Rb}(5d_{3/2,5/2})$. [The fine structure splitting in $\text{Rb}(5d)$ is not resolved in these experiments, and the $5p_{1/2}$ - $5d_{3/2}$ transition is outside the bandwidth of the laser.] The lower and upper transition wavelengths are $\lambda_1 = 780.03 \text{ nm}$ and $\lambda_2 = 775.79 \text{ nm}$, respectively. Both transitions lie within the approximately 30 nm bandwidth of

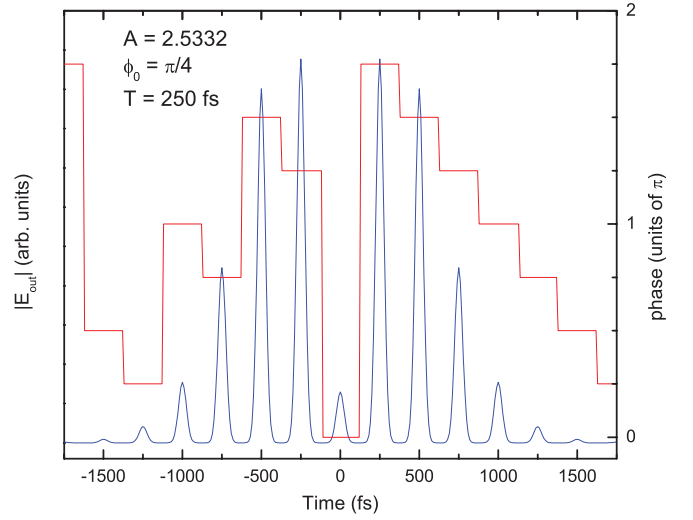


FIG. 1. (Color online) The effect of a sinusoidal spectral phase, like that of Eq. (4), on a Gaussian transform-limited input pulse. Here the curve of pulselike structures is E_{out} (blue online), while the boxy curve shows the absolute phase of each pulse, relative to some arbitrary zero (red online).

the laser. For convenience, Eq. (3) is put into its corresponding wavelength form, and an offset wavelength of $\lambda_0 = 790 \text{ nm}$ is used. (This does not change the physics but allows for more readily measured slopes and simpler comparison with the predictions of the simple intuitive model.) Equation (3) then becomes

$$\lambda = \left[\lambda_0^{-1} - \left(2\pi c \frac{dT}{d\phi_0} \right)^{-1} \right]^{-1}. \quad (6)$$

In the experiment, $\text{Rb}(5d)$ is detected through photoionization by the same train of optical pulses that is used for excitation, resulting in Rb^+ . The ions are detected using a time-of-flight spectrometer [11]. The Rb^+ signal is shown plotted versus T and ϕ_0 in Fig. 2. For the data in Fig. 2, $A = 2.5332$, resulting in a train of approximately 11 pulses. (See the appendix for a discussion on the choice of A .) Several features can be observed in this plot, the most obvious being the diagonal structures, which may be described as ridges and valleys. A careful inspection of the diagonal structures reveals a gash across the ridge. Furthermore, when the figure's signal scale is expanded, a small ridge parallel to the gash can also be seen cutting across the valley.

The solid lines in the figure were fit to the main features of the ridges and valleys. When inserted into Eq. (6), the slope of the solid lines yields a wavelength of 781.6 nm, approximately corresponding to the $5s_{1/2}$ - $5p_{3/2}$ transition. The dashed line follows the gash. When inserted into Eq. (6), the slope of the dashed line yields a wavelength of 778.0 nm, approximately corresponding to the $5p_{3/2}$ - $5d_{5/2}$ transition. The structures and their slopes, then, were the results expected from the simple model.

Although a simple extension of the two-level model to our three-level ladder would seem to predict constructive interference when both transition frequencies simultaneously satisfy Eq. (1), in our experiments, we clearly measure destructive interference between the two excitation channels.

