Delayed Electron Emission in the Ionization of Rydberg Atoms with Half-Cycle THz Pulses

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(Received 20 March 1997)

Electron emission of cesium Rydberg ($n \approx 21$) atoms ionized with half-cycle THz pulses is investigated using an atomic streak camera. The THz pulses have a peak electric field of 17 kV/cm and a duration of less than 1 ps. It is found that the atoms do not always emit their electrons instantaneously. Under certain conditions up to 50% of the emitted electrons are ejected 11 ps after the time of interaction. The delayed electron emission shows a strong dependence on the direction in which the electrons are kicked, and is interpreted as scattering off the atomic core. [S0031-9007(97)03703-4]

PACS numbers: 32.80.Rm, 42.50.Hz

In recent years it has become possible to generate extremely short electrical pulses with peak field of up to 100 kV/cm [1,2]. The duration of the pulses (0.5–1 ps) is short compared to the time it takes a highly excited atomic Rydberg electron to orbit the nucleus. The pulse corresponds to a half cycle of electromagnetic radiation with a frequency spectrum extending to 2 THz. In the short pulse limit, $\tau_{\text{pulse}} < \tau_n$, with $\tau_n = 2\pi n^3$ the classical round trip time of the Rydberg electron, the interaction can best be described as a kick. The electron hardly moves during the pulse. Under this assumption the transferred momentum Δp to the electron is given by the time-integrated electric field $\vec{F}(t)$ [2,3] (atomic units are used throughout),

$$\Delta \vec{p} = -\int_{-\infty}^{\infty} dt\, \vec{F}(t)\,. \tag{1}$$

Strictly this integral is zero for a freely propagating electromagnetic pulse, and the short unipolar pulse is followed by a very long, weak tail of opposite electric field [1]. This tail typically lasts several tens of picoseconds and has an amplitude of 1% or less of the main peak. However, ionization of the atom takes place on the time scale of the short pulse, and it is assumed that in the description of the ionization the long weak tail can be neglected, so that the effective change in momentum is indeed nonzero. The change in momentum leads to an energy change

$$\Delta E = \frac{1}{2}[2\vec{p}_0 \cdot \Delta \vec{p} + (\Delta p)^2]. \tag{2}$$

In general the energy transfer to the electron depends on its initial momentum $\vec{p}_0$ at the time of the kick and therefore varies over the orbit. This feature becomes apparent when a Rydberg wave packet is ionized, in which case the ionization probability oscillates in time with a period corresponding to the Kepler orbit time [4].

The direction of the kick not only influences the amount of transferred energy. If the kick is large compared to the original momentum p_0, then the electron moves in the direction of the kick after the pulse is gone. With the half-cycle pulse (HCP) a Rydberg state becomes the perfect starting point for a well-controlled electron-ion collision. Both the impact parameter and the energy of the collision can be chosen by varying the direction and the strength of the kick that is imparted on the electron. Until recently the analysis of such collisions consisted of recording the integrated ionization yield after illumination with an HCP. However, if scattering off the ionic core occurs before ionization takes place this is likely to give rise to delayed emission of the electrons.

In this Letter time-resolved data on the ionization of Rydberg states with THz half-cycle pulses are reported. The time-dependent emission is recorded with an atomic streak camera, which has a picosecond time resolution. We find that the recorded time-dependent emission shows a high sensitivity on the exact direction of the kick.

