Ramsey Interference in Strongly Driven Rydberg Systems

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We have studied the evolution of regular and "dark" radial wave packets under the influence of two coherent, temporally separated 100 fs laser pulses. The atomic states are probed by ramped-field ionization. The final state populations exhibit Ramsey oscillations with periods ranging from 2.6–1000 fs as a function of the delay between the two pulses. This time-domain bound-state interferometry reveals several new aspects of the dynamic evolution of wave packets in the presence of strong laser fields. Lowest order perturbation theory is inadequate to describe wave packet formations, even at the lowest intensities studies.

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A considerable amount of experimental and theoretical effort has been exerted over recent years in an attempt to understand the complexities of atomic dynamics in an intense laser field. Recently, it has been shown that multiphoton couplings can either enhance or inhibit photoionization, and population transfer between resonantly coupled states may or may not occur depending on experimental parameters [1,2]. Laser intensity, pulse duration, and frequency as well as the principal and angular quantum numbers of the coupled states are extremely important in determining the details of the laser-atom interaction [1,2].

We have implemented an optical double resonance technique to study the effects of coherent population transfer in an intense laser field. Two identical laser pulses probe the atomic system at different times, creating a time-domain bound-state Ramsey interferometer [3]. This system is analogous to a Mach-Zender optical interferometer. The first pulse acts as a beam splitter, coherently splitting the initial state into two or more final states. The states interact again at another "beam splitter" during the second laser pulse. During the time between the two pulses, all states acquire phase at different rates due to their different energies (analogous to different optical path lengths). After the second pulse, the population in each state exhibits a sinusoidal variation as a function of the delay between the first and second pulses. This modulation is completely analogous to the intensity variation at the output of an optical interferometer as a function of optical path length.

We discuss two different experiments which were performed to study coherence in strongly driven atomic systems using time-domain bound-state interferometry: (1) the formation of a radially localized Rydberg wave packet [4]; and (2) the formation of a "dark" radial wave packet [5]. In these two particularly simple examples, the experimental results are in good agreement with a numerical integration of the time-dependent Schrödinger equation using a small essential state basis. Lowest order perturbation theory, traditionally used to describe wave packet dynamics, is inadequate, even for relatively weak laser pulses. Stimulated Raman processes and other higher order effects which are ignored in lowest order are an important part of the wave packet formation [6]. We conclude with some general remarks concerning the applicability of this technique to studies of coherence in higher order multiphoton processes, where finite basis calculations are no longer feasible.

The laser system which generates the short, coherent laser pulses uses chirped pulse amplification (CPA) of 100 fs pulses from a Ti:Al_2O_3 self-mode-locked oscillator. The amplified pulses have energies up to 10 mJ at 772 nm. A Michelson interferometer splits a single laser pulse into two spatially overlapped, temporally separated pulses. The tie delay between the pulses is controlled by changing the optical path length in one leg of the interferometer using two different schemes. For coarse path length adjustments (> 1 μm) the physical length of one of the interferometer legs is changed by translating the retroreflecting mirror using a precision translation stage. For fine adjustments, the optical path length in one of the legs is altered by rotating a glass slide which is inserted between the beam splitter and retroreflecting mirror in the interferometer. Pulse separations are controllable to a precision of 0.02 fs. The effect of increasing the pulse length in the delayed beam due to group velocity dispersion in the glass slide is negligible.

In the first experiment, a single 100 fs laser pulse creates a localized radial wave packet in K [4]. A second, identical pulse can either deexcite the original wave packet or excite a second wave packet depending on the delay between the two pulses. The population remaining in the individual Rydberg states is monitored as a function of the delay between the two 100 fs pulses.

