Excitation of a Three-Dimensionally Localized Atomic Electron Wave Packet

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(Received 9 June 1999)

We have prepared an atomic electron in a three-dimensionally localized wave packet that travels along a highly eccentric orbit between the nucleus and a turning point approximately 1000 Å away. State preparation consisted of two distinct steps. First the electron wave function was localized to the desired orbit by exciting a stationary extreme Stark state from the $n = 30$ manifold of a sodium atom in a dc electric field. Then the dynamic wave packet was formed by applying a terahertz electromagnetic half-cycle pulse polarized along the direction of the dc field. The localized state is an ideal starting point for further quantum control proceeding along classical orbits.

PACS numbers: 32.60.+i, 32.80.Rm, 42.50.Ct

One of the primary goals from the earliest days of atomic electron wave packet research has been to produce a three-dimensionally localized wave packet moving along a classical orbit [1]. There have been a number of beautiful experiments producing various types of electron wave packets with localization along either the radial or angular coordinate [2]. There have also been experiments in two electron wave packets [3]. While there have been a large number of theoretical studies of the properties of fully three-dimensionally localized wave packets [4], and proposals for how this wave packet could be created [5,6], to our knowledge no one has reported the actual creation of this wave packet. In this paper we describe an experiment in which we created such a wave packet and carried out a series of experiments to verify that the desired wave packet was produced.

While the creation of this wave packet is important for studies of the classical limit of a highly excited atom, it is perhaps more important because it offers a launching point for further research in quantum control. The quantum mechanical electron has two complementary descriptions: in terms of waves, or as an ensemble of particles. Previous techniques for quantum control have dwelt on the wave properties of the quantum system [7]. Here we emphasize the classical ensemble description.

It has been shown that Rydberg wave packet states are sufficiently near to the classical limit in their properties that an ensemble of particles following classical dynamics provides a very accurate description, so long as the ensemble does not shear all the way around the orbit allowing quantum interference to occur [8]. Such descriptions show the utility of studying ensembles of classical particles for developing an intuition for wave packet evolution. In this regime the problem of quantum control reduces to the problem of controlling an ensemble of classical particles. If one wishes to control a classical ensemble of particles, moving them from their initial locations to some well-defined final location, the problem is made much more difficult if the initial ensemble is too spread out in phase space. In the spread wave packet various members of the ensemble will react differently to any control pulse depending on their individual initial conditions. An efficient strategy is first to localize the ensemble, and then to move the localized ensemble along a common path to the desired destination.

The technique that we used to make the three-dimensionally localized wave packets was first presented by Gaeta *et al.* [5] and consists of two distinct steps. First one excites a particular Rydberg state that has a stationary wave function localized along the desired elliptical wave packet trajectory [9]. Then one uses a short electromagnetic half-cycle pulse (HCP) polarized along the orbital direction to further localize the wave function into a wave packet moving along the elliptical orbit. Previous experiments have used HCPs [10] to excite dynamic wave packets from initial, low-angular-momentum, energy eigenstates [11]. However, because such initial states do not define a unique elliptic trajectory, the resulting wave packets are not well localized and do not follow an elliptical orbit.

The proposed technique allows the production of wave packets moving along elliptical orbits of arbitrary eccentricity, ranging from a circle to a line. The wave packet that we chose to excite has a trajectory in the shape of the ellipse with the maximum eccentricity: a line on one side of the atom's core. Thus the first step was to excite a stationary state that was localized along this line. For hydrogenlike atoms, such states are eigenstates of the atom in a dc electric field that have the maximum Stark shift. For a given n manifold (and for fields lower than the field at which states from neighboring manifolds cross), there are two such states. The highest energy state for the manifold is called the *blue* state, while the lowest is called the *red* state [12]. Note that, for a Stark field in the $+z$ direction, the blue state is confined to a line on the $+z$ side of the core, while the red state is on the $-z$ side.

The physical arguments that led to this excitation technique are based on classical simulations of the interaction of a hydrogen atom and HCP. In order to verify the efficacy of the technique for a real sodium atom we carried

out a full quantum simulation of our actual experiment. Our quantum calculation consisted of integrating the time-dependent Schrödinger equation in an essential state basis of 1400 states, using sodium dipole moments and our measurement of the HCP electric field [13]. They show that, for an initial *red* Stark state, the resulting wave packet consists primarily of the initial state and *red* Stark states from neighboring manifolds in a roughly Gaussian distribution in energy. For $\bar{n} = 30$, the orbital period T_K is 4.1 ps, and the wave packet orbits along on a line on one side of the core. As the wave packet oscillates, it disperses and revives due to the anharmonicity of the Coulomb potential. This wave packet is illustrated in Fig. 1. At a time 2.0 T_K after the HCP, the uncertainty product $\Delta z\, \Delta p_z$ is only twice the minimum allowed by Heisenberg's uncertainty principle.

