## **Revival structure of Stark wave packets**

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The revival structure of Stark wave packets is considered. These wave packets have energies depending on two quantum numbers and are characterized by two sets of classical periods and revival times. The additional time scales result in revival structures different from those of free Rydberg wave packets. We show that Stark wave packets can exhibit fractional revivals. We also show that these wave packets exhibit particular features unique to the Stark effect. For instance, the wave functions can be separated into distinct sums over even and odd values of the principal quantum number. These even and odd superpositions interfere in different ways, resulting in unexpected periodicities in the interferograms of Stark wave packets. [S1050-2947(97)02501-8]

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A free Rydberg wave packet initially follows the motion of a charged particle in a Coulomb field. However, after several cycles it collapses and a cycle of full and fractional revivals and superrevivals commences [1-6]. This behavior holds not only for hydrogenic wave packets but also for wave packets in alkali-metal atoms [7], where the energies have quantum defects causing shifts in the classical period and in the revival and superrevival times. Theoretical descriptions of these wave packets as squeezed states are known [8].

In this paper we examine the revival structure of Stark wave packets, which evolve in the presence of a static electric field. We prove below that under certain conditions Stark wave packets can exhibit full and fractional revivals. Moreover, we show the existence of new wave-packet behavior that does not occur for free wave packets and that is experimentally accessible.

To create a Stark wave packet, an atom is first placed in a static electric field that splits and shifts the energy levels. A short laser pulse is then applied in the presence of the electric field, resulting in a coherent superposition of Stark levels. For a hydrogen atom in a small electric field, the energies in atomic units are  $E_{nk} = -(1/2n^2) + 3nkF/2$ , where *n* is the principal quantum number,  $k = n_1 - n_2$  with  $n_1$  and  $n_2$  being parabolic quantum numbers, and *F* is the magnitude of the electric-field strength.

Stark wave packets have been produced and studied experimentally. The production of wave packets consisting of a superposition of k states in one Stark manifold with a fixed value of n is described in Ref. [9]. The oscillation of these parabolic wave packets corresponds to an oscillation of the eccentricity of the orbit. The dynamics of Stark wave packets above the classical field-ionization threshold  $F_c$  is examined in Refs. [10,11]. Simultaneous quantum beats in both the radial motion and angular motion are observed.

Stark wave packets with long lifetimes can be created by forming combinations of states below the classical fieldionization threshold. Superpositions of *k* states with n=23-25 have recently been produced in cesium [12]. Although the Stark spectra for alkali-metal atoms show strong avoided crossings, experiments indicate the behavior of these Stark wave packets is similar to ones in hydrogen. This is because on average the energy spacings between states of an alkalimetal atom are similar to those in hydrogen, and it is these spacings that determine the motion of the wave packet.

We are interested in the revival structure of a Stark wave packet  $\Psi(t)$  formed as a coherent superposition of states  $\phi_{nk}$ with energies  $E_{nk}$ . This is an example of a quantum system with energies depending on two quantum numbers n and k[13]. We write  $\Psi(t) = \sum_{n,k} c_{nk} \phi_{nk} \exp[-iE_{nk}t]$ . Here, the quantum number k is even or odd according to whether n is odd or even. The two-unit jump of adjacent values of k for fixed n requires special handling in the treatment of fractional revivals and results in additional interference effects in the interferograms of Stark wave packets. We suppose that the superposition is weighted around central values  $\overline{n}$  and  $\overline{k}$ of the two quantum numbers. The energy can then be expanded in a Taylor series around  $E_{\overline{nk}}$ .