A detailed description of the atomic streak camera can be found elsewhere [5]. Here we outline its operation. The cesium atoms are excited to a Rydberg state ($n \approx 21$) with a nanosecond dye laser between two field plates separated by 1 cm, which are at a potential of -4.0 and -2.0 kV, respectively (see Fig. 1). After excitation to a particular Rydberg state, the atoms are irradiated with the picosecond THz pulse. Emitted electrons are accelerated (to the right in Fig. 1), and pass through a 1 cm long, 300 μm wide slit in the field plate. The actual operation of the streak camera takes place between the two deflection plates that the electrons pass through after they have been accelerated. The electric field between these two plates is ramped with a few V/ps. Consequently, the electric field experienced by the electrons depends on the time at which they pass between the plates. Electrons that arrive at later times are deflected more by this field. The difference in deflection angle is converted into a vertical displacement when the electrons reach the multichannel plate detector. At this detector each electron is converted with high efficiency into a cloud of electrons that leads to a measurable signal on the phosphor screen placed behind the multichannel plates. The light from this phosphor screen is recorded with a CCD camera and read by a computer. The time resolution obtained with this streak camera is approximately 1 ps.

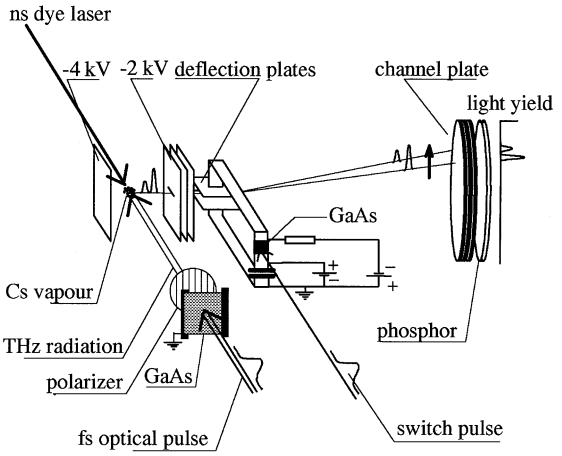

FIG. 1. Principle of the atomic streak camera. Operation of the camera is discussed in the text.

The subpicosecond THz pulses are generated in a way similar to the technique described by Jones *et al.* [2]. The optical switch pulse comes from a colliding pulse mode-locked (CPM) laser, operated at 620 nm which is amplified to a pulse energy of up to 300 μJ, and a pulse duration of 200 fs [6]. The CPM beam is expanded to illuminate a GaAs wafer with an area of 2×2 cm, as depicted in Fig. 1. The GaAs surface is connected on opposite sides with two electrodes. On these electrodes an electric field is applied. The field strength across the wafer can be as high as 7.5 kV/cm. The radiation that is emitted in the forward direction passes through a wire grid polarizer, to eliminate imperfectly polarized components that may originate from small inhomogeneities of the electric field in the GaAs wafer. The direction of the electric field of the HCP can be chosen continuously with respect to the static field between the two field plates in the streak camera. The semiconductor wafer and the polarizer are mounted together on a rotatable mount. The radiation is sent into the vacuum system containing the atomic streak camera, after passing through a quartz window that absorbs less than 15% of the incident radiation. The surface area of the GaAs wafer is large compared to the wavelength of the generated light, so that the generated beam is well collimated. The THz radiation is not focused into the interaction region. An unfocused configuration was preferred to avoid possible problems when trying to focus all the frequencies that are present in the broadband radiation. As a result, only states that lie just below the ionization threshold can be ionized by the HCP.

The THz field strength has been calibrated at the interaction spot by ionizing field free Cs d states ($n \approx 40$), of which the ionization threshold with HCP with a FWHM of 0.5 ps is known from experiments by Jones *et al.* [2]. The measured maximum peak is approximately 17 kV/cm assuming a pulse duration of 0.5 ps. At this maximum output the bias over the GaAs wafer is approximately 7.5 kV/cm. The HCP duration was independently measured with a cross correlation technique, and found to be 0.5–1 ps (FWHM) [7].

The output of a nanosecond dye laser is frequency doubled to produce light around 320 nm that is used to excite the cesium atoms to a high lying Rydberg state. The Rydberg states are excited in the state electric field (2.0 kV/cm) that is required for the operation of the streak camera. Therefore, the states that are excited are not pure p states, but l-mixed Stark states. Examining the properties of Rydberg states ($n^* \approx 21$) at an energy around the ionization threshold in 2.0 kV/cm, it is found that the radial oscillation (Kepler) time is 1.5 ps. The HCP pulse duration, although not negligible with respect to this radial oscillation time, is still a factor of 2 to 3 shorter, so that the impulse approximation is a reasonable one.