Our experiment consists of illuminating a sodium atomic beam with optical pulses and HCPs, and detecting ions as a function of the delay between pump and probe HCPs. Figure 2 shows the geometry of the interaction region formed by three copper plates, each measuring 10 cm $\times$ 10 cm. The beams intersect between the bottom two plates which are separated by 1 cm. We apply a voltage across these plates to provide the dc Stark field, as well as a pulsed collection field that pushes ions towards our detector, an electron multiplier.

The first step of the excitation process is to excite the red Stark state in sodium from the $n = 30$ manifold, for Stark fields less than the 75 V/cm field at which states from neighboring manifolds cross. To accomplish this we use a pair of nanosecond-pulse, tunable dye lasers, one tuned to the $3s$-$3p$ resonance (589 nm) and the other tuned to take population from the $3p$ state to the final Rydberg Stark state (411 nm). The next step of the excitation uses a HCP to coherently redistribute population from the initial state to other red Stark states in neighboring manifolds. We generate HCPs inside the vacuum chamber using standard large-aperture photoconductive antennas fabricated from a 25 mm-square piece of semi-insulating GaAs that is biased with up to 11 kV [14]. The emitters are triggered using a 130 fs pulse from a Ti:sapphire regenerative amplifier that uniformly illuminates the surface of the emitter with a fluence of 40 μJ/cm^2.

Measurements of HCPs [13,14] show that their electric field consists of a short positive peak followed by a longer, low-amplitude tail. The width of the main peak at half maximum is ~400 fs, whereas the tail extends for more than 3 ps. As explained by diffraction theory [15], such pulses must have no dc component and thus have *zero area:* the areas of the positive peak and negative tail cancel. It has been previously shown that zero-area pulses cannot redistribute population from an initial state if they are much shorter than the state's Kepler period [5]. Our numerical simulations show, however, that because the tail of our pulse extends for almost a full Kepler period, it does not significantly alter the redistribution produced by the short positive peak.

We sample the motion of the wave packet using a second, delayed HCP to ionize the wave packet. After this probe HCP, a 50 V, 1 μs pulse is applied to the bottom plate to push any ions created by the HCP through a slit in the middle plate and on towards the detector (an electron multiplier). The field-free region between the middle and upper plates (which were separated by 4 cm) serves to delay the arrival of the ions at the detector so that our ion signal is not obscured by noise from the HCP emitters. As there is an 8° angle between the pump and probe HCP beams, we use a 1 mm wide slit that bisects the HCP beams to ensure that the pump-probe delay for atoms below the slit varies by no more than 460 fs. We find zero delay by ionizing the wave packet using two HCPs with electric fields that point in opposite directions but are equal in magnitude. At zero delay, the pulses cancel each other, producing a minimum in the ionization signal.

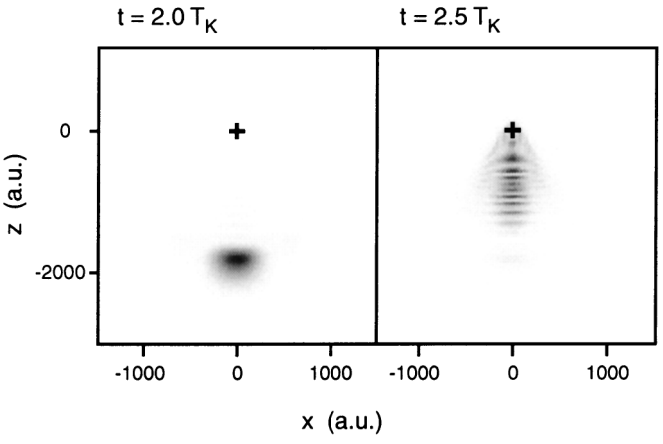

FIG. 1. Linear-orbit wave packet at the outer and inner turning points of its orbit. The plots show the probability distribution projected onto the x-z plane at two times following the HCP. The Kepler period, T_K, is 4.1 ps for $\bar{n} = 30$.