A 10 ns dye laser at 928.7 nm excites a thermal beam of K atoms from the $4s$ ground state to the $3d$ state via a two photon transition. After a 10 ns delay a single 100 fs, 772 nm laser pulse crates a coherent superposition of nf Rydberg states ($12 < n < 22$) from the $3d$ state. A second identical 772 nm pulse stimulates emission out of the wave packet to the $3d$ state and also drives additional population from the $3d$ level to the Rydberg states. The

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populations in all of the different Rydberg states are then measured using state selective field ionization by applying a slow (~ 5 μs) 10 kV pulse to a set of capacitor plates which straddle the interaction region. Electrons created via field ionization are pushed through a 0.5 mm diam hole in the upper capacitor plate toward a microchannel plate (MCP) detector. Electrons in different energy levels are ionized at different electric fields. Therefore, the population in each Rydberg level is detected as a separate peak in the time of flight spectrum. The dye laser and 772 nm pulses are weakly focused to a ~ 1 mm diam spot, therefore we can neglect an intensity variation for those electrons which are detected.

Previous experiments have employed photoionization to observe the evolution of wave packets [4,5]. The use of ramped-field ionization detection in this experiment improves the signal by 10–100, eliminates the main source of background, i.e., photoionization of ground state atoms, and also permits separate detection of individual states in the wave packet.

Figure 1 shows the population in the $16f$ state as a function time delay between the two pulses. The rapid 2.57 fs optical Ramsey fringes are always accompanied by a slow beat frequency even at low intensities ($\sim 10^{10}$ W/cm^2). The slow modulations correspond to energy differences between any two Rydberg states in the wave packet. At higher intensities ($\sim 10^{12}$ W/cm^2) the temporal interferograms become increasingly complicated, often to the point of resembling random noise. However, by taking the Fourier transform of the time delay spectra, we obtain clear frequency spectra which show all of the energy levels which contribute to the population in a given eigenstate. We have identified up to 18 discrete frequencies in the transform spectrum of a single final state. Frequencies ranging from 20 to 13000 cm^{-1} can be observed in a single scan.

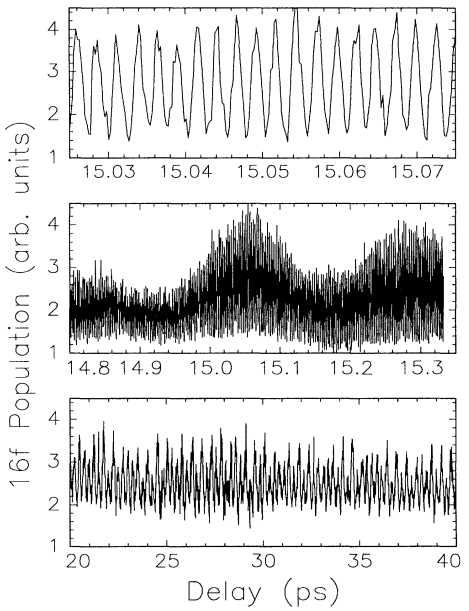

FIG. 1. Electron signal due to population in the $16f$ Rydberg level as a function of delay between the two short laser pulses. The initial state for the excitation is $3d$ and the laser intensity is $\sim 10^{11}$ W/cm^2. Three different time scales are displayed to show the temporal modulations which vary over 3 orders of magnitude. All three scans were taken under identical experimental conditions.

As predicted by Noordam, Duncan, and Gallagher [7], our time delay spectra have a far greater signal to noise ratio than any previously reported Rydberg wave packet experiments due to the extremely high detection efficiency associated with field ionization as opposed to photoionization. A typical scan over a delay range of 50 ps shows approximately 100 oscillations of the wave packet and nearly 20 complete "collapses" and "revivals" [4]. Furthermore, state selective field ionization allows us to study the population in each individual Rydberg state. Such a careful analysis of the final state distribution is not possible using photoionization.

The appearance of low frequency Ramsey fringes is caused by depletion of the $3d$ state and stimulated emission out of the Rydberg states within a single laser pulse. *Hence, lowest order perturbation theory is invalid even at the lowest intensities that we have studied.* We are unable to use the closed form expressions developed by Noordam, Duncan, and Gallagher [7], which are derived from first order perturbation theory. Fortunately, we are able to reproduce the observed temporal and frequency spectra satisfactorily using a numerical integration of the Schrödinger equation with only 22 states—$3d$, $14p$ through $20p$, and $11f$ through $24f$. The calculation assumes a Gaussian laser intensity profile with a 100 fs full width at half maximum (FWHM). We neglect any nonresonant effects between our finite basis set and the other states of the atom, as well as photoionization out of the $3d$ or Rydberg levels.

