Indications of High-Intensity Adiabatic Stabilization in Neon

M. P. de Boer, J. H. Hoogenraad, R. B. Vrijen, L. D. Noordam, and H. G. Muller
Foundation for Fundamental Research on Matter (FOM)-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands
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We study the stabilization behavior of the circular $5g$ state in neon in a two-pulse experiment. A first pulse prepares the state. Comparison of the single-photon ionization yield, due to a second laser pulse for both short (0.1 ps) intense and long (1 ps) less intense pulses, shows an intensity-dependent suppression of ionization. The smaller yield due to the short pulses, at intensities of several times 10^{13} W/cm^2, is in accordance with recent predictions of stabilization.

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The photoionization rate of an atom will usually increase when the atom is exposed to increasing intensities. Recently, however, different mechanisms have been proposed that could prevent photoionization of an atom, even at the highest intensities. Collectively these are called stabilization. One such mechanism is dynamic or transient stabilization, in which part of the population is temporarily inaccessible for ionization [1].

Adiabatic stabilization [2–5] is a different mechanism that suppresses photoionization. More precisely, it corresponds to an ionization rate which *decreases* with *increasing* intensity. This type of stabilization is predicted to occur in the high-frequency regime, where the photon energy is much larger than the binding energy. Classically, this means that the electron motion due to the light field is much faster than its motion in the Coulomb potential. Quantum mechanically, it means that the electron wave function cannot adiabatically respond to the oscillating light field. As a result, in the high-frequency regime, the wave function of the electron will be driven in an oscillatory motion by the light field.

Adiabatic stabilization may occur once the amplitude of the oscillatory motion becomes comparable to the size of the unperturbed wave function, which usually implies a high intensity [6]. In the rest frame of the oscillating electron cloud (the Kramers-Henneberger frame), the atomic core will appear to oscillate. The electron wave function is determined by the potential of the atomic core, which, in this rest frame, is time dependent. However, in the high-frequency regime, it is valid to time average the motion of the atomic core. The potential which results is no longer Coulombic, but is deepest at the outer turning points of the oscillation, where the atomic core spends most of its time. In response the probability of finding the electron is largest at these points. One result of the electron spending more of its time further away from the nucleus is that ionization is suppressed and stabilization occurs. Ionization depends on transfer of momentum from the core to the emitted electron and once the wave function is far away from the core this becomes impossible [7].

Originally, stabilization was predicted for the ground state of atomic hydrogen [2], and was later confirmed by other calculations [3]. However, with present-day laser technology, the stabilization regime can only be reached when the initial state is a Rydberg state. For that case, different theoretical groups [4,5] give similar predictions for the intensities and frequencies at which stabilization occurs. To our knowledge, this Letter reports the first experimental indication for adiabatic stabilization.

Our experiment relies on a pump-probe setup [8]. The pump pulse excites a circular Rydberg state. Of all the Rydberg states with the same quantum number n, the circular state is the one with the highest magnetic quantum number m ($m=l=n-1$). The one-photon ionization yield due to a subsequent, linearly polarized, probe laser is measured. Because of the selection rule $\Delta m=0$, this light only couples the circular Rydberg state to other states with the same m value. No deeper bound states are accessible and the circular state is the ground state of the $m=l$ manifold. For the probe pulse high-frequency theory applies since the photon energy is much higher than the binding energy of the Rydberg state.

Stabilization is easily visualized as vigorously shaking the electron cloud. For a circular state the electron cloud resembles a doughnut. To fulfill high-frequency conditions the light should be linearly polarized along the quantization axis (the symmetry axis of the doughnut). Intuitively, the oscillatory motion will only increase the average distance of the doughnut to the nucleus and hence such a motion stabilizes the wave function.

In an experiment, before the stabilization regime is reached an atom may already experience relatively high intensities during the rising edge of the pulse. In principle the ionization rate could be so high, and the corresponding lifetime could become so short that all atoms are ionized before the stabilization regime is reached [9]. This has become known as the "Death Valley" problem [10]. A circular state has a small photoionization cross section [11] due to its large angular momentum. Therefore the minimum lifetime in "Death Valley" is relatively long: "Death Valley" may be crossed with 0.1 ps pulses and stabilization can be observed. In addition, in our setup, the peak intensity of the probe pulse is equal for all the excited atoms because the focus of the probe laser is much larger than that of the pump laser that creates the