For definiteness, consider an expansion centered around the values  $n = \overline{n}$  and  $k = \overline{k} = 0$ , and take the quantum number *m* associated with the third component of the angular momentum to be zero. In this case, we introduce

$$T_{\rm cl}^{(n)} = \frac{2\pi}{(\partial E/\partial n)_{\bar{n},\bar{k}}} = 2\pi\bar{n}^3, \quad T_{\rm cl}^{(k)} = \frac{2\pi}{2(\partial E/\partial k)_{\bar{n},\bar{k}}} = \frac{2\pi}{3F\bar{n}},$$
(1)

$$t_{\text{rev}}^{(n)} = \frac{2\pi}{\frac{1}{2}(\partial^2 E/\partial n^2)_{\overline{n},\overline{k}}} = \frac{4\pi}{3} \overline{n}^4,$$
$$t_{\text{rev}}^{(nk)} = \frac{2\pi}{2(\partial^2 E/\partial n \partial k)_{\overline{n},\overline{k}}} = \frac{2\pi}{3F}.$$
(2)

There is no revival time  $t_{rev}^{(k)}$  associated with the quantum number k since  $\partial^2 E/\partial k^2 = 0$ . Note that the definitions for  $T_{cl}^{(k)}$  and  $t_{rev}^{(nk)}$  contain factors of 2 that compensate for the two-unit jumps of adjacent k values. Note also that the mixed-derivative term generates a time scale  $t_{rev}^{(nk)}$ , which we call the cross-revival time. Substituting these definitions into  $\Psi(t)$  and keeping terms to second order yields the expression

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$$\Psi(t) = \sum_{n,k} c_{nk} \phi_{nk} \exp\left[-2\pi i \left(\frac{(n-\bar{n})t}{T_{cl}^{(n)}} + \frac{kt}{2T_{cl}^{(k)}} + \frac{(n-\bar{n})^2 t}{t_{rev}^{(n)}} + \frac{(n-\bar{n})kt}{2t_{rev}^{(nk)}}\right)\right].$$
(3)

For small *t*, the first two terms of the time-dependent phase in Eq. (3) dominate. They represent beating between the two classical periods  $T_{cl}^{(n)}$  and  $T_{cl}^{(k)}$ . We call  $T_{cl}^{(n)}$  and  $T_{cl}^{(k)}$  commensurate if  $T_{cl}^{(n)} = aT_{cl}^{(k)}/b$ , where *a* and *b* are relatively prime integers. If this relation holds, the time evolution of  $\Psi(t)$  on short-time scales exhibits a period  $T_{cl}$  $= bT_{cl}^{(n)} = aT_{cl}^{(k)}$ .

For larger times, the revival time scales  $t_{rev}^{(n)}$  and  $t_{rev}^{(nk)}$  become relevant and modulate the initial behavior, causing the wave packet to spread and collapse. We find that the wave packet undergoes full revivals provided the revival times  $t_{rev}^{(n)}$  and  $t_{rev}^{(nk)}$  are commensurate and obey  $t_{rev}^{(n)} = rt_{rev}^{(nk)}/s$ , where *r* and *s* are relatively prime integers. If this relation is satisfied, then there exists a revival time  $t_{rev} = st_{rev}^{(n)} = rt_{rev}^{(nk)}$  at which both second-order terms in the phase are integer multiples of  $2\pi$ . Near  $t_{rev}$ , the phase is again controlled by the first-order terms, and the shape and motion of the wave packet resembles that of the initial wave packet, i.e., a full revival occurs.

The commensurability of the time scales depends on  $\overline{n}$  and F. Restricting F to below the classical field-ionization threshold  $F_c = 1/16\overline{n}^4$  places limits on the ratios a/b and r/s. We find a/b < 3/16 and r/s < 1/8. By tuning F, specific commensurabilities and different types of revival structure can be selected.

To illustrate some of the possibilities, consider two examples. Let one have a/b=2/13, corresponding to a periodicity  $T_{cl}=2T_{cl}^{(k)}$ , while the other has a/b=1/6, corresponding to  $T_{cl}=T_{cl}^{(k)}$ . In the first case, peaks in the autocorrelation function should appear every two cycles in the period  $T_{cl}^{(k)}$ , while in the second peaks should appear every cycle. Since  $a/b=3F\bar{n}^4$ , these two different types of commensurability can be obtained using two values of the field strength in the ratio 12/13.

