Ionization suppression of Rydberg atoms by short laser pulses

J. H. Hoogenraad, R. B. Vrijen, and L. D. Noordam
FOM Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands
(Received 20 October 1993; revised manuscript received 25 May 1994)

One-photon ionization from the $6s27d\,^3D_1$ state in barium is measured with short (0.25–2.7 ps), high-intensity laser pulses. Fermi's golden rule predicts that only the fluence (time-integrated intensity) determines the yield. We observed a decrease in the yield for fixed-fluence pulses shorter than the Kepler orbit time of the Rydberg electron (2.2 ps). This is explained semiclassically: The wave function of a Rydberg electron performs a Kepler-like orbit. Only the wave function near the core can be ionized. Not all of the wave function nears the core during a short pulse, and therefore the wave function far away from the core is stable against ionization. A quantum-mechanical calculation based on Raman transitions over the continuum agrees well with experimental observations and the semiclassical explanation.

PACS number(s): 32.80.Rm, 42.50.Hz

I. INTRODUCTION

One-photon ionization of atoms is a process for which low-intensity results are well known as Fermi's golden rule: The ionization rate is proportional to both intensity of the laser light and the initial-state population. The number of ionized atoms (the *yield*) is proportional to the fluence (time-integrated intensity) of the applied light. Even if the initial state becomes depleted during the pulse, the fluence of the (pulsed) light field determines the yield, regardless of the temporal pulse shape. We demonstrate here experimentally and theoretically that this dependence on fluence breaks down for very short laser pulses. We will show that for pulses shorter than the Kepler orbit time of the electron, complete photoionization is prohibited irrespective of the fluence. Calculations on this phenomenon (known as *ionization suppression* or *transient stabilization*) have been reported previously [1–4].

The electron wave function of a Rydberg state is spatially extended (for an $n = 24$ state, 600 Å). Radial confinement of the Rydberg wave function can be achieved by exciting a superposition of Rydberg states (a wave packet) with a short laser pulse. After the pulse, the wave packet oscillates between the core and the outer turning point. Ionization of the Rydberg wave function occurs only close to the core [5], because the combination of momentum and energy conservation requires that the core absorb recoil momentum of the electron. Therefore ionization by a probe pulse is only observed whenever the wave packet passes the core [6]. The oscillation period of a Rydberg electron is the Kepler orbit time ($2\pi n^3$, 2.1 ps for $n = 24$) of the coherently excited states, equal to the inverse of the level spacing between the states ($\Delta E = 1/n^3$).

Let us now consider a single, stationary, Rydberg state irradiated by a pulse that is very short compared to its Kepler orbit time. In the initial stationary Rydberg state, the wave function has appreciable amplitude both near the core and far away. Ionization will take out wave function near the core. During the short pulse, no wave function from the outer region can be supplied and therefore ionization is limited by depletion of the wave function near the core. The created hole in the wave function is called an *antiwave packet* [3]. In quantum mechanics, this antiwave packet has to be described as a coherent superposition of states: The ionizing pulse not only releases wave function into the continuum, but also populates some adjacent Rydberg states [3,7] by Raman transitions via this continuum [1,2]. The two photons required for this stimulated Raman transition are available within the bandwidth of a short laser pulse. For this process, the bandwidth must be larger than the spacing between the states. This statement is equivalent to requiring the pulse to be shorter than the Kepler orbit time.

In this article we present an experimental study of ionization of a stationary Rydberg state and a fully quantum-mechanical description of the observations.

II. EXPERIMENT

For our experiment, we prepared barium atoms in the $6s27d\,^1D_2$ Rydberg state. This state has a Kepler orbit time of 2.2 ps. Subsequently, the atoms were photoionized with pulses with a duration ranging from much shorter (0.25 ps) to somewhat longer (2.7 ps) than the Kepler orbit time. The number of emitted electrons per laser shot was measured as a function of the pulse duration and energy of the ionizing pulses.

The barium Rydberg state was excited from the $6s^2\,^1S_0$ ground state in two steps. We used a pulsed dye laser with a wavelength of 553.7 nm to populate the intermediate $6s6p\,^1P_1{}^0$ state, followed by a second dye laser at 420.39 nm to populate the $6s27d\,^1D_2$ state. The pulses overlapped in time, as was verified using the Sr $9s$ two-photon resonance at 410 nm. The dye lasers were pumped by one 5-ns, Q-switched 10-Hz Nd-doped yttrium aluminum garnet (Nd:YAG) laser.

Pulses of 0.1 ps duration at a central wavelength of

620 nm were generated by a colliding-pulse mode-locked (CPM) dye laser system [8]. The pulses can be lengthened up to 3 ps with a pulse shaper [8] either by reducing the bandwidth or by applying a *chirp* (separating the frequency components in time). We chose the latter method since it does not change the fluence of the pulses. Also, artifacts from changing the wavelength of the light, possibly changing the cross sections, are avoided. Changing the chirp to stretch the pulse can only be used with one-photon processes where the phase within the laser pulse is not relevant [9]. This requirement was proven to be valid for the system under study [10]. Note that the redistribution of population to neighboring Rydberg states is due to a Raman transition over the continuum, where states within the (large) bandwidth of a short pulse are populated. For a pulse stretched by chirp, the bandwidth in any time interval is decreased, even though the total bandwidth is constant. The repopulation is a fast process, and therefore only perceives the small "instantaneous bandwidth," corresponding to a long pulse.

