Relative Phase Measurement of a Stark Wave Packet in the Vicinity of the Saddle Point

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A gas of Rb atoms in a static electric field has been photoexcited to just above the classical ionization threshold by a phase-locked sequence of two far infrared pulses. A single laser pulse generates a series of ejected electron packets emerging at the saddle point of the potential; each of the ejected packets is characterized by a phase and a chirp. We calculate and measure these phases and chirps using the time dependent interference of the electronic wave function controlled by the delay between the two light pulses.

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The dynamics of Rydberg wave packets [1] in atomic systems have been studied for more than a decade using optical pump-probe techniques. In the most common form of the experiment, the optical Ramsey method [2], the total photoexcitation yield due to a sequence of two identical pulses is monitored as a function of the delay time between the two pulses. This experimental arrangement probes the return of the wave packet to the ground state, and the Fourier transform of the signal is simply the absorption spectrum [2]. The Ramsey interference technique has been used to study complicated systems (e.g., Ref. [3]). More recently, new techniques have been developed which enable researchers to probe other regions of the extended Rydberg orbit. The atomic streak camera measures the time dependence of the outgoing electron flux [4,5], while THz momentum retrieval methods yield the momentum distribution of the wave packet as it travels through its orbit [6,7]. The *phase* of the Rydberg wave packet at the core is easily measured using the optical Ramsey method. Recently, the phase of the wave packet has been calculated at all points in space using measurements of the phase at the core [8]. However, to our knowledge, there have been no reports of the direct measurement of the phase of the Rydberg wave function at other positions along its orbit.

In this Letter, we describe a method by which the relative phase of a wave packet in a static electric field (Stark wave packet) is measured in the vicinity of the saddle point. To give an idea of how we accomplish this, Fig. 1 shows a schematic graph of the electron probability leaving the atom near the saddle point of the potential after the wave packet has been created by the pump laser pulse. The energy of the electron is well above the saddle point energy, thus whenever the wave packet reaches the saddle point, it can escape: ionization occurs. Once ionized the properties of the sequence of ejected packets (e.g., width, spacing, etc.) is preserved as the electron is accelerated by the static electric field from the region near the saddle point to macroscopic distances where the electron flux is measured [9]. Typically, a single laser pulse generates a sequence of electron pulses. The first electron pulse is

from the part of the packet directly ejected from the atom after the laser excitation; the next pulse is from the part of the packet excited relatively up-potential into the closed region of phase space returning to the interaction region near the nucleus, elastically scattering from the core electrons into the down-potential direction, and then being ejected; the succeeding peaks are from the second and third return of the packet to the nucleus. The information obtained from experiments using cw lasers [10] can be used to obtain $\langle \Psi(0) | \Psi(t) \rangle$ but not the real time ejection of the electron.

For the simplified case shown in Fig. 1, each of the packets has a simple peaked shape and each of the packets has a phase and a chirp (variation of frequency with time) associated with it. The phase and chirp arise from the motion of the packet in the closed channel [11]; thus we expect the phase and chirp of each succeeding pulse to accumulate
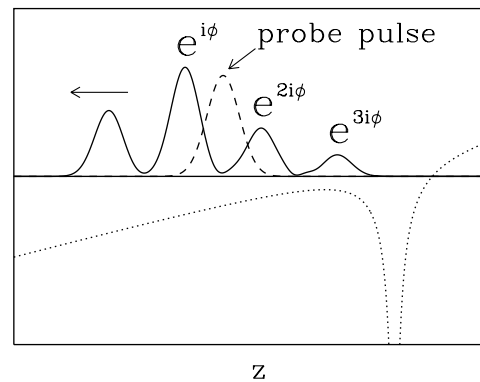

FIG. 1. Schematic drawing of the potential on the field axis (dotted line) for an alkali atom in a static electric field as a function of distance (*i*) along the field. Also sketched is the electron probability (solid line) roughly 50 ps following excitation by a single laser pulse. For physical reasons discussed in the text, simplified arguments lead to the expectation that the change in phase, ϕ, between successive electron pulses is constant. We also expect that the change in chirp between successive pulses is roughly constant. The dashed line represents an electron pulse from a probe source which will be used to interfere with the electron pulses generated by the pump laser pulse.

by roughly the same amount because the electron roughly repeats its motion after each period. The phase and chirp of each of the electron pulses can be measured through interference with a reference electron pulse if the time dependence of the ejected electron flux is measured and if the reference electron pulse is not highly chirped. In the experiment, the reference electron pulse is generated with a probe laser pulse that is phase locked to the pump laser pulse. Although the probe laser generates a train of reference pulses, the principle is unchanged from the schematic picture of Fig. 1. The outgoing flux at each moment in time arises from the coherent sum of the pump and of the probe temporal wave functions. The phase difference of these two amplitudes determines the outgoing probability. Hence by measuring the outgoing flux in a time-resolved manner, we obtain phase information on the wave packet as it is near the saddle point. Finally, shaped laser pulses have been used to control the products in chemical reactions [12] and the radial and angular momentum character of Rydberg states [13]; in this paper, we show that we can control when an electron is ejected from an atom by controlling the time delay between two laser pulses.