Behavior of this type has been seen experimentally in Stark wave packets of cesium with  $\overline{n} \approx 24$  [12]. Two different commensurabilities were observed in measured interferograms, one with peaks every other  $T_{cl}^{(k)}$  cycle, and the other with peaks every cycle. The ratio of the two measured field strengths agrees with the ratio 12/13.

These features can be displayed in plots of the absolute square the autocorrelation function  $|A(t)|^2$ of  $=|\langle \Psi(0)|\Psi(t)\rangle|^2$  as a function of t. We consider two plots of  $|A(t)|^2$  for wave packets with values of F having the ratio 12/13. With  $\overline{n}=24$ , this requires using  $F \approx 794.8$  V/cm for a/b=2/13 and  $F \approx 861.0$  V/cm for a/b=1/6. The superposition of interest lies near  $\overline{n}=24$  with m=0 and k=0, so the sum in  $|A(t)|^2$  can be restricted to the three *n* values n=23, 24, and 25 with only the upper part of the n=23 manifold and the lower part of the n=25 manifold included. For the distribution in k, we choose a broad Gaussian centered on k=0 with  $\sigma_k=6$ . This distribution matches the shape of the weighting coefficients obtained in a short-pulse laser excita-



FIG. 1. The autocorrelation vs time in picoseconds for a Stark wave packet with  $\overline{n}=24$ . The electric-field strength is  $F \approx 794.8$  V/cm, corresponding to the ratio  $T_{\rm cl}^{(n)}/T_{\rm cl}^{(k)} \approx 2/13$ .

tion from a low-energy p state to a superposition of Stark states with s character. A similar distribution would also occur in a superposition of m=1 states in an excitation from a low-energy s state to a superposition of Stark states with p character.

Figure 1 displays  $|A(t)|^2$  as a function of t for the case where a/b=2/13. As expected, the periodicity equals  $2T_{cl}^{(k)}$ , with odd multiples suppressed. Figure 2 shows the plot for a/b=1/6. In this case, there are peaks every cycle with period  $T_{cl}^{(k)}$ . In both figures, there is an overall decrease in the size of the peaks as the time increases. This is caused by the revival times, which destroy the initial periodic motion. Our analysis here uses hydrogenic energies, and effects of core scattering to the continuum as would occur in a Stark wave packet for an alkali-metal atom are ignored. A more detailed theoretical treatment incorporating higher-order Stark effects and quantum defects could be performed analytically using the methods of Ref. [14], but this lies outside the scope of the present work.

For fractional revivals to form in Stark wave packets, the wave function  $\Psi(t)$  in Eq. (3) must be expressible as a sum of distinct subsidiary wave functions. This can only occur at times  $t=t_{\text{frac}}$  that are simultaneously irreducible rational fractions of the two revival times scales. We define

$$t_{\rm frac} = \frac{p_1}{q_1} t_{\rm rev}^{(n)} = \frac{p_{12}}{q_{12}} t_{\rm rev}^{(nk)} \,. \tag{4}$$



FIG. 2. The autocorrelation vs time in picoseconds for a Stark wave packet with  $\overline{n}=24$ . Here, the electric-field strength is  $F \approx 861.0$  V/cm, corresponding to the ratio  $T_{\rm cl}^{(n)}/T_{\rm cl}^{(k)} \approx 1/6$ .

Here, the pairs of integers  $(p_1,q_1)$  and  $(p_{12},q_{12})$  are relatively prime.

In the Appendix, we outline a proof that subsidiary waves form at the times  $t_{\text{frac}}$  and discuss some additional effects due to the parity of k and the extra factors of 2 in the phase. We also show that  $\Psi(t)$  in Eq. (3) can be written as a sum  $\Psi_{\text{odd}}(t) + \Psi_{\text{even}}(t)$  consisting of separate sums over odd and even values of n.