In Fig. 2 two ionization spectra, obtained with the atomic streak camera, are shown. The spectra were obtained by exciting a Rydberg state at $\lambda = 321.36$ nm, 2 meV below the ionization threshold in the electric field, and subsequently ionizing the Rydberg atom with a HCP. The two orientations of the electric field of the THz pulse that are shown in Fig. 2 are 0° (parallel to the static electric field) and 180° (antiparallel). Note that there is a difference between the two cases because of the unipolar character of the HCP [8]. Both spectra show a large amount of ionization at $t = 0$, when the HCP arrives. However, the 0° spectrum, taken with the kick in the same direction as in which the electron is pulled by the static electric field, shows a significant amount of ionization at later times. This second burst of ionization peaks around $t = 11$ ps. Note that the time-integrated ionization yield

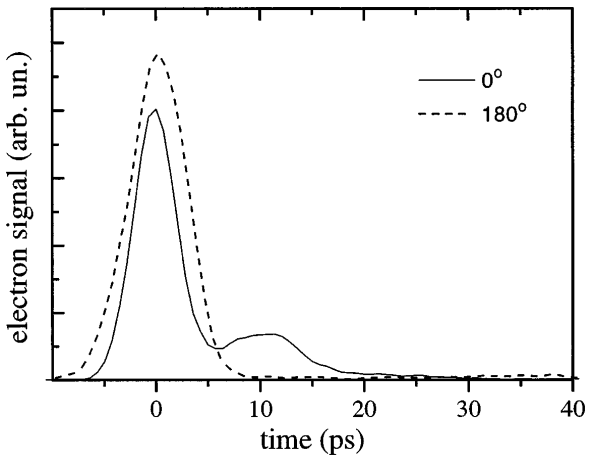

FIG. 2. Time-dependent electron emission from the THz ionization of a cesium Rydberg state, in a static electric field of 2.0 kV/cm. The Rydberg state was excited at a wavelength $\lambda = 321.36$ nm. The THz pulse had a peak electric field of 17 kV/cm and a duration of 0.5 ps. Two traces are shown, one for parallel alignment of the THz electric field with the static electric field (0°, solid line) and one for antiparallel alignment (180°, dashed line).

is the same for both orientations. A possible explanation for the observed delay for 0° orientation is that part of the wave function scatters off the core and follows a different trajectory that leads to delayed emission. However, the delay of 11 ps is very long compared to the radial oscillation time of 1.5 ps.

Apparently the delayed emission is not simply caused by a single bounce off the core in the direction opposite to the direction in which the electron is kicked. It is more likely that the electron is scattered into an orbit which has no overlap with the saddle point in the potential. Such orbits exist and can give rise to long lifetimes in the continuum [9], up to tens of picoseconds. Directly after scattering off the nucleus the electron has low angular momentum, because the impact parameter has to be small to allow the collision to occur. This angular momentum is not conserved in the electric field. The precession time of the angular momentum is proportional to the inverse of the energy splitting of the Stark states: $\tau_k = 2\pi/3Fn$. For states that have an energy close to the ionization threshold in the field of 2.0 kV/cm, this oscillation time is about 6 ps. The angular momentum has to return to low values before the electron can rescatter off the nucleus, possibly into an orbit that has overlap with the saddle point and thus allows emission of the electron. In previous experiments rubidium atoms were excited from the ground state to continuum states just above the ionization threshold in an electric field, using picosecond laser pulses [9]. In those experiments it was found that under some conditions the decay of the continuum wave packet showed a strong peak at the second recurrence of the oscillation of the angular momentum, i.e., at $2t_k$. Possibly such trajectories are followed in the current experiments by the part of the wave function that scatters of the nucleus after illumination with the HCP, and is emitted after 11 ps, approximately twice the precession period of the angular momentum.