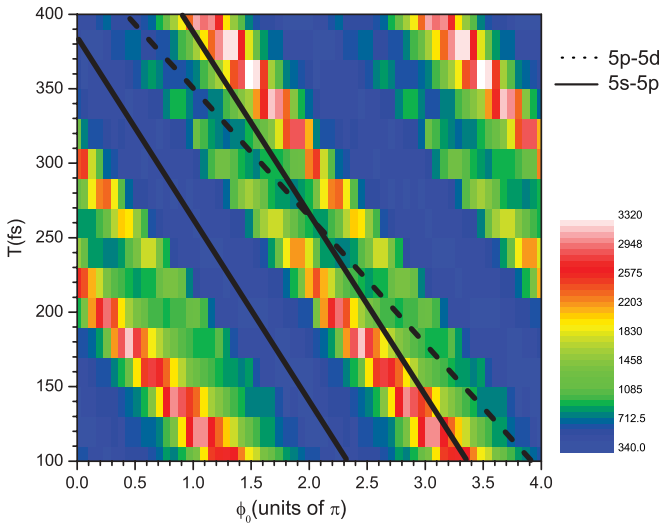


FIG. 2. (Color online) Landscape plot of Rb^+ counts vs. T and ϕ_0 for $A = 2.5332$, which gives rise to a train of 11 significant pulses. The structures correspond to constructive and destructive interferences of Eqs. (1) and (2) from the $5s-5p$ and $5p-5d$ transitions. In taking the data, ϕ_0 ranged from 0 to 2π . In order to make clearer the diagonal structures, the data were copied and replotted so as to artificially extend the range of the ϕ_0 axis to 4π .

We do not know the cause of this discrepancy, which is both clear and reproducible. However, we point out that there are some differences, which may be significant, between the simple model system and the one measured here. The first is that the simple model neglects both spontaneous and stimulated emission. Spontaneous emission is negligible on the time scale of T , the time between pulses, or indeed, on the time scale of the entire pulse train, which is less than 4 ps.

Recent experiments have shown [12] that the details of interference effects in the excitation of ladder systems by ultrafast optical pulses depend on whether the system is in the strong or weak field regime. We believe that our experiments are in the weak field regime because we reduced our laser pulse energy by an order of magnitude and saw no difference in our results except for a corresponding reduction in signal rate. Nevertheless, even with the reduced intensity, it is not clear that we were in the weak field regime. Furthermore, even in the weak field regime, one cannot neglect stimulated emission. However, as already mentioned, while stimulated emission may affect the magnitude of the excitation, it should not affect the general form of the landscape plot.

As a weak check on whether we are in the weak or strong field regime, we made a simple second-order perturbation theory calculation [13] for the $\text{Rb}(5s)-\text{Rb}(5p)-\text{Rb}(5d)$ three-level ladder system. The results [14] were only partially in agreement with experiment: The large diagonal structures were present, but the gashes cutting through them were not. Furthermore, the contrast in the calculated landscapes was far smaller than in the measured case. We can think of no reason for a difference between this calculation and our measurements other than the fields were too strong for a perturbative model to be valid.

A second difference between our system and that in the simple model is the extra π phase shift for odd negative-order

pulses in our train. It is not at all obvious to us how this could give rise to the destructive interference that we observe between the two excitation channels.

Finally, even though the slopes of our structures yield transition wavelengths that are qualitatively consistent with the $5s-5p$ and $5p-5d$ transitions through the use of Eq. (6), they differ quantitatively by more than expected from fitting errors. It is possible that this is because of poor calibration of our pulse shaper, for which the absolute spectral uncertainty was approximately ± 2 nm. (The relative spectral uncertainty is less than 0.1 nm.)

The simple model is completely analogous to the multislit wave interference. In diffraction, the greater the number of slits, the sharper the diffraction image. In the process described here, the greater the number of pulses in the train, the better resolved the interference structures should be. We therefore created a series of landscape plots like that of Fig. 2, but with the parameter A chosen so as to give different numbers of pulses in the train. The results are shown in Fig. 3. All the plots of Fig. 3 show the same diagonal structures. However, as N is increased from 3 to 7 to 11, the contrast in the interferences becomes clearer and sharper, as expected. One can readily imagine that if N were increased to several thousand, as in a frequency comb, then the resolution in the interferences would become extremely sharp such that the slopes of the structures could be used to determine the transition frequencies with very high precision, as is done in direct frequency comb spectroscopy [4]. In principle, one could obtain frequency comb resolution with a sinusoidal pulse train by decreasing T and choosing A to allow for an arbitrarily large number of pulses in the train. In practice, however, N is limited by the