The time dependent measurements of the electron flux are performed with the atomic streak camera, which has been described in detail elsewhere [4]. A diffuse gas of Rb atoms are evaporated from an oven in a vacuum system (background pressure $\sim 10^{-6}$ Torr). These atoms drift into the interaction region situated between two metal plates upon which a voltage is applied. A frequency-doubled visible dye laser, operating at 622.70 nm, is used to prepare the gas of atoms into the $11p_{3/2}$ Rydberg state. Subsequently, midinfrared pulses from a free electron laser (FELIX [14], The Netherlands) are used to excite the Stark wave packets above the saddle point. The FEL is a continuously tunable laser with a wavelength range of 4.5–250 μm, which generates a pulse train of roughly 5000 pulses (called micropulses) of picosecond duration, with an interpulse spacing of 1 ns. For these experiments, the FEL is tuned to 7.37 μm, with a spectral bandwidth of 35 nm FWHM (6.8 cm^{-1}), corresponding to a pulse duration of 2 ps. Before being focused on the Rb ($11p$) atoms, the FEL beam is directed into a Michelson interferometer with a variable delay stage on one arm, to generate the optical pulse pair. The relative timing is such that the UV laser excites the gas of Rb atoms several hundred nanoseconds before the FEL macropulse arrives. Both lasers are polarized parallel to the static electric field. The emitted electrons, accelerated by the field, pass between a pair of deflection plates, upon which a fast sweep voltage is applied. The rapid electric field ramp converts the longitudinal profile of the electron ionization (i.e., the time axis) into a transverse profile. The resulting electron image is amplified by a multichannel plate and is then imaged by a phosphor screen and a CCD camera. For the phase-sensitive measurements described here, data are collected by employing a "synchroscan" technique [15].

The calculations are performed in the manner discussed in Refs. [5,9]. The continuum wave functions of the alkali atom in the field are superposed with the amplitude for excitation. The amplitudes depend on the dipole matrix elements connecting the initial to the final states and on the amplitudes of the frequency components of the laser field. The dipole matrix elements are calculated by numerical integration of the initial $11p$ and the final Rydberg wave function. Since the asymptotic form of the wave function is known, we can calculate the flux entering a detector that is a macroscopic distance from the atom.

The measured and calculated time dependent electron flux is shown in Fig. 2 when only the pump laser pulse of ~ 2 ps width excites the Stark wave packet. At a field strength of 1650 V/cm, the 7.37 μm infrared light excites the Rb atoms to an energy roughly 32 cm^{-1} above the classical saddle point. A series of emission peaks can be seen in Fig. 2, most of which occur long after the optical pulse is over.

The time separation of these electron emission peaks is roughly 14 ps, which we denote T. These peaks correspond to trajectories in which the electron escapes over the saddle point after several quasibound orbits. Previous work [4,5,9] has demonstrated that in a static electric field the probability of ionization is strongest when the electron has low angular momentum and is in the vicinity of the core. Strong emission peaks are therefore expected at the beat frequency between the radial oscillation frequency $(1/n^3)$ and the angular momentum oscillation frequency $(3Fn)$, where n is the principle quantum number and F is the electric field strength. For the conditions under which this experiment is performed, the angular momentum oscillation period is roughly 7–8 ps. Thus, since the emission peak separation is 14 ps, recurrences between the radial and the angular momentum oscillation frequency occur every second oscillation of the angular momentum, resulting in electron ejection.
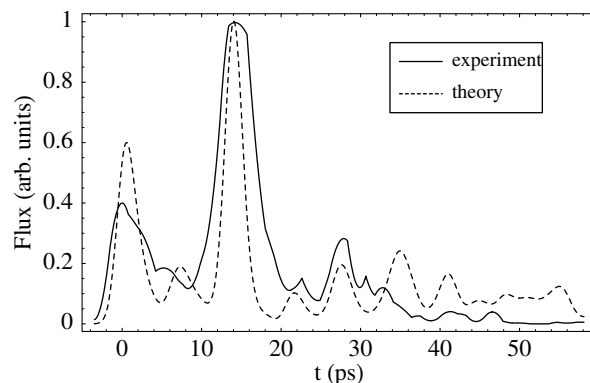

FIG. 2. Measured (solid line) and calculated (dotted line) time dependent electron emission from a single midinfrared (7.37 μm) laser pulse. The observed beating period (14 ps) matches twice period of the angular momentum precession in the static electric field.

