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Atomic electron wave packets in an electrical field

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We have observed the periodic motion of a parabolic electron wave packet. The wave packet was created by coherent excitation with 7-ps laser pulses of several parabolic states of atomic rubidium in a dc electrical field. The dispersion of the wave packet was small because the spacing between subsequent energy levels of the Stark map is almost constant. By measuring the photoionization yield in a pump-probe experiment, as many as 10 oscillations were observed.

There has been an increasing interest in atomic wave packets during the last few years. Wave packets are quantum beat phenomena, and they describe the evolution of the system in space and time. Recently, we reported the observation of radially localized electron wave packets.[1,2] Also the observation of wave packets localized in the angular coordinates was reported.[3] As a result of the new experimental possibilities, there is a renewed interest in the theory.[4-6] The concept of a wave packet, which is as old as quantum mechanics itself,[7] forms a bridge between the quantum-mechanical stationary wave functions and the classical notion of a localized, moving particle. In the case of a radial wave packet, the localization is rapidly lost because of the varying spacing between the energy levels. This spreading of the wave packet can be avoided by choosing a system where the energy levels have a constant spacing, like a harmonic oscillator. In this Rapid Communication we report the observation of wave packets which are formed by a coherent superposition of parabolic states. For an atom in a dc electrical field, the energy levels are split and shifted by the Stark effect. The spacing between the energy levels in one n manifold is almost constant. This leads to a wave packet with little dispersion, so many oscillations could be observed.

To get more insight in the time evolution of parabolic wave packets, we will briefly discuss the case of a hydrogen atom in an external dc electrical field. For small field strengths, the Stark shift is linear and the energy levels are given by $E_{nk} = -1/2n^2 + \frac{3}{2}Fnk$, where n is the principal quantum number, F the electrical field strength, and $k = n_1 - n_2$, with n_1 and n_2 the parabolic quantum numbers (atomic units are used throughout this paper). The spacing between the parabolic energy levels is given by $\Delta E = 3Fn$, leading to a beating period

$$\tau = \frac{2\pi}{3Fn}, \tag{1}$$

in the case of coherent excitation. This excitation can be performed by applying a laser pulse with a duration τ_p shorter than this period. In that case, a parabolic wave packet is created, which exhibits oscillating behavior with period τ. This period is the same for all neighboring k components of the wave packet, and therefore dispersion is absent.

The nature of the oscillation is easier to understand if we write the wave function as a coherent superposition of l states rather than k states. Only certain l states are populated from the ground state as a result of the selection rule $\Delta_l = \pm 1$ for each absorbed photon. The electrical field F mixes all l states, so the populations of the different l states will vary in time. In order to calculate these time-dependent populations, we project the l distribution at $t = 0$ on the k basis. The time evolution of the wave packet described in this basis is trivial, because k is a good quantum number and the energy levels are known. The population of each l state at time t is calculated by projecting wave packet at time t back from the k states on the l states. The results of such a calculation are shown in Fig. 1. The calculation is done for $n = 23$ and $F = 35$ V/cm, leading to an oscillation period τ of 324 ps (the electrical field strength F is chosen smaller than in the experiments in order to provide a clear example). The exciting pulse is Gaussian and has a duration of 7 ps. Assumed is that after the two-photon excitation from the ground state, only the d state is populated. The evolution shown starts at the end of the pump pulse when some mixing to other low l states has already taken place. We see that the distribution spreads over higher l states until it is localized near the highest one, $l = 22$. This happens at $t = 162$ ps, which is equal to $\tau/2$. In the second half of the oscillation, the reverse development takes place, until the original distribution reappears at $t = \tau$. The coherent excitation of parabolic states leads to a quantum beat in the popula-

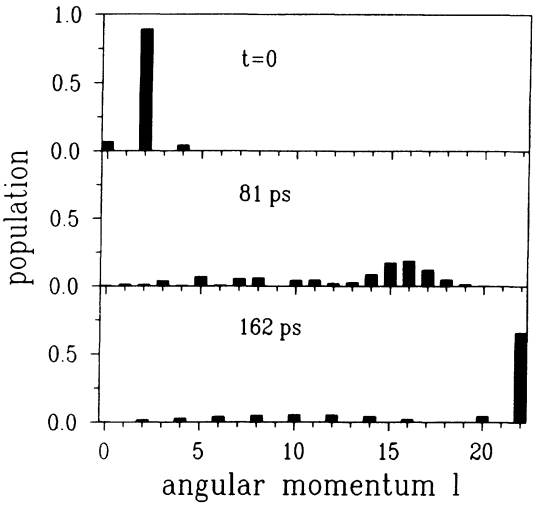

FIG. 1. The population of the different l states at four different times ($t=0$, 81, and 162 ps). The calculation is carried out with $n=23$, $F=35$ V/cm, and a Gaussian pulse of 7 ps. At $t=0$, only $l=2$ is populated, apart from some l mixing during the pump pulse. The population starts to mix and spreads out over higher l states, until it is almost completely localized in the highest one, $l=22$, at $t=162$ ps. This moment is half of the oscillation period τ.

tions of the l states.