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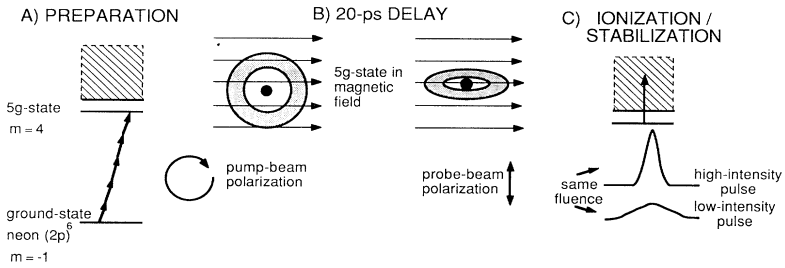

FIG. 1. Experimental scheme: (a) A pump pulse excites the $5g$ circular ($m=4$) state in neon. (b) We allow the atom to rotate due to a magnetic field (Larmor precess) until the atom is circular with respect to the vertical polarization of the probe laser. (c) Subsequently, the photoionization yield is measured using high- and low-intensity pulses of the same fluence.

circular state [12].

A circular Rydberg state can be excited through multiphoton absorption, by using a circularly polarized pump pulse [11] (see Fig. 1). We used five photons to excite a $5g$ state ($n=5$, $l=4$, $m=4$) in neon. The binding energy (0.544 eV) is sufficiently small compared to the energy of the probe photon (1.99 eV) for high-frequency theory to apply. The only transition which has $\Delta m=+5$, prescribed by the selection rule for five-photon absorption, is then the excitation of an $m=-1$ electron of the $2p^6$ shell to the circular, $m=4$ state. The angular momentum of the excited state, projected on the polarization axis of the probe pulse, will be maximum after a quarter Larmor period. We then rely on a magnetic field, perpendicular to the propagation direction of the laser beam, to rotate the atom through Larmor precession. Subsequently, a probe pulse arrives. In our experiment we compare the yield due to intense and less intense probe pulses with the same fluence. A decrease in yield for pulses with the same fluence but higher peak intensity will indicate stabilization.

After excitation spin-orbit and other angular couplings cause a rapid exchange of angular momentum, resulting in the excitation of three $5g$ states, $P_{3/2}$ [11/2], $P_{1/2}$ [9/2], and $P_{3/2}$ [9/2] in the intermediate coupling scheme [13]. The couplings cause the magnetic quantum number of the outer electron to oscillate somewhat between $m=4$ and $m=3$. Since the $m=3$ contribution never exceeds 13% we will ignore it.

A more detailed description of the setup now follows. A colliding-pulse mode-locked dye laser (CPM) is amplified at 10 Hz in four Bethune type dye cells. After recompression in a folded four-prism sequence it delivered 100-μJ, subpicosecond pulses [14]. Part of the resulting beam was used for continuum generation in a water cell. For this experiment we amplified a 3-nm-wide part of the white-light continuum around 571.5 nm in five dye cells (diameters 1, 1, 3, 8, and 16 mm) up to an energy of 1.5 mJ. The resulting pulses were collimated down to 8 mm, and frequency doubled in a 4-mm-long potassium diphosphate (KDP) crystal to obtain up to 200 μJ of 286-nm ultraviolet light in an approximately 1-ps-long pulse. The remaining light at the fundamental frequency was filtered out using a dichroic mirror. After frequency doubling the pump polarization was adjusted to circular polarization using a quarter-wave plate.

The remainder of the amplified CPM pulses were sent through a pulse shaper [14] to adjust the frequency chirp. Using this device the pulse duration could be adjusted to be as short as 0.1 ps or as long as 3 ps. The durations were measured by pulse-energy-resolved autocorrelation, which revealed no correlation of duration with energy. The bandwidth of the resulting pulse was measured to be 5 nm around 623 nm. After the shaper, diaphragms could be put into the beam to vary the final pulse energy and consequently the maximum fluence in the focus. Changes in beam direction or profile due to possible misalignments in the pulse shaper or the influence of the diaphragms were prevented from propagating through the system by subsequent spatial filtering to the diffraction limit. The pulses were reamplified in two dye cells (diameters 3 and 8 mm) and recompressed once more in a folded four-prism sequence. A diaphragm was used to select the 5-mm-wide center of the beam, containing up to 55 μJ energy per pulse. The relative timing of the two light pulses could be adjusted. It was checked that changing the delay between the two pulses did not vary the spatial overlap between the two foci.

The electron spectrometer is of the magnetic-bottle type [15], containing a magnetic field of about 0.9 T. The ionization chamber was filled with 3×10^{-4} mbar of neon, while the background pressure was 5×10^{-9} mbar.