At the fractional revivals, each of the wave functions  $\Psi_{odd}(t)$  and  $\Psi_{even}(t)$  can be written as a sum of subsidiary waves  $\psi_{cl}^{(odd)}$  and  $\psi_{cl}^{(even)}$ , respectively, with arguments shifted relative to *t* by certain fractions of the corresponding periods. At the times  $t = t_{frac}$ , the result is

$$\Psi(t) = \sum_{s_1=0}^{l_1-1} \sum_{s_2=0}^{l_2-1} a_{s_1s_2}^{(\text{odd})} \psi_{\text{cl}}^{(\text{odd})} \left( t + \frac{s_1}{l_1} T_{\text{cl}}^{(n)}, t + \frac{s_2}{l_2} T_{\text{cl}}^{(k)} \right)$$
$$+ e^{-i\pi(p_{12}t_{\text{rev}}^{(nk)}/q_{12}T_{\text{cl}}^{(k)})} \sum_{s_1=0}^{l_1'-1} \sum_{s_2=0}^{l_2'-1} a_{s_1s_2}^{(\text{even})} \psi_{\text{cl}}^{(\text{even})}$$
$$\times \left( t + \frac{s_1}{l_1'} T_{\text{cl}}^{(n)}, t + \frac{s_2}{l_2'} T_{\text{cl}}^{(k)} \right).$$
(5)

The functions  $\psi_{cl}^{(odd)}$  and  $\psi_{cl}^{(even)}$ , the coefficients  $a_{s_1s_2}^{(odd)}$  and  $a_{s_1s_2}^{(even)}$ , and the integers  $l_1, l_2, l'_1$ , and  $l'_2$  are defined in the Appendix.

The functions  $\psi_{cl}^{(odd)}$  and  $\psi_{cl}^{(even)}$  are doubly periodic functions with periods  $T_{cl}^{(n)}$  and  $T_{cl}^{(k)}$ . The evolution of the wave packet exhibits the beating of these two classical periods. At the fractional revivals, the sums in Eq. (5) exhibit periodicities that are fractions of the time scales  $T_{cl}^{(n)}$  and  $T_{cl}^{(k)}$ . The behavior of the quantum number k causes the functions  $\psi_{cl}^{(odd)}$  and  $\psi_{cl}^{(even)}$  to obey

$$\psi_{\rm cl}^{(\rm odd)}(t + \frac{1}{2}T_{\rm cl}^{(n)}, t) = -\psi_{\rm cl}^{(\rm odd)}(t, t),$$
  
$$\psi_{\rm cl}^{(\rm even)}(t + \frac{1}{2}T_{\rm cl}^{(n)}, t) = \psi_{\rm cl}^{(\rm even)}(t, t).$$
(6)

This additional dependence on  $T_{cl}^{(n)}/2$  causes the unconventional revival structure of Stark wave packets.

As an illustrative example, consider the case  $\overline{n}=24$ , and set  $t_{rev}^{(n)}/t_{rev}^{(nk)} = r/s = 1/12$  by tuning the electric-field strength to  $F \approx 645.8$  V/cm. For this example,  $t_{rev} = t_{rev}^{(nk)} = 12t_{rev}^{(n)}$ . Using the expressions in the Appendix, for  $t \approx t_{rev}$  we find  $\Psi(t) \approx \psi_{cl}^{(odd)}(t,t) + \psi_{cl}^{(even)}(t,t)$ . These sums are in phase and combine as a single total wave packet, producing the full revival at  $t_{rev}$ .

At  $t = t_{rev}/2$ , however, we find a time phase between  $\Psi_{odd}(t)$  and  $\Psi_{even}(t)$ , with the full wave function reducing to

$$\Psi(t) \approx \psi_{\rm cl}^{(\rm odd)}(t, t + \frac{1}{2}T_{\rm cl}^{(k)}) + \psi_{\rm cl}^{(\rm even)}(t + \frac{1}{4}T_{\rm cl}^{(n)}, t).$$
(7)

We see that this fractional revival consists of two subsidiary wave functions out of phase with each other.