Focusing on the behavior at 0°, several streak spectra were taken at various values of the peak electric field of the HCP, by varying the bias field on the GaAs wafer. The results are shown in Fig. 3 at the same absolute scale. As can be seen in Fig. 3 the total ionization yield decreases as the peak field of the HCP decreases, as expected. However, the results show that the decrease of the ionization yield is mainly caused by a decrease in the first burst of ionization. Only when the HCP peak field drops to even lower values, resulting in hardly any ionization, the second peak also decreases in amplitude. At those fields both ionization bursts of the same size; approximately 50% of the emitted electrons are delayed by 11 ps. The fact that the second peak grows with respect to the first peak favors the interpretation of the results in terms of scattering off the core. In general scattering cross sections increase with decreasing energy. If the second peak is caused by part of the wave function that scatters off the core, this process is likely to be more efficient for kicks of smaller amplitude.

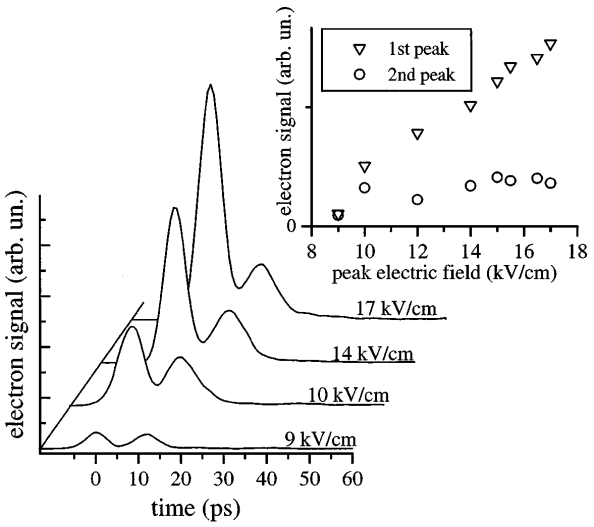

FIG. 3. Time-dependent emission from a cesium Rydberg state excited after ionization with THz pulses of various field strength (all at 0° orientation). All curves are plotted on the same absolute scale. The inset shows the amplitude of both peaks as a function of the electric field strength of the HCP.

The dependence of the time-resolved ionization on the THz orientation was further investigated by varying the alignment with only small angles from 0°. Figure 4 shows the traces taken under the same conditions as those in Fig. 2, but with the THz orientation varied from −25° to 25°. A rapid disappearance of the delayed ionization peak is observed, if the alignment with the static field is slightly different from 0°. The electron needs to be kicked into a very well-defined direction, in order to delay the ionization. The observed dependence is another indication for scattering. The Rydberg states are prepared in the electric field with a dye laser that is polarized parallel to the direction of the static field, defined as the z axis. Because the dye laser excited p character, the states with the largest cross section for excitation resemble a p_z wave function, with lobes extending along the z axis. If such a wave function is to be kicked onto the nucleus, the direction of the kick needs to be fairly well aligned.

A known feature of THz pulses generated from semiconductor wafers is the fact that they have a small subpulse, caused by internal reflection at the back plane of the wafer [10]. The amplitude of this reflection is generally less than 10% of the main pulse and the delay between the two pulses is approximately 10 ps. Although such a subpeak would not explain the observed dependence of the streak traces on alignment angle and peak field strength, the possibility that this subpulse is responsible for the delayed ionization was explicitly excluded. A second GaAs wafer was placed between the first, THz-generating wafer, and the Rydberg atoms. This second wafer was illuminated, by a split off fraction of the CPM fs-pulse beam, at the backside of the second wafer, antiparallel with the THz beam. The second wafer is switched from transmitting to