At our experimental wavelength of 286 nm, the $5g$ state ac Stark shifts into resonance at an intensity of 8.6×10^{13} W/cm^2 and a reasonable amount of population is transferred. For the pulse duration of 1 ps the saturation fluence for photoionization of the $5g$ state by the pump laser, 400 J/cm^2, corresponds to an intensity of 40×10^{13} W/cm^2. This intensity is never reached, so that almost all the excited population survives the pump pulse.

We will now discuss our experimental results. In the absence of the probe beam, the electron spectrum [see Fig. 2(a)] contains a broad peak at 3.6 eV, corresponding to nonresonant ionization with six photons. An enhancement at the position of the $3d$ resonance (2.8 eV) is visible. Selection rules forbid this transition using circularly polarized light, so the enhancement is due to either a remnant of linear polarization or linear polarization in-

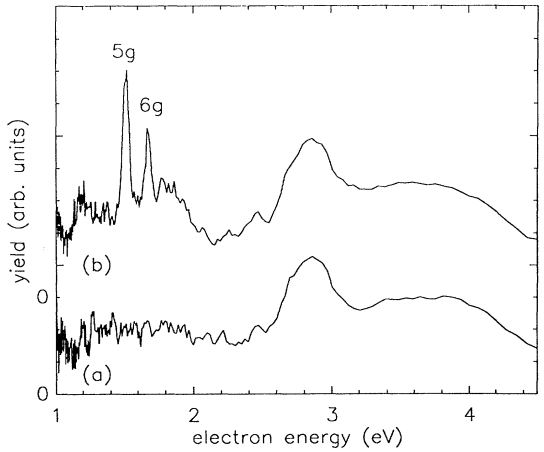

FIG. 2. (a) Electron energy spectrum of neon with pump pulses only (1 ps, 286 nm, circularly polarized). The broad bump at 3.6 eV corresponds to six-photon ionization via off-resonant continuum g states. As a result of a small fraction of linear polarization, there is still some resonant enhancement by the $3d$ state, leading to the peak at 2.8 eV. (b) When the probe pulse (1 ps, 620 nm, vertical polarization) arrives 40 ps after the pump pulse well-resolved additional peaks are seen. They correspond to excitation of Rydberg g states by the pump beam and subsequent one-photon ionization by the probe laser.

duced by the tight focusing.

When the probe pulse arrives after the pump pulse, well-resolved overlap peaks are observed [see Fig. 2(b)]. In this case the probe pulse was chirped out to a pulse duration of 1 ps. Under these conditions the probe beam alone does not cause any signal. The overlap peaks correspond to single-photon ionization of different Rydberg g states (the principal quantum number n ranging from 5 to 8 and $m=4$), which were resonantly populated by the pump laser as well, since they ac stark shift into resonance at lower intensities. Note that Raman redistribution through the continuum [1,16] does not play a role since both the laser bandwidth (16 meV) and the extrapolated perturbative lifetime (30 meV at 1.2×10^{14} W/cm^2) are much smaller than the spacing between the levels.

To study the saturation behavior of the $5g$ states, the magnitude of the corresponding overlap peak was measured as a function of the fluence of the probe pulse and for two different pulse durations. In principle, the yield will depend on the fluence, the peak intensity (stabilization or no stabilization), and the distribution of population over various m states. If the stabilization regime is reached with the short pulses, the yield should be smaller than that due to the long pulses for the same fluence. We adjusted the delay, $\Delta t=20$ ps, so that the probe pulse arrived after one-quarter Larmor precession and the $5g$ population was in the $m=4$ circular state. The perturbative saturation fluence is the fluence which leaves a fraction $1/e$ of the excited population un-ionized. This fluence, 15 J/cm^2 for the circular $5g$ state, was of the order of our maximum available fluence, 12 J/cm^2. Indeed,

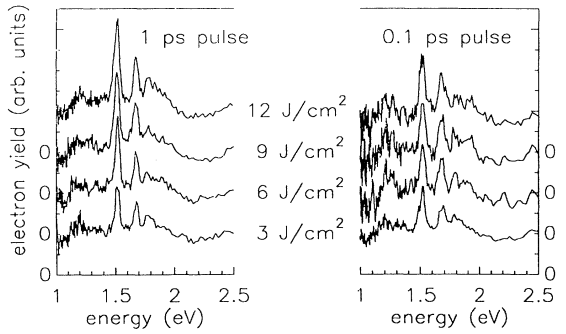

FIG. 3. Electron energy spectra for various fluences and two different pulse durations. The spectra are averaged over 2000 and 4000 shots for the long and short pulses, respectively. The background due to ionization when only the probe pulses are present has been subtracted. The peaks at 1.5 eV correspond to one-photon ionization of the $5g$ state. For the short pulses the photoionization yields hardly increase with fluence.

with a 1-ps pulse, the yield still increases with fluence (see Figs. 3 and 4). Theoretically no stabilization is predicted for the maximum attained peak intensity of 1.2×10^{13} W/cm^2.