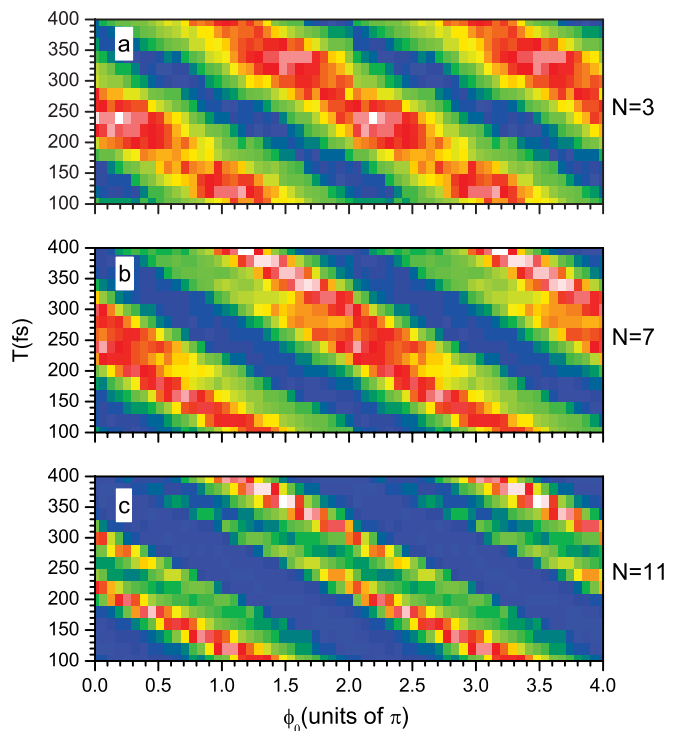


FIG. 3. (Color online) Landscape plots of Rb^+ counts vs. T and ϕ_0 for various numbers, N , of pulses in the train. Plots (a), (b), and (c) have $N = 3, 7$, and 11 , corresponding to $A = 0.3672, 1.3152$, and 2.5332 , respectively.

temporal width of the pulses in the train. For example, we have observed that the structures in our landscape maps wash out below $T \sim 50$ fs because our 35 fs pulses begin to overlap. One can also imagine arbitrarily increasing N by increasing the temporal length of the train itself. In practice, the train's length is limited by the pulse shaper. In our case, the train is less than 4 ps.

In summary, the application of a sinusoidal spectral phase to a transform-limited ultrafast optical pulse was used to test a simple model of cumulative phase effects in a three-level ladder system. The simple intuitive model was found to be in very good agreement with the data, except for the interference between the structures originating from different transitions in the ladder and for a slight quantitative discrepancy in the measured transition wavelengths. The landscape data that were taken can be used to test the much more comprehensive model of [3]. It would be instructive to solve the optical Bloch equations, in a nonperturbative scheme using the intrapulse phase jumps employed in our experiment, in order to see if the interference discrepancy can be resolved. When the number of pulses in the train was increased, the structures in the landscape plots became dramatically sharper, just as optical spectra are sharper in multislit interference than in two-slit interference experiments. The sharpening of the interference structures can also be explained by the fact that more pulses in the time domain gives rise to narrower comb teeth in the frequency domain.

The structure in the landscape of Fig. 2 is reminiscent of a quantum carpet [15]. Indeed, in both cases, the structure is caused through constructive and destructive interferences. Thus, even though the processes by which the interferences are created are different, the underlying physics is quite similar.

The physics behind the interaction of a train of ultrafast pulses with an atomic or molecular system is completely general, and one can imagine enhancing experiments [16] that use attosecond pulse trains [17] (APT) by scanning their pulse-to-pulse temporal spacing. This can readily be done since the time between pulses in an APT is simply half the optical period of the carrier electric field. Therefore a pulse shaper can be used to select the carrier frequency of the fundamental pulse before high-order harmonic generation. In the future, we hope to extend our measurements to APT, interacting with a suitable system, in order to test the general nature of this interaction.