From this elementary model, which does not incorporate the effects of higher-order Stark shifts, saturation, quantum defects, and spin-orbit coupling, we can already draw several conclusions. First, we can see how the wave packet evolves in space. Since the eigenfunctions in the l basis can be written as a product of a radial part $R_{nl}(r)$ and an angular part $Y_{lm}(\theta,\phi)$, we can study the evolution of these parts separately. The radial part of the wave function is located between the classical turning points, given by

$$r_{1,2}=n^2\mp n[n^2-l(l+1)]^{1/2}. \quad (2)$$

For $l\ll n$, the classical turning points approach the values $r_1\approx\frac{1}{2}l(l+1)$ and $r_2\approx 2n^2$ so that the wave function is spread out between the core and $2n^2$. The classical analogue is a highly eccentric elliptic orbit, with the electron passing close to the atomic core. In contrast, if l approaches $n-1$, both classical turning points approach the value of n^2. Now the wave function is located in a small region around $r=n^2$, like it is in a classical circular orbit. The atomic wave packet is oscillating between these two extremes as a result of the beating l populations. At the same time, the angular part of the wave packet is oscillating between a more or less spherical distribution for low l states and a distribution which is strongly oriented along the z axis for l close to $n-1$ (for the case that $m=0$). The fact that the angular momentum l is not a conserved quantity in a dc electrical field, corresponds classically to the changing eccentricity and orientation of the elliptical orbit. In this respect it should be noted that localization is only present in the parabolic coordinates, and not in r, θ, and ϕ.

The periodic motion of the wave packet will be reflected in photoionization. The one-photon ionization probability of a Rydberg state decreases rapidly with increasing l, as a result of the decreasing wave-function overlap.[8] The physical reason for this is that absorption of a visible photon can only take place near the atomic core.[5] For high l states, the electron is always far away from the core and, consequently, it has a low ionization probability. At the beginning of each period, only low l states are populated, leading to a considerable ionization signal. At half of the period, only the highest l state is appreciably populated and no ionization takes place. By measuring the photoionization signal in a pump-probe experiment as a function of the delay between the exciting pump and the ionizing probe pulse, the period of the oscillating wave packet shows up as peaks in the ionization yield at delay 0, τ, $2\tau,\ldots$.

The experiment is performed in the following way (see also Ref. 2). The output of a synchronously pumped dye laser (5-ps pulse duration, wavelength around 600 nm) is amplified in a three-stage amplifier. The dye amplifier is pumped by the second harmonic of a Q-switched Nd:YAG laser (20 Hz). The pulse duration of the 500-μJ amplified pulses was measured to be 7 ps. Each pulse is divided by a beamsplitter, reflected on a mirror, and sent back to the beam splitter. The interference fringes between the two pulses continue over $\sim$4 ps, indicating that the bandwidth of the pulses lies close to the Fourier transform limit. One of the mirrors is mounted on a delay line in order to vary the time difference between the two pulses. After this Michelson setup, the beam is led into a vacuum vessel with a background pressure of 2×10^{-7} mbar. The beam is focused by an $f=16$-cm lens, enters a metal box, and crosses the rubidium vapor, which comes out of an orifice in a tube. With this focal length of 16 cm, a pulse energy of less than 1 μJ, and a beam diameter of 2 mm, we calculate a maximum laser intensity in the focus of 10^9 W/cm^2. This means that the ac Stark shift is negligible. The laser focus lies between two condenser plates that create a homogeneous dc electrical field. The polarization of the light is chosen parallel to the electrical field, leading to the selection rule $\Delta m_l=0$. In the upper plate (the anode), the center is replaced by a wire grid through which all the photoelectrons are accelerated in the direction of the detector. The detector consists of a set of channel plates. The output is amplified, monitored on an oscilloscope, averaged over 500 laser shots, and sent to a computer. In order to minimize the detection of slow electrons, which could have been created after the pulse sequence, we use a time gate of about 30 ns for the signal. In practice, the number of measured slow electrons arising from long time-scale processes is negligible. With this experimental setup, the (2+1)-photoionization signal of Rb atoms in an electrical field can be measured as a function of the delay between the pump and probe pulse.

The excitation process from the ground state of Rb is performed by the absorption of two photons, so only the s and the d states are populated from the ground state. From spectra obtained with ns pulses, it appears that the population of the s state is negligible. In order to populate the manifold, the d state has to be mixed with it. A com-

plicating factor is the rather large quantum defect of the d state of Rb, $\delta = 1.347$. As a result of this, the d state lies at $\frac{1}{3}$ between two succeeding manifolds in the zero-field situation. In order to mix the population from the d state to the nearest manifold, the electrical field strength must be larger than half of the n mixing value ($F_n \approx 1/3n^5$). Therefore, F is chosen between $\frac{1}{2}F_n$ and F_n, while the central wavelength is resonant with the center of the n manifold ($E = -1/2n^2$). By tuning the wavelength to different n states, different manifolds can be excited leading to different oscillation periods.

