Nondispersing Wave Packets

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We have formed nondispersing wave packets from atomic Li Rydberg eigenstates by adding a small microwave field approximately resonant with the $\Delta n = 1$ transition. The orbital motion of the Rydberg electron in states of $70 \leq n \leq 78$ becomes phase locked to a 17.5 GHz microwave field and remains so for 900 ns, roughly 15 000 orbits of the Rydberg electron. A Floquet approach provides a reasonable quantum-mechanical description.

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To show that the classical picture of a particle moving in a potential could be represented by the stationary eigenstates of quantum mechanics, Schrödinger constructed wave packets, coherent superpositions of energy eigenstates [1]. When properly constructed, they have localized peaks in the probability of finding the particle that move as would a classical particle in the same potential. Wave packets remained theoretical objects until the advent of mode-locked lasers, although quantum beats, which are few level wave packets, were observed long ago [2]. With mode-locked lasers it has been possible to create vibrational wave packets in molecules and electronic wave packets in atoms [3–5]. In the first Rydberg-atom wave-packet experiments a ps laser was used to excite atoms from the ground state, forming electronic wave packets that were superpositions of n states of the same ℓ. Here n and ℓ are the principal and orbital angular momentum quantum numbers of the Rydberg electron. Initially, just after the laser pulse, these wave packets are localized near the ionic core, and they then oscillate between small and large orbital radii just as would a classical electron in a highly eccentric orbit. However, the localization of the wave packet persisted for only a few orbits because of the dispersion in the energy level spacings [6]. Schrödinger used the harmonic oscillator to construct his wave packet, and the delocalizing effect of dispersion in any other potential was noted coincidentally by Lorentz [7].

Rydberg atoms are attractive systems for wave packets because the dispersion is small. The energies of the np states, for example, are given by $W_n = -1/2(n - \delta_p)^2$ where δ_p is the quantum defect of the np series, and the $\Delta n = 1$ spacing between levels is $\Delta W = W_{n+1} - W_n \cong 1/n^{*3}$, where $n^* = n - \delta_p$. We use atomic units unless units are expressed explicitly. For high n, ΔW is almost, but not quite, constant. Since the orbital period is given by $T \cong 2\pi n^{*3}$ the dispersion in energy level spacings implies that the orbital periods of the higher n states in the wave packet are slightly longer than those of the lower n states in the wave packet, and thus the initial spatial localization of the wave packet disappears after a few orbits. Since there is a finite number of states in the wave packet, typically about five, at a later time, the revival time, the wave-packet rephases such that there is again a localized wave packet, and the observation of several revivals has been reported [8–10]. However, even in the best of cases only tens of orbits were observed before the localization disappeared, presumably due to technical dephasing.

The question of how to construct nondispersing Rydberg wave packets has attracted considerable attention. ten Wolde *et al.* constructed wave packets of Stark states, which in hydrogen are equally spaced by $3nE$, where E is the electric field [11]. While they observed less dispersion than seen in radial wave packets at the time, it was still present, and the wave packets were short lived [11]. A different route to constructing nondispersing wave packets is to add a weak periodic perturbation that phase locks the motion of the Rydberg electron [12–18]. The first case to be discussed was the application of an electric field rotating in a plane at the orbital frequency [13]. A classically stable periodic orbit exists in which the electron's motion is circular in the plane of the field and phase locked to the field so that the force from the rotating field on the electron is always radially inward toward the ion core. Such wave packets have been termed Trojan wave packets because of the analogy to the Trojan asteroids of Jupiter orbiting in the Sun-Jupiter gravitational system [13]. Adding a magnetic field perpendicular to the plane of the orbit allows further control of the electron's orbit [14,15]. It has also been suggested that an electron can be trapped in a phase-locked circular orbit in which the force from the applied field is always radially outward, forming an anti-Trojan wave packet [16]. Particularly useful is the notion that such a nondispersing wave packet is a stationary Floquet eigenstate [17]. Finally, Kalinski *et al.* have suggested that nondispersing wave packets can be constructed in two electron planetary atoms in which the motion of the inner electron phase locks the motion of the outer electron [19].