The calculation and data clearly show a general increase in the number of frequency components which contribute to the Ramsey interferogram with increasing laser intensity. Saturation of the more strongly coupled $3d$-nf transitions as well as depletion of the $3d$ state contribute to the complicated frequency spectra [6]. The dynamics of the wave packet created by the first pulse are also changed, and although localization occurs, different periodicities are apparent in the wave packet oscillation. Thus, varying the intensity of the exciting laser can actually shape the radial wave packet and determine its motion. (This intensity effect is distinctly different from ponderomotive effects [6,8].)

A more controllable method of tailoring wave packets is to alter the frequency spectrum of the exciting laser. A simple example is to eliminate laser frequencies which are responsible for exciting one or several different Rydberg components in the wave packet. This technique is particularly simple using a mask in the dispersed beam in the

grating expander in our laser system. We first study the behavior of a wave packet which has three dominant components: $14f$, $15f$, and $16f$. The wave packet oscillates with the characteristic Kepler period of the $15f$ state and difference frequencies corresponding to the $14f$-$15f$ and $15f$-$16f$ energy differences are the dominant features in the Fourier transform of the Ramsey spectra for the three states. Figure 2 shows the frequency spectrum of a typical interferogram for the $16f$ state. When a small notch is placed in the laser spectrum, there is no excitation of the $15f$ state, but transitions to $14f$ and $16f$ are unaffected. The characteristic frequency of the wave packet changes by approximately a factor of 2. The dominant feature in the Ramsey interferogram is now the $14f$-$16f$ energy separation as shown in Fig. 2. Of course much more complicated frequency tailoring schemes are possible, giving enormous control over the oscillation period and even dispersion compensation within the Rydberg wave packet.

In our second experiment, two tunable dye lasers are used to prepare Rydberg atoms in a single nl eigenstate ($n > 12, l = 0$ to 3). Rydberg states with $l = 0$ or 2 are excited via the $6p_{3/2}$ level, and states with $l = 1$ or 3 are excited via the $3d$ state. The Rydberg atoms are then subjected to a short 772 nm pulse. State selective field ionization is used to determine the Rydberg state population after the intense pulse.

Previous experiments in Ba have shown that population in a single Rydberg level can be redistributed over several different states when the initial level is exposed to an intense short laser pulse [5]. This redistribution is caused by the creation of a dark wave packet or hole near the ion core due to partial photoionization of the Rydberg state wave function. In the frequency domain the effect is attributed to stimulated Raman processes in the continuum which result in population redistribution due to the finite bandwidth of the laser pulse.

In contrast to previous results in Ba, we observe no population redistribution in K via stimulated Raman processes in the continuum even at intensities as high as 10^{13} W/cm^2. However, Rydberg redistribution due to bound state couplings can be seen at intensities below 10^{11} W/cm^2. The nf ($12 < n < 18$) levels in K are resonant with the $3d$ state within the bandwidth of the 772 nm laser pulse. The states which are most heavily depleted are the $14f$ and $15f$ states which have the strongest coupling to the $3d$ state at this frequency. Initially populated np states exhibit no observable redistribution, presumably due to their reduced cross section for stimulated emission to the $3d$ state. However, the redistribution of the f states implies that a coherent superposition state has been created which we can probe using a second laser pulse and the optical Ramsey method.

Once again we measure the population in several different Rydberg states as a function of delay between the two 100 fs laser pulses. As in the case of direct wave packet excitation, these scans generally become extremely complicated as the laser power is increased, corresponding to an increase in the number of frequency components which are present in the wave packet motion. The Fourier transform of a typical Ramsey interferogram is shown in Fig. 3.