We could also zero the chirp of the probe pulses to given short pulses of 0.1-ps duration, while the used fluences remained the same. In this case the peak intensities are much higher (up to 12×10^{13} W/cm^2). This intensity is higher than the theoretically predicted stabilization intensity for the $5g$ state of 5.5×10^{13} W/cm^2 [5]. In this case the yield hardly increases with increasing fluence (see Figs. 3 and 4). Comparison of the yields for both types of pulses therefore indicates stabilization: The one-photon ionization rate depends on the peak intensity

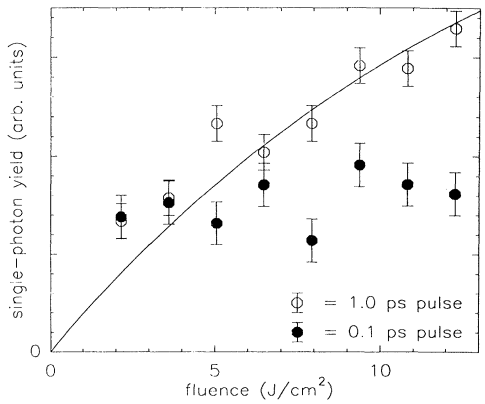

FIG. 4. Photoionization yield of the $5g$ circular state as a function of the fluence. The open circles were measured with low peak intensity (up to 1.2×10^{13} W/cm^2, 1 ps). The curve represents a fit using the theoretical perturbative rate (including depletion) and an adjustable asymptotic value. The solid circles were measured using shorter pulses, with the same fluence but more intense (up to 12×10^{13} W/cm^2, 0.1 ps). The yield due to these pulses hardly increases with fluence, which indicates stabilization.

for pulses with the same fluence.

For the short pulses (0.1 ps), the probe peak intensity at the highest fluence used is enough to cause some ionization of ground-state neon with the probe pulse alone [17]. This small but significant background had to be subtracted from the pump-probe spectra since the probe-only spectra also showed some resonance enhancement at the exact position of the $5g$ state. For this subtraction we used the spectra where the probe arrived 20 ps before the pump. Note that the background is hardly affected by ground-state depletion due to the pump pulse, since it originates from a roughly 100 times larger volume.

The enhancement peak in the probe-only spectra is due to excitation of the $5f$ state ($m=0$) with twelve red photons, which occurs at a Stark shift of 2.86 eV. Assuming the state shifts with the quiver energy this corresponds to an intensity of 8.0×10^{13} W/cm^2. This gives us the most accurate intensity calibration of our spectra and agrees with the measured energies, focal sizes, and pulse durations.

Our setup allowed us to measure the size of the probe focus. We measured the magnitude of the $5g$ overlap peak, due to the presence of both beams, when the probe arrives 40 ps after the pump. This gives us a measure for the red probe intensity at the pump focus. The result was consistent with a diffraction-limited probe focus diameter of 11 μm (FWHM), and a pump focus that was significantly smaller (≈ 3 μm). Thus all prepared Rydberg states essentially see the same peak intensity of the red pulse. In the pump focus, at a pressure of 3×10^{-4} mbar, there are approximately 1700 atoms. Since we detect approximately 50 electrons per laser shot and our detection efficiency is approximately 25%, this means that we can neglect depletion of ground-state atoms. On average the pump pulse excites about 1 atom per shot to the $5g$ state.

A possible complication is that the $5g$ population may be photoionized by two photons instead of one photon when using the shortest, and therefore most intense, pulses. However, even for the 0.1-ps pulses, the only signal we observed due to the pump-probe combination corresponds to one-photon ionization. Considerations from high-frequency theory predict that the one-photon ionization peak should dominate the electron spectrum, mainly because higher-order peaks require a larger momentum exchange with the nucleus [18]. The hard collisions, required for the high-order peaks, are unlikely to occur in a circular g state, even in the perturbative regime, because the electron is localized so far from the atomic core. They are especially unlikely to occur in a stabilized state, which is localized even further from the core.

In conclusion, we have observed a strong indication for stabilization of circular $5g$ states in neon which occurs for intensities above several times 10^{13} W/cm^2. This result is in excellent agreement with calculations for the lifetime minimum by Potvliege and Smith [5] for the states involved. A detailed experimental test of their lifetime curves is not yet possible due to the poorly known temporal profile of our laser pulses.

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