A classically stable phase-locked orbit analogous to the one described above is predicted to occur in a linearly polarized oscillating field, which is monochromatic [18] or composed of trains of pulse [20]. The essential notion for a monochromatic field is shown in Fig. 1. If an

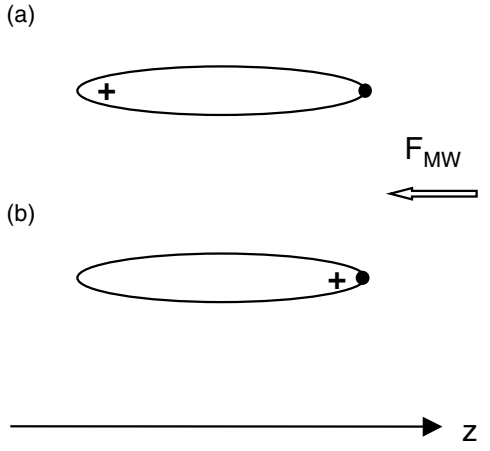

FIG. 1. The electron (●) in the two highly eccentric orbits about the ionic core (+) that become phase locked to the weak microwave field. The electron orbits are aligned along the z axis as is the microwave field. When the force of the field on the electron F_{MW} is at its maximum in the $-z$ direction the electron in the orbits (a) and (b) are at their outer and inner turning points, respectively.

electron is in a highly eccentric, almost one-dimensional orbit along the z axis it can be phase locked to a nearly synchronous z-polarized microwave field that forces the electron toward (away from) the ion core at the outer (inner) turning point of the orbit. As shown, there are equivalent stable orbits on either side of the atom. Here we report for the first time the use of a 17.5 GHz, linearly polarized microwave field to observe experimentally nondispersing wave packets of Li Rydberg atoms that live for more than 15 000 orbits.

In the experiment a beam of Li atoms passes in front of a microwave horn where the atoms are excited to Rydberg np states of $n \approx 70$ by three tunable 5 ns dye-laser pulses using the excitation sequence $2s \rightarrow 2p \rightarrow 3s \rightarrow np$. The atoms are then exposed to a 17.5 GHz microwave pulse that is turned on in 1 to 10 ns and is polarized in the z direction. The microwave field can convert the eigenstate into a wave packet in which the electron's motion becomes phase locked to the microwave field. To detect whether or not the electron's motion has become synchronous with the microwave field we expose the atoms to a sub-ps half-cycle field pulse (HCP), which is a time-resolved detector of the Rydberg electron's momentum [21]. The HCP is synchronized with the microwave field and polarized in the z direction. The HCP ionizes atoms in which the z component of the electron's momentum exceeds a value determined by the HCP amplitude, as will be discussed in more detail shortly. The HCP is followed by a 5–10 V/cm field pulse to drive the resulting ions to a microchannel-plate detector. The ion signal is captured by a gated integrator and recorded in a computer.

The HCP is generated by illuminating a biased GaAs wafer in the vacuum chamber with a 200 fs, 815 nm Ti:sapphire laser pulse, and it freely propagates to the point where the atoms are excited by the lasers [22]. The microwave oscillator is phase locked to the 230th harmonic of the 76 MHz signal from a fast photodiode that detects the output of the Ti:sapphire oscillator. The coarse timing of the HCP pulse relative to the microwave-pulse envelope is set electronically, and we make fine adjustments (0–240 ps) in the delay of the HCP by changing the optical path length of the 815 nm pulse illuminating the GaAs wafer. This adjustment changes the phase of the microwave field when the HCP reaches the atoms while the first does not. The timing uncertainty between the microwave field and the HCP is 5 ps.

The duration of the HCP is short compared to the orbit time of the electron, so it gives the electron an impulse $\Delta p\hat{z}$ which changes the energy of the Rydberg electron by $W_I = 2\Delta p\ p_0 + \Delta p^2$, where p_0 is the z component of the momentum of the electron just prior to the impulse from the HCP. If $W_I > 1/2n^{*2}$ the electron gains enough energy to escape from the atom.

Typically we set the amplitude of the HCP so as to ionize ~50% of the atoms in an np eigenstate. We thus ionize the half of the atoms in which the electron has a z component of velocity in the same direction as $\Delta p\hat{z}$. We then scan the fine time delay of the HCP to vary the phase of the microwave field at which it reaches the atoms. In Fig. 2 we show the ionization signals vs the fine time delay of the HCP pulse when we excite the Li $72p$ state, for which $T = 56$ ps, and use a 17.528 GHz (57 ps period)