Reference [11] discussed the theoretical aspects of the time dependent decay of a system excited by two, phase-locked laser pulses. The important point is that the wave function for the excited wave packet can be written as $\Psi(t) + e^{i\zeta}\Psi(t - \tau)$ where ζ is a phase change which varies from 0 to 2π over a change in time delay of one optical cycle. Since the periods of the electron wave packet (14 ps from Fig. 2) are *much* longer than the period of an optical cycle (25 fs), the phase can be treated as an independent parameter that can be varied in an experiment by changing the delay over one optical period.

In Fig. 3 we show how two laser pulses with control over the phase ζ allow us to measure properties of the phase of the outgoing electron pulses in Fig. 2. Figure 3 shows the measured and calculated time dependent electron flux as the phase of the probe laser pulse is varied. The time delay of the probe pulse is chosen to be 14 ps; thus, the first electron peak from the probe laser pulse is at the same time as the second peak from the pump laser, the second electron peak from the probe laser pulse is at the same time as the third peak from the pump laser, etc. The calculated and measured flux show a very striking pattern. The ejected electron flux from the probe laser pulse interferes in the same manner with all of the peaks from the pump laser pulse. The fact that the interference for all peaks is in phase implies that each of the successive peaks in Fig. 2 change phase as discussed for Fig. 1.

Much more information is available in this measurement than in the similar Ramsey interference technique. The relationship between the traditional Ramsey measurement and the technique presented here is analogous to the relationship in optics between an optical spectral measurement and a frequency resolved optical gating (FROG) measurement [16], whereby not only the amplitude but the phase of the optical field is measured. The usual Ramsey measurement is the time integral of the electron flux; thus a data point is an integral of Fig. 3 along t for a fixed delay between the laser pulses. The Ramsey fringe for the measurement in Fig. 3 has a contrast (ratio of maximum to minimum signal) of ~5.5; the contrast for the third electron peak from the pump laser ($t \sim 28$ ps) is 25.

In Fig. 4, we show the relative phase, φ, of the interference as a function of emission time. The relative phase is derived from the data in Fig. 3. For each t, we obtained the relative phase by performing a χ^2 fit of the ejected flux to $F_0(t) + F_1(t)\cos[2\pi t_d + \varphi(t)]$ where t_d is the time delay in optical cycles, F_1 is the amplitude of the interference and $\varphi(t)$ is the relative phase. The darkness of the symbol corresponds to the magnitude of the oscillation, F_1, with respect to change in delay of the second laser pulse; thus, dark symbols correspond to minimum uncertainty in the measurement. Both the calculated and measured relative phase are nearly independent of emission time which is reflected in the fact that all of the peaks constructively interfere at the same optical phase. This behavior is expected for this type of quantum system [11] but is a clear example of measuring the relative phase of the wave packet far from the core.
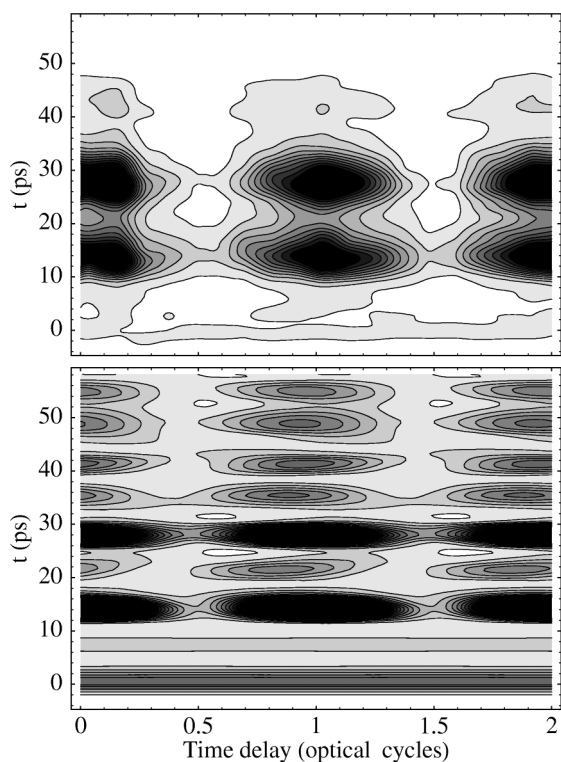