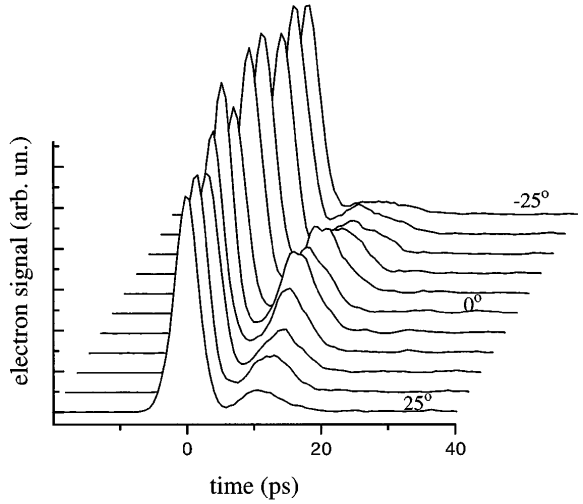

FIG. 4. Time-dependent emission with THz pulses (12 kV/cm) with varying orientation. The THz orientation is measured with respect to the static electric field of 2.0 kV/cm. 0° means parallel alignment of the electric field of the THz pulse with the static electric field.

reflecting on a picosecond time scale by varying the delay between the optical and the THz beam. By inspecting the two peaks in the streak spectrum for different delays between the two beams, it was verified that both peaks disappear at the same, single delay. This means that the atoms are really struck by a single pulse, and nevertheless emit electrons in a double burst.

Although results are presented for only one particular Rydberg state, excited at $\lambda = 321.36$ nm, the described behavior was also observed for other Rydberg states, excited with one and two photons (in the latter case with the fundamental output from the dye laser, exciting *s* and *d* character). All the states that yielded a significantly large THz ionization yield to allow recording of the time-dependent emission by the streak camera, showed a second burst of ionization for the 0° orientation of the HCP, at approximately 11 ps.

For a more quantitative analysis of the observed results, especially in terms of scattering, it is necessary to know the initial Rydberg wave function. The wave function of the Rydberg state that was excited in the experiment has been calculated by Robicheaux [11], using the method of Robicheaux and Shaw [12]. The theoretical wave function does not show a large asymmetry in the amount of probability localized on the $+Z$ and $-Z$ direction from the nucleus, if the static electric field of 2 kV/cm points in the Z direction. Therefore the asymmetry in the results obtained for a THz pulse with 0° or 180° orientation cannot simply be ascribed to an asymmetry in the wave function. However, a tentative explanation may be the following. First, it is assumed that electrons that do not scatter off the core are emitted almost immediately and are detected in the first burst of emission. For the 0° orientation of the kick, electrons that are on the $+z$, or high energy side of the field, are kicked onto the nucleus. On that side of the nucleus more relatively stable orbits exist because there is little overlap with the saddle point in the potential: the probability to scatter into a long-lived state is relatively high. For the 180° orientation, the part of the wave function on the $-z$ side of the nucleus is kicked onto the nucleus. On that side of the nucleus, however, hardly any stable states exist because of the proximity of the saddle point. Although scattering may occur, the electron is unlikely to scatter into a long-lived state. Further theoretical investigations are required to confirm this intuitive explanation.

Summarizing, we have investigated the time-dependent electron emission from Rydberg atoms in a static electric field that are ionized with subpicosecond half-cycle pulses. The data show a delayed burst of emission at 11 ps after the interaction with the HCP. The relative amplitude of the second peak is maximum if the electron is kicked in the same direction as the static electric field and the HCP barely ionizes the atom. Both the dependence on peak electric field and the orientation indicate that the electron scatters off the nucleus into a metastable orbit before it is emitted.

It is a pleasure to acknowledge R. R. Jones for fruitful discussions. This work is part of the research program of the Stichting Fundamenteel Onderzoek van de Materie (FOM), which is financially supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO).

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