We will present two typical experimental results for different n manifolds. In Fig. 2, the photoionization signal is plotted against the delay between the pump and the probe pulse. The signal due to the direct three-photon ionization of both pulses separately was measured, and has been subtracted from the total ionization yield. In the case of Fig. 2(a), the $n = 23$ manifold was excited with a central wavelength of 597.3 nm, while the applied field strength F was 247.5 V/cm ($F_n = 266$ V/cm). We see periodic peaks in the ionization signal at delay values $\Delta t = 46$, 87, and 132 ps. This is in good agreement with the theory: Eq. (1) predicts an oscillation period of 45.8 ps. Around delay zero, there is a large peak in the ionization yield. This is the result of the so-called coherent spike in the light intensity, due to interference between the two beams of the Michelson interferometer. The width of all peaks is approximately equal to the pulse duration 7 ps, and does not increase yet. In another experiment [Fig. 2(b)], the $n = 19$ manifold was excited with a wavelength of 599.2 nm, while the applied field was 645 V/cm ($F_n = 692$ V/cm). The peaks in the ionization yield lie at $\Delta t = 22, 41, 62, \ldots, 204$ ps. This is, again, in good agreement with the predicted oscillation period of 21.3 ps.

The most important result of Fig. 2 is the observation of as many as ten oscillations of the parabolic electron wave packet. The dispersion of this wave packet is much smaller than for a radial wave packet, where we observed only two oscillations.[2] Still, the dispersion is clearly not equal to zero. In the case of Rb, there are two effects that could easily lead to dispersion.[9] First, the s and p states have large quantum defects. They mix with the manifolds, and deform it by avoided crossings. The spacing between the levels will not be perfectly constant, and this leads to dispersion of the wave packet. Second, the fine structure can, in principle, not be neglected and may slightly change the position of the energy levels. However, this second effect is expected to be small around $k = 0$ for the used field strengths. In Fig. 2(b) the ionization signal is decreasing for longer delays. This decrease of the ionization signal during the scan (starting at delay zero) is due to the decreasing pulse energy, which was monitored with a photodiode. On a single-shot basis, the signal could be corrected for differences in the laser intensity I, because in a three-photon process the ionization yield is proportional to I^3. However, in our experiment, both the ionization yield and the measured light intensity were averaged over 500 laser shots. This leads to an ambiguity in the power of I in the normalization procedure. Therefore, the results are given without correcting for light intensity differences.

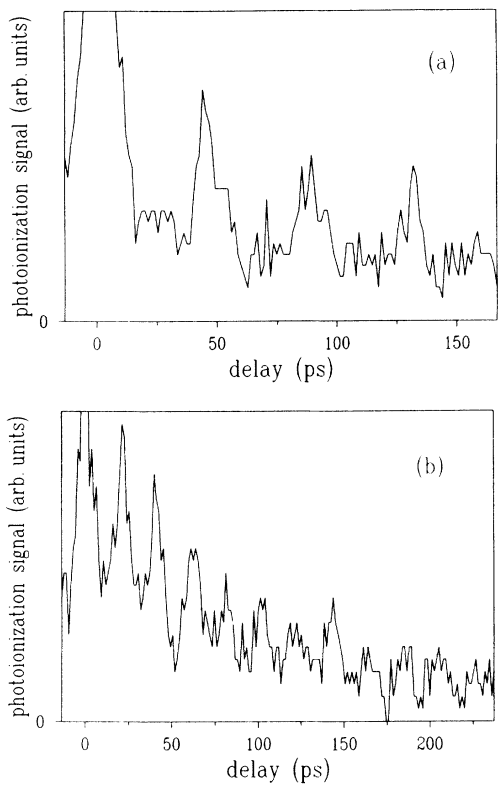

FIG. 2. The pump-probe ionization signal as a function of the time difference between the two pulses, for two different experimental situations. Around zero delay, the coherent spike in the light intensity gives rise to a very high ionization rate. The periodic peaks in the ionization signal for increasing delay reflect the oscillation of the parabolic wave packet. In (a), $n = 23$ is excited, while $F = 247.5$ V/cm, leading to a period of 45.8 ps. In (b), $n = 19$ is excited, while $F = 645$ V/cm, leading to a period of 21.3 ps; 10 oscillations are observed.

In conclusion, a parabolic electron wave packet has been created with ps laser pulses. The superposition of the k states leads to a beat in the angular momentum. The periodic evolution of the wave packet manifests itself in photoionization because only states with low l can be efficiently ionized. The spacing between k states is almost constant, leading to a wave packet with little dispersion.

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