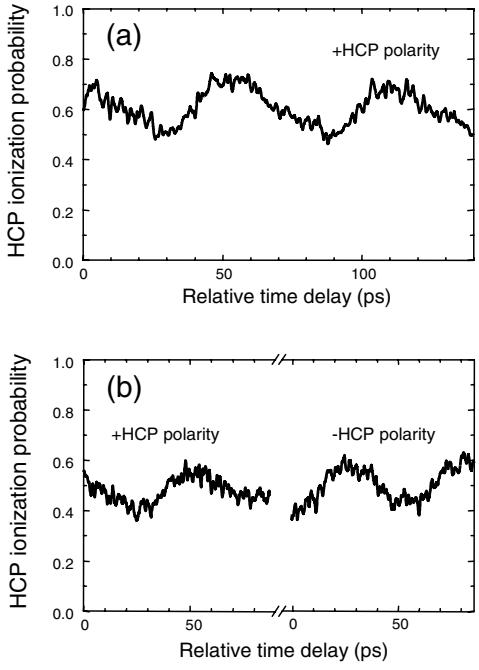

FIG. 2. Ionization probability vs fine time delay of the HCP 100 ns after the beginning of the 17.528 GHz, 1 V/cm microwave pulse: (a) HCP amplitude set to ionize ~60% of the atoms, (b) HCP with amplitude set to ionize ~50% of the atoms with the same polarity as (a) and reversed polarity. In all cases the modulation with period 56 ps indicates that the wave packet is phase locked to the microwave field. In (b) the phase of the signal reverses with HCP polarity as expected.

microwave pulse of amplitude $E = 1$ V/cm. For comparison, the atomic Coulomb field at the outer turning point of an $n = 72$ electron orbit is 48 V/cm. The coarse timing is set so the HCP reaches the atoms ~100 ns after the beginning of the microwave pulse. The signals obtained with two amplitudes and both polarities of the HCP are shown in Fig. 2. There is a clear variation in the ionization signal with the 57 ps period of the microwave field, showing that the electron's motion is, indeed, phase locked to the microwave field. Note that the modulation shown in Fig. 2 is as deep as that observed when using a HCP to detect radial wave packets produced by a fs laser [23]. When the polarity of the HCP is reversed, the sign of the modulation of the ion signal is reversed as well [Fig. 2(b)].

By changing the coarse delay of the HCP relative to the start of the microwave pulse, we have observed that the modulation in the ion signal grows from zero to the amplitude shown in Fig. 2 in ~2 ns and remains at this amplitude for at least up to ~900 ns. The modulation in the signal has the same phase relative to the microwave field for all coarse delay times. In Fig. 2(a) the times ~28 and ~56 ps, where the minimum and maximum ionization signals are observed, correspond to the highest and lowest electron momenta in the z direction. We infer that the wave packet is at the inner and outer turning points of the orbits of Fig. 1 at times ~42 and ~70 ps in Fig. 2. With the HCP polarized perpendicular to the microwave field, the modulation in the ionization signal is reduced by a factor of 10 from that shown in Fig. 2, indicating that the changes in momentum of the Rydberg electron are largely confined to one dimension and that a one-dimensional model should contain most of the physics [18,24].

Only states for which the $\Delta n = 1$ transitions are nearly resonant with the microwave frequency ω can become part of the wave packet. The sharpness of this resonance is shown in Fig. 3, a plot of the amplitudes of the signals obtained when exciting different np states with the laser and exposing them to a 1 V/cm, 17.528 GHz microwave field. Somewhat surprising is the double-peaked structure, with peaks at $n = 72$ and 76. It persists when the field amplitude E is reduced, although the signals become smaller. At higher E the plots analogous to Fig. 3 cover a broader range of n, with multiple, smaller peaks. The small negative signals of Fig. 3 denote signals with a negative amplitude, or a 180° phase reversal. When the microwave frequency is changed, the peaks of Fig. 3 move to different n, as expected. For example, with a microwave frequency ω of 16.850 GHz the peaks in the structure analogous to those shown in Fig. 3 move from $n = 72$ and 76 to $n = 73$ and 77.

Viewing the wave packets in quantum-mechanical terms provides further insight. We can easily estimate the expected half-width of the resonance peak shown in Fig. 3 by equating the $\Delta n = 1$ Rabi frequency, $\sim 0.3n^2E$, to the detuning due to dispersion, $(3n^{-4})|\Delta n|$. For a $E =$ 1 V/cm and $n = 72$ this leads to a half-width of $|\Delta n| = 2.7$, in reasonable agreement with the data shown in Fig. 3. Using a one-dimensional atom, which gives a reasonable representation of ionization in linearly polarized microwave fields [24], we have found the Floquet eigenvalues and eigenvectors [25]. The Floquet energies vs 17.528 GHz microwave-field amplitude E are shown in Fig. 4. Each of the Floquet eigenstates is periodic, and thus nondispersing, although they have varying amounts of moving spatial localization. The highest-energy state, labeled A, corresponds most closely to the classical motion of Fig. 1. States that shift down in energy correspond to