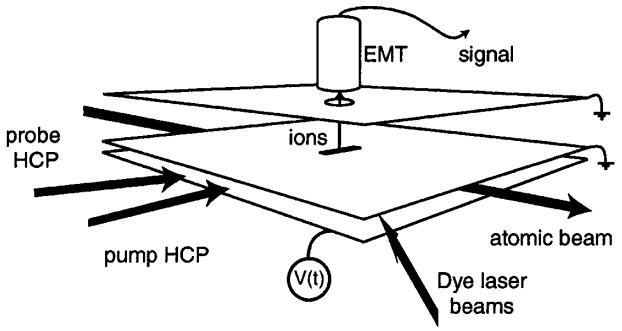

FIG. 2. Schematic of the interaction region inside the vacuum chamber. The sodium atomic beam passes between the lower plates where it intersects the dye laser and HCP beams. The voltage $V(t)$, applied across the bottom and middle plates, provides the dc Stark field as well as a pulsed field that pushes ions through the slit towards the electron multiplier tube (EMT).

We use the probe HCP to time resolve the motion of the wave packet in the following way. As shown previously, a HCP ionizes the wave packet by giving it a kick [16]. The energy absorbed by the atom depends on the direction of motion of the wave packet during the probe pulse. For a classical electron moving with velocity $\mathbf{v}(t)$, the change in energy ΔW is given by

$$\Delta W = \int_{-\infty}^{\infty} \mathbf{F}(t') \cdot \mathbf{v}(t')\, dt', \tag{1}$$

where the HCP electric field $\mathbf{E}_{\mathrm{HCP}}$ exerts a force on the electron of $\mathbf{F}(t) = -e\,\mathbf{E}_{\mathrm{HCP}}(t)$. Thus the ionization signal, as a function of delay between the pump and probe HCPs, shows the motion of the wave packet.

Our simulations show that the wave packet follows a linear orbit along a line on one side of the core, as defined by the initial Stark state. One could expect that this asymmetry could lead to variations in the pump-probe signal as the relative orientation of the trajectory and probe pulse were changed. To test this, we measured the signal for two probe field directions. We changed the direction of the probe field by simply changing the polarity of the bias voltage applied to the HCP emitter. Note that we did not change the way that we made the wave packet.

Figure 3 shows our pump-probe data for the following two cases. In both cases the pump pulse formed the wave packet by kicking the atom's electron in the $-z$ direction. There are a number of interesting features displayed in the data, some of which are the subject of continuing research. We can, however, explain all the qualitative features observed. When the pump and probe pulses are in the same direction (top figure), the signal at zero delay is a maximum. This maximum occurs because the pump and probe pulses add constructively, increasing the ionization probability of the initial Stark state. If, however, they are in opposite directions (bottom figure), the pump pulse reduces the amplitude of the probe pulse to produce a signal that is initially at a minimum. Furthermore, if the probe pulse kicks the electron in the $-z$ direction (top figure), the signal shows strong modulation at the Kepler period. But if the probe pulse kicks the electron in the $+z$ direction (bottom figure), after two Kepler periods the modulation is greatly reduced. In addition, the signal level is lower by almost a factor of 4.

One can explain the reduction in signal level by taking into account the motion of the wave packet during the ionizing probe pulse. When the probe pulse kicks the wave packet in the $-z$ direction, the wave packet is most likely ionized if it is moving away from the core in the $-z$ direction, along the direction of the kick. During the probe pulse a wave packet moving in this direction will continue to do so, and, as shown by Eq. (1), the atom will absorb energy throughout the pulse. In a similar way, if the kick is in the $+z$ direction, the wave packet is most likely ionized when moving towards the core, in the $+z$ direction. In this case, however, there is a chance during the pulse that the wave packet can pass the point at which it is closest to the core (perihelion) and change direction to travel in the $-z$ direction. Thus the remainder of the probe pulse actually lowers the energy of the wave packet, reducing the probability of ionization. Effects of this kind were seen in HCP ionization of oriented, *stationary* states [17]. Here we are seeing the effect of motion of the wave packet during the pulse. This effect also produces a reduction of modulation when the probe pulse kicks the wave packet towards the core.