Figure 3 shows the absolute square of the autocorrelation function as a function of time. Here,  $t_{rev} \approx 403.4$  psec,  $T_{cl}^{(n)} \approx 2.1$  psec, and  $T_{cl}^{(k)} \approx 16.8$  psec. Since  $T_{cl}^{(k)}$  is an integer multiple of  $T_{cl}^{(n)}$ , we expect peaks at times equal to multiples of  $T_{cl}^{(k)}$ . These are apparent in Fig. 3. The full revival is



FIG. 3. The revival structure of Stark wave packets is displayed in a plot of the autocorrelation function as a function of the time in picoseconds. The Stark wave packet has  $\overline{n}=24$ . The electric-field strength is  $F \simeq 645.8$  V/cm, which sets  $t_{rev}^{(n)}/t_{rev}^{(nk)} \simeq \frac{1}{12}$  and  $t_{cl}^{(n)}/T_{cl}^{(k)} \simeq \frac{1}{8}$ .

evident and has the anticipated periodicity. The fractional revival near  $t = t_{rev}/2$  has peaks corresponding to those from two wave packets half a classical period  $T_{cl}^{(k)}$  out of phase, in agreement with our predictions.

For the odd-*n* superposition in the Stark wave packet, additional interference occurs at the  $t = t_{rev}/2$  revival. The additional interference is caused by the antiperiodic behavior of  $\psi_{cl}^{(odd)}$  and can be seen explicitly from the form of the autocorrelation function. The subsidiary wave functions  $\psi_{cl}^{(odd)}$  and  $\psi_{cl}^{(even)}$  are orthogonal since they consist of separate sums over odd and even *k*. We can therefore calculate  $A(t) = \langle \Psi(0) | \Psi(t) \rangle$  using Eq. (7) for times  $t \approx t_{rev}/2$ , disregarding the cross terms. This gives

$$A(t) = \langle \psi_{cl}^{(odd)}(0,0) | \psi_{cl}^{(odd)}(t,t+\frac{1}{2}T_{cl}^{(k)}) \rangle + \langle \psi_{cl}^{(even)}(0,0) | \psi_{cl}^{(even)}(t+\frac{1}{4}T_{cl}^{(n)},t) \rangle.$$
(8)

Suppose the functions  $\psi_{cl}^{(odd)}$  and  $\psi_{cl}^{(even)}$  are spatially localized. Then, at times that are multiples of  $T_{cl}^{(k)}/2$ , A(t) reduces to the first term since the second term vanishes. Conversely, at multiples of  $T_{cl}^{(k)}$ , A(t) reduces to the second term. Since  $\psi_{cl}^{(odd)}$  is antiperiodic in the first time argument with period  $T_{cl}^{(n)}/2 \approx 1.05$  psec, we expect nodes in the autocorrelation function occurring with this periodicity at times that are multiples of  $T_{cl}^{(k)}/2$ . However, since  $\psi_{cl}^{(even)}$  is periodic, nodes need not appear in A(t) at multiples of  $T_{cl}^{(k)}$ .

Figure 4 shows an enlargement of the autocorrelation



FIG. 4. An enlargement of Fig. 3 in the vicinity of the fractional revival at  $t_{rev}/2$ .

function near the fractional revival at  $t_{rev}/2$ . Alternate peaks have different interference patterns, as expected. The peaks at multiples of  $T_{cl}^{(k)} \approx 16.8$  psec are single with no interference. These arise from  $\psi_{cl}^{(even)}$  in Eq. (8). The peaks at multiples of  $T_{cl}^{(k)}/2$  arise from the  $\psi_{cl}^{(odd)}$  terms in A(t). Nodes in A(t) with the periodicity  $T_{cl}^{(n)}/2 \approx 1.05$  psec are apparent.

The experiment described in Ref. [12] observed Stark wave packets for delay times of about 150 psec, after which the signal was lost due to dephasing. To detect the fractional revival at  $t_{rev}/2$  described in this paper, the alignment of the interferometer would need to be maintained for delay times of at least 200 psec. Observation of the full revival predicted would require a delay time of 400 psec. Delay times greater than these have already been achieved in studies of Rydberg wave packets in the absence of external fields. Note that our treatment has disregarded core scattering and fine structure. The importance of these effects could therefore be determined in part by comparison of experiments to our predictions for the fractional and full revivals.