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APPENDIX: BEST CHOICE OF A FOR N PULSES IN THE TRAIN

In general, the sinusoidal phase of Eq. (4) gives rise to an infinite train of pulses in the time domain, as indicated in Eq. (5). However, practically speaking, the sum in Eq. (5) is effectively truncated and runs from $-N'$ to $+N'$, where N' is related to the total number of significant pulses N by $N = 2N' + 1$. The choice of A in Eq. (4) is determined by the

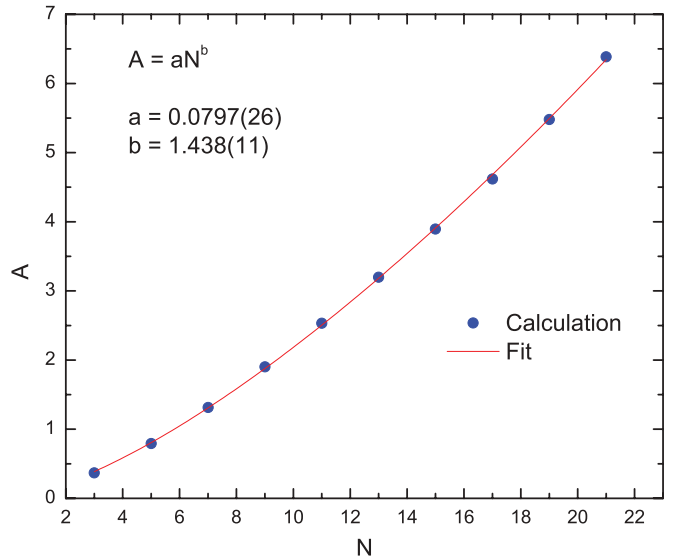


FIG. 4. (Color online) Values of the Bessel function argument as a function of the desired number of pulses in the train for $f = 0.05$. The points are from direct computation, while the solid curve is a fit to the indicated power law.

desired number of pulses in the train and by the definition of *significant*. For example, one could claim that the $(N' + 1)$ th pulse is not significant if its amplitude is less than or equal to some fraction of the average amplitude of all the pulses having $|n| \leq (N' + 1)$; that is, we can define f as

$$f \equiv J_{N'+1}(A)/\bar{J} = N J_{N'+1} \quad (\text{A1})$$

and require that the last significant pulse has an amplitude greater than f times the amplitude of the average acceptable pulse in the train. Our procedure, then, was to run a simple iteration program that determined A for desired values of N and f such that the condition of Eq. (A1) was satisfied. As shown in Fig. 4, when these values of A were plotted versus N , we found that they fit very well to a power law

$$A' = aN^b, \quad (\text{A2})$$

where a and b are functions of f and a prime is used to distinguish between the fitted value of A and the directly computed one. Our fitted values of a and b , along with the fit errors, are given in Table I for several values of f .

For the work done here, we chose $f = 0.05$, for which case Eq. (A2) yields the values of A' (with associated fit errors) shown in Table II. For comparison, we also show the directly computed value of A .

TABLE I. Table of fitted parameters to Eq. (A2) for several values of f .

f	a	b
0.03	0.0565(9)	1.554(6)
0.04	0.0661(15)	1.500(8)
0.05	0.0797(26)	1.438(11)
0.06	0.0936(3)	1.385(10)
0.08	0.1054(38)	1.352(13)
0.10	0.0981(39)	1.384(14)

TABLE II. Table of the arguments of the Bessel function of Eq. (4) used in this work. Here N denotes the total number of significant pulses in the train; the column labeled A denotes the value of A used in Eq. (4); and the column labeled A' denotes the value of A obtained using the coefficients of Table I for $f = 0.05$. The error in A' comes from the fitting error in a and b .

N	A	A'
3	0.3672	0.387(13)
5	0.7933	0.807(28)
7	1.3152	1.308(50)
9	1.9012	1.878(70)
11	2.5332	2.506(95)

Note that this approach assumes that no pulses having $|n| < N'$ fall below this critical amplitude. In general, for a given A , pulse amplitudes can increase or decrease until n reaches some critical value, whereupon they monotonically and rapidly decrease. However, it is clearly possible that for some unfortunate choice of A and n , one could accidentally be near a zero in the Bessel function, giving rise to a missing or negligible pulse in the train. We intend the approach of this appendix to serve as a convenient guide for choosing A . However, once A is chosen, the entire pulse train given by Eq. (5) should still be examined for its suitability. We believe this numerical inverse function treatment of Eq. (5) to be useful.

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