FIG. 3.   Measured (top) and calculated (bottom) ejected electron flux for two laser pulse excitations where the probe laser is delayed by ~14 ps. The flux depends on small changes in the time delay which is given on the x axis as a fraction of the optical cycle (25 fs).
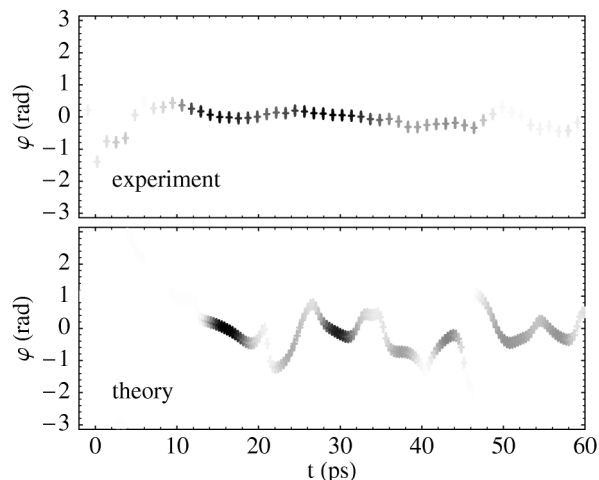

FIG. 4.   Measured (top) and calculated (bottom) relative phase of the interference for the case of Fig. 3. The phase at time t is derived from a horizontal cut of the intensity plot of Fig. 3. The darkness of the symbol corresponds to the magnitude of oscillation. This shows that the change in phase and chirp between successive ejected electron pulses is nearly constant as indicated in Fig. 1.
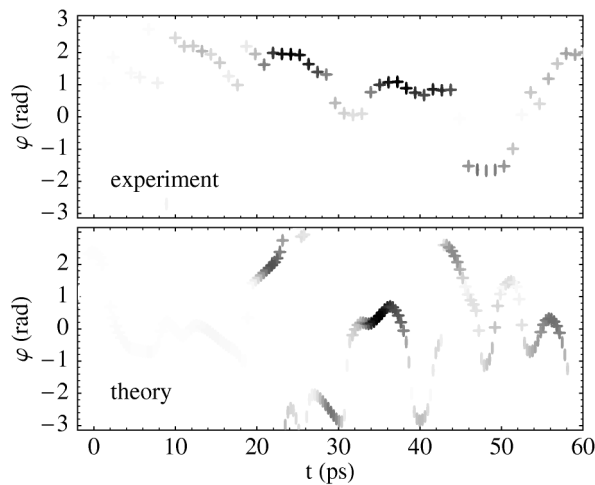
FIG. 5.   Same as Fig. 4 but now the probe laser is delayed by ∼22 ps so that the first electron pulse from the probe laser overlaps both the second and third electron pulses from the pump laser, and the second electron pulse from the probe laser overlaps both the third and fourth electron pulses from the pump laser, etc. Because a single electron pulse from the probe laser interferes with two electron pulses from the pump laser, the phase shows a series of steps with emission time that reflect the change in phase between successive electron pulses. Chirp of the electron pulses are visible as oscillations around the plateau of the step.

In Fig. 5, we show the changing phase of the successive packets when we delayed the probe laser pulse by ∼22 ps so that the first electron pulse from the probe laser overlaps both the second and third electron pulses from the pump laser, and the second electron pulse from the probe laser overlaps both the third and fourth electron pulse from the pump laser, etc. This is similar to the case sketched in Fig. 1. Clearly Fig. 5 is more complicated than Fig. 4 because the change in phase and chirp with successive electron packets now has a nontrivial effect. The main points of comparison are the strong interferences near 22 and 36 ps. The main feature is the change in phase of the main interferences at 22 and 36 ps. From the data of Fig. 5, we can estimate that the phase change between successive peaks is roughly 1 rad for the experiment and 1.5 rad for the theory. This demonstrates that the phase is not constant during an individual emission peak.

For the simplest case, we expect a sequence of flat steps in the relative phase. To take an example, the first electron pulse from the second laser overlaps the second electron pulse from the pump laser giving interference with a relative phase that is different from the interference with the third electron pulse from the pump laser. The difference in relative phase is the change in phase between the second and third electron pulse in Fig. 2. In Fig. 5 such steps are indeed observed but they are not flat. This is due to the chirp of the electron pulses. In other words the phase is not constant during an individual peak in the outgoing flux. The calculation shows more chirp than in the experiment, but the main features, where the interference is large and therefore the uncertainty is small, are in agreement.

In conclusion, we have made a measurement of the relative phase and chirp of several electron pulses in the vicinity of the saddle point of an atom in a static electric field, using the powerful capabilities of the new synchroscan streak camera. These experiments represent the first direct measurement of this phase at any point in the atomic orbit other than the core, and thus this technique provides a valuable new observable for the dynamics of wave packet evolution. An appealing variation of the scheme introduced in this Letter is to use a similar optical pump-probe pulse pair and replace the streak camera detection by THz ionization. It has been demonstrated that ionization by a half cycle THz pulse reveals the momentum distribution of wave packet [6]. In that way the relative phase of the different momentum components of the wave packet can be measured.

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