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## APPENDIX

This appendix proves that fractional revivals occur in Stark wave packets. First, rewrite  $\Psi(t)$  in Eq. (3) by shifting  $(n-\overline{n}) \rightarrow n$  and separating the series into odd and even sums over *n*. This gives  $\Psi(t) = \Psi_{odd}(t) + \Psi_{even}(t)$ . Then, let  $k \rightarrow 2k$ in the sum over odd *n*, and  $k \rightarrow 2k+1$  in the sum over even *n*. We then define the doubly periodic wave functions  $\psi_{cl}^{(odd)}$ and  $\psi_{cl}^{(even)}$ ,

$$\psi_{\rm cl}^{(\rm odd, even)}(t_1, t_2) = \sum_{n \text{ odd, even}} \sum_k c_{nk} \phi_{nk}$$
$$\times \exp\left[-2\pi i \left(\frac{nt_1}{T_{\rm cl}^{(n)}} + \frac{kt_2}{T_{\rm cl}^{(k)}}\right)\right]. \tag{A1}$$

The higher-order terms in the time-dependent phases of  $\Psi_{\text{odd}}(t)$  and  $\Psi_{\text{even}}(t)$  at  $t = t_{\text{frac}}$  are given by

$$\theta_{nk}^{(\text{odd})} = \frac{p_1}{q_1} n^2 - \frac{r}{s} \frac{p_1}{q_1} nk, \qquad (A2)$$

$$\theta_{nk}^{(\text{even})} = \frac{p_1}{q_1} n^2 - \frac{r}{s} \frac{p_1}{q_1} nk - \frac{r}{s} \frac{p_1}{q_1} \frac{1}{2} n.$$
(A3)

Here, *n* is odd in Eq. (A2) and even in Eq. (A3). We seek the minimum periods  $l_1$ ,  $l_2$ ,  $l'_1$ , and  $l'_2$  such that  $\theta_{n+l_1,k}^{(odd)} = \theta_{nk}^{(odd)}$ ,  $\theta_{n,k+l_2}^{(odd)} = \theta_{nk}^{(even)}$ ,  $\theta_{n+l'_1,k}^{(even)} = \theta_{nk}^{(even)}$ . These relations yield four conditions for the periods  $l_1$ ,  $l_2$ ,  $l'_1$ , and  $l'_2$  in terms of *n*, *k*, and  $t_{\text{frac}}$ . Since the functions  $\psi_{cl}^{(odd)}$  and  $\psi_{cl}^{(even)}$  with *t* shifted by appropriate fractions of  $T_{cl}^{(n)}$  and  $T_{cl}^{(k)}$  have the same periodicities in *n* and *k* as  $\theta_{nk}^{(odd)}$  and  $\theta_{nk}^{(even)}$ , respectively, we may use these functions as a basis for an expansion of the wave functions  $\Psi_{odd}(t)$  and  $\Psi_{even}(t)$  at the times  $t_{\text{frac}}$ . The result is the expansion (5), where the coefficients  $a_{s_1s_2}^{(odd)}$  and  $a_{s_1s_2}^{(even)}$  are given by

$$a_{s_{1}s_{2}}^{(\text{odd})} = \frac{1}{l_{1}l_{2}} \sum_{\kappa_{1}=0}^{l_{1}-1} \sum_{\kappa_{2}=0}^{l_{2}-1} \exp(2\pi i \,\theta_{\kappa_{1}\kappa_{2}}^{(\text{odd})}) \\ \times \exp\left(2\pi i \,\frac{s_{1}}{l_{1}} \,\kappa_{1}\right) \exp\left(2\pi i \,\frac{s_{2}}{l_{2}} \,\kappa_{2}\right), \quad (A4)$$

with a similar expression for  $a_{s_1s_2}^{(\text{even})}$  in terms of  $l'_1$  and  $l'_2$ .

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