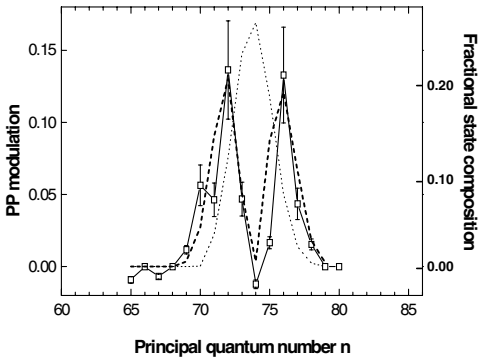

FIG. 3. Peak-to-peak amplitude of the detected wave-packet ion signals vs initial n state for a 17.528 GHz, 1 V/cm microwave field (□). The solid line simply connects the points. Fractional n-state composition of the highest energy Floquet state, A (dotted line) and the adjacent Floquet state, B (dashed line) calculated with a 4 V/cm microwave field of the same frequency, but shifted in n by 1 as described in the text. The states A and B are labeled in Fig. 4.

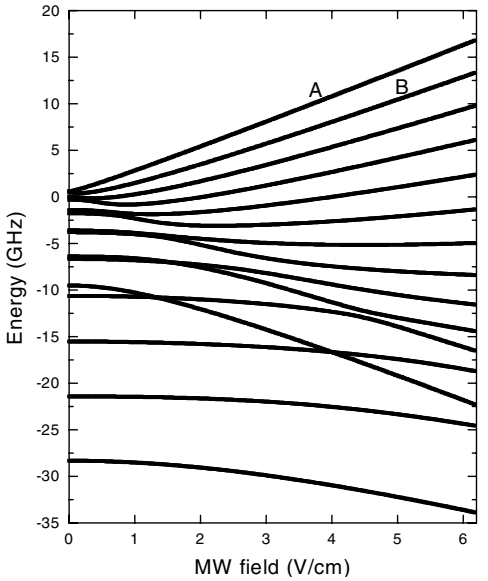

FIG. 4. Floquet energy vs field for a one-dimensional atom in a 17.528 GHz field. The energies are plotted relative to the energy of the $n = 72$ state. The zero-field Floquet energy of state n is shifted from its normal energy by $(72 - n) \times$ 17.528 GHz.

classical motion 180° out of phase, and the lowest energy states, which exhibit minimal field dependence, resemble conventional stationary eigenstates at low fields. How efficiently a Floquet state can be prepared depends upon the excitation scheme, and to explore this issue we have excited the atoms to the Li np states during, as well as before, the microwave pulse of $E = 1$ V/cm. There is no discernible difference between the signals in the two cases, so we focus on the well-defined case in which we excite the atoms in the microwave field. The laser bandwidth is ~6 GHz and the pulse is ~5 ns long. Consequently, we can expect to excite an incoherent superposition of the Floquet eigenstates shown in Fig. 4. We presume that we are exciting primarily the ones that have positive energy shifts in the microwave field, for their spatial oscillations will be approximately at the phase shown in Fig. 1.

In Fig. 3 we show the calculated fractional composition of the two highest-energy Floquet eigenstates of Fig. 4 in terms of the initial n eigenstates for a 4 V/cm, 17.528 GHz microwave field. Specifically, we show the squared projections from the initial n state onto these Floquet states. In plotting the squared projections we have shifted them down in n by 1 to match the experimental observations. This shift is comparable to the shift of resonances observed in microwave ionization of Li relative to H [26,27] and arises from the finite size of the Li^+ ionic core. As shown, while the width of the calculated curves matches the width of the experimental resonance width, the double-peaked structure does not. While our data resemble the projection of the np states onto the Floquet state labeled B in Fig. 4, the agreement is probably fortuitous, as the one-dimensional model does not account for the fact that the Li eigenstates have a definite parity and that the Li np oscillator-strength distribution in the presence of a field is not hydrogenic [28].

In conclusion, we have observed nondispersing wave packets that are formed by phase locking the Rydberg electron's motion to a small, near-resonant microwave field that suppresses wave-packet spreading due to both dispersion and technical dephasing. These long-lived wave packets are evidently the source of the classical resonant stabilization in microwave ionization [24,26,27], and they may be useful in applications impractical with short-lived wave packets or as a means of trapping Rydberg atoms [29].

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