There are several advantages of time-domain atomic interferometry over previous schemes for observing dark or antiwave packets [5]. The first advantage is angular momentum identification. The Fourier transform of the Ramsey interferograms yields frequencies which have widths < 1 cm^{-1}. With this high resolution we can easi-

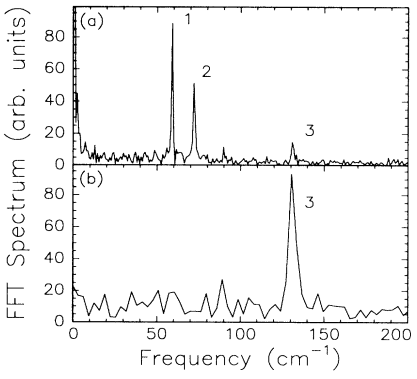

FIG. 2. Discrete Fourier transform spectrum of the Ramsey interferogram for the $16f$ state using (a) an ordinary laser frequency spectrum and (b) a notched frequency spectrum as discussed in the text. The labeled features correspond to energy differences between Rydberg states: (1) $15f$-$16f$, (2) $14f$-$15f$, and (3) $14f$-$16f$. When the $3d$-$15f$ transition cannot occur, the dominant frequency in the wave packet motion increases by approximately a factor of 2. The poor resolution in (b) is due to a short (10 ps) sample time.

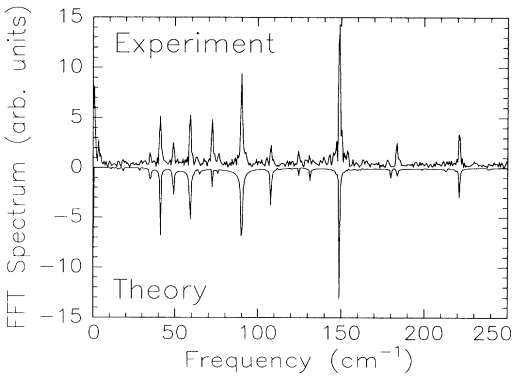

FIG. 3. Experimental and theoretical Fourier transform spectra corresponding to the population in the $18f$ level after the interaction of the $15f$ Rydberg eigenstate with two laser pulses at 2×10^{11} w/cm^2. The theoretical prediction is shown with negative amplitude. The laser intensity used in the calculation is within our experimental uncertainty of approximately a factor of 2.

ly determine which low angular momentum states are contributing to the spectrum due to the large quantum defects of these states. In fact we observe that only f states contribute to the redistribution in this experiment and to the wave packet formation described previously. Second, the dominant signal in our time-of-flight field ionized electron spectrum comes from the initial state. Saturation of the MCP detector prevents us from observing redistribution to lower n states which ionize at higher electric fields, and therefore, at later times. This same problem prevented Noordam *et al.* [5] from determining whether or not lower lying n states were populated in their experiment. Since our transform spectra show difference frequencies corresponding to all energy levels which are populated by the short pulses, we are able to determine that transitions to lower n states do occur in our redistribution process. Thus, time-domain atomic interferometry allows us to determine which states play a dominant role in the laser-atom interaction, even if the population in those states is not directly observable.

We use the same set of essential states in the numerical integration routine and in general the results of the calculation are in excellent agreement with the data as shown in Fig. 3. Again higher laser intensities correspond to complicated wave packet motion due to significant depletion of the initial Rydberg state [6]. As discussed by Noordam *et al.* [5] the dark wave packet manifests itself (at low intensities) as a hole or notch moving through the otherwise stationary initial state wave function. In our experiment, significant depletion of the initial state is possible at higher intensities, and no apparent stationary features remain. In fact, at high intensities the motion of the "regular" and "dark" wave packets do not appear drastically different from each other.

In conclusion, we have demonstrated that time-domain bound-state interferometry can be an extremely useful tool for studying the interaction of atoms with intense laser radiation. We have used this technique to observe intense field effects in the creation of ordinary and dark Rydberg radial wave packets. The interferometric method has proven to be effective in characterizing the motion of a "tailored" wave packet which was created from a laser pulse with a masked frequency spectrum. Furthermore, it has given additional insight into which angular momentum states participate in dark wave packet formation as well as in demonstrating the presence of energy eigenstates in the wave packet whose population is not readily detectable. We believe that the ability of the interferometric Ramsey technique to identify states which are populated during the interaction of an atom with an intense laser field without any direct observation of population in these levels can greatly benefit studies of atoms in intense laser fields.

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