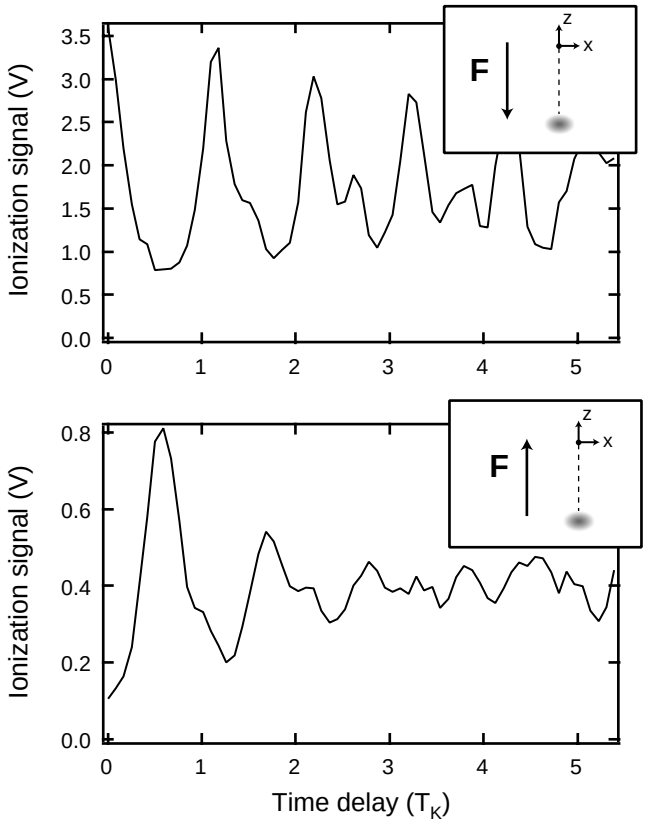

FIG. 3. Linear-orbit wave packet signal. The time delay between the pump and probe pulses is in units of the 4.1 ps Kepler period. The pump HCP had a 1 kV peak field in the $+z$ direction. The probe pulse had a 3 kV peak field in either the $+z$ or $-z$ direction. The insets show the relative orientation of the wave packet (when at its outer turning point) and the kick **F** it receives from the probe HCP. Note that when the probe kicks the wave packet away from the core the signal is larger and shows increased modulation.

An interesting signature of a linear-orbit wave packet is the effect the Stark field has on its orbital period. If the Stark field is applied in the $+z$ direction, it raises the potential on the $+z$ side of the core, and lowers it on the $-z$ side. A wave packet moving along the $-z$ side of the core can travel farther in this distorted potential before it reaches the classical outer turning point (aphelion). Thus its orbital period is increased slightly, about 2% for a 40 V/cm field. One can calculate the period of a

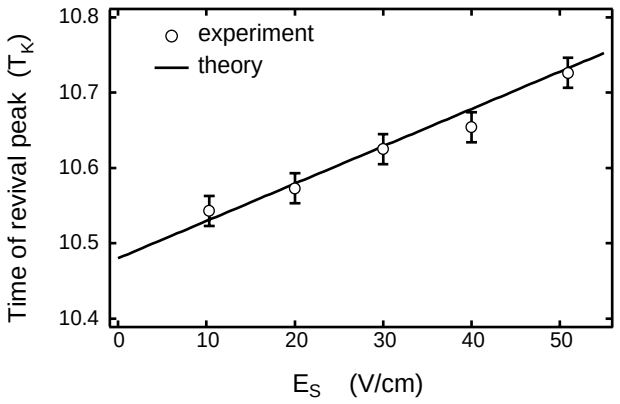

FIG. 4. Measurements showing the effect of the Stark field on the wave packet motion. The time of the revival peaks is plotted versus the Stark field, E_S. The points are experimental data, with the error bars indicating the uncertainty in determining the location of the revival peak. The line is calculated using theory presented in the text.

linear-orbit wave packet formed from red Stark states from the separation of the states as a function of the Stark field, E_S. The change in the period (ΔT) given a change in the Stark field (ΔE_S) is, in atomic units,

$$\Delta T \simeq 3\bar{n}^4 \Delta E_S \,. \tag{2}$$

To see this change we waited until the wave packet revival, some 10–11 Kepler periods after the wave packet's formation. Figure 4 shows the times of the peaks in the pump-probe signal in the vicinity of the revival for a range of Stark fields. Also shown is the prediction derived using Eq. (2), namely, $T_{\rm rev}(E_S) = T_{\rm rev}(0)\{1 + 3\bar{n}^4 \Delta E_S\}$, which agrees well with our experimental observations.

This work was supported by the U.S. Army Research Office through Contract No. DAAH04-96-1-0284 and through the MURI Rochester Center for Quantum Information, by the National Science Foundation under Contracts No. ECS-94-13989 and No. PHY 94-15538. We would also like to acknowledge useful conversations with Z. Gaeta, M. Noel, J. West, and J. Corless. J. B. acknowledges support from the Laboratory for Laser Energetics of the University of Rochester.

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