The pulses were attenuated to the desired energy by an aperture. A spatial filter with an overfilled pinhole followed to ensure that none of the spatial beam parameters was affected upon changing the pulse duration or energy. The (sub)picosecond pulses were amplified in dye cells pumped by the same Nd:YAG laser as the excitation dye lasers. The pulse durations of the amplified pulses were measured with a resolution of 0.1 ps using a scanning autocorrelator. After excitation of the Rydberg state, some population remains in the intermediate $6s6p\,^1P_1{}^0$ state. The (sub)picosecond light was delayed approximately 100 ns with respect to the excitation lasers to let the population in this short-lived state decay to the ground state. The lifetime of the prepared Rydberg states is much longer than this delay.

The laser light crossed an effusive beam of barium from an oven, resistively heated to 600 K. The (sub)picosecond pulses were focused to a spot of 10 μm diameter. The excited Rydberg-state population was approximately constant in volume around the focus of the (sub)picosecond light as the foci of the preparation lasers were five times as wide. A disadvantage of focusing is that all intensities up to the peak intensity are present near the focus. The volume in which a certain minimal fluence is reached increases rapidly with increasing peak fluence, resulting in more ionization. We present the data as measured (averaged over all fluences present) and include the focal effect in the calculations.

We detected the electrons emitted by ionization with a multichannel plate detector at 5 cm distance (50-ns electron flight time) from the interaction center. Its signal was fed directly into a digitizing oscilloscope with a sampling time of 5 ns. Using the time of flight (TOF) of the electrons, the system had a limited energy resolution of about 1 eV.

Two processes allow unwanted ionization of barium, and can contribute to a background in the measurements: ionization with the excitation laser and, when the intensity of the (sub)picosecond light is high, three-photon ionization of barium ground-state atoms. Electrons emitted during the excitation could be distinguished because of the 100-ns delay of the (sub)picosecond pulses. We exploited the energy resolution to reduce signal from direct ionization from the barium ground state, and above-threshold ionization (ATI) from the ground state. Due to this procedure, the small signal from ATI from the Rydberg state was excluded from our measurement.

A calibrated photodiode was used to register the energy (proportional to the fluence) of each excitation and (sub)picosecond pulse separately. Time of flight traces were recorded for each laser shot and were summed by a computer for laser shots with an energy within one of the contiguously set energy intervals. The intensity distribution of the laser shots was much wider than the width of one interval, filling each interval evenly.

III. RESULTS

We have performed the measurements of the photoionization of the $6s27d\,^1D_2$ state as a function of both the fluence and the pulse duration of the ionizing pulse. In Fig. 1 the total electron yield for pulses with a fixed fluence of 7.8 J/cm^2 in the center of the focus is plotted as a function of the pulse duration. According to Fermi's golden rule, the ionization yield should not be dependent on the pulse duration. We observe indeed that for long pulses, the pulse duration does not matter. For pulses shorter than the Kepler orbit time (2.2 ps), the ionization yield starts to decrease (not all the wave function nears the core). For the shortest pulses, the yield becomes almost proportional to the pulse duration (this agrees with only ionizing the fraction of the wave function that actually nears the core during the ionizing pulse). In other words, the photoionization is observed to be strongly reduced for short pulses: Ionization suppression.

In Fig. 2 the yield is plotted as a function of the fluence

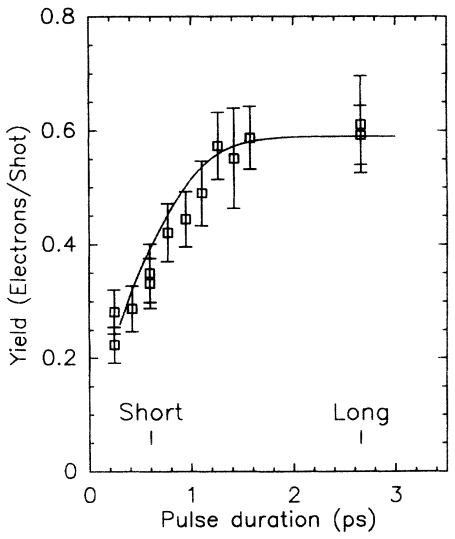

FIG. 1. The measured photoionization yield of the $6s27d\,^1D_2$ state in barium as a function of laser pulse duration for a constant fluence of 7.8 J/cm^2. The curve is calculated with the model presented in the text.

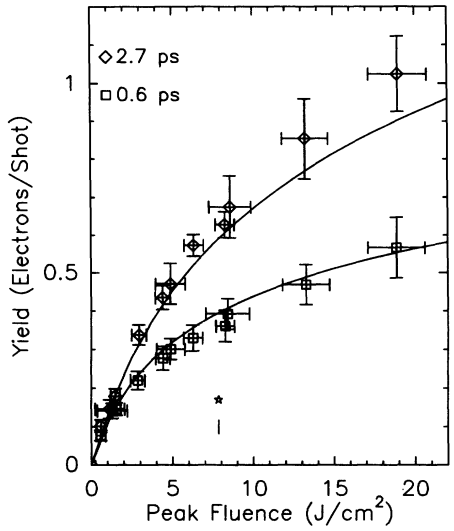

FIG. 2. The measured photoionization yield of the $6s27d\,^1D_2$ state in barium as a function of laser pulse fluence for pulses with a duration of 0.6 ps and 2.7 ps ("short" and "long" in Fig. 1). The curves are calculated with the model presented in the text. The asterisk at the bottom of the figure indicates the fluence at which Fig. 1 was measured.

in the focus for the long (2.7 ps) and short (0.6 ps) pulse durations denoted in Fig. 1. For low fluences, both curves start linearly with the same slope, as Fermi's golden rule is still a good approximation. Any deviation from a linear curve shows a form of depletion. The local depletion of the wave function is determined by the laser *intensity* rather than the fluence. Hence depletion starts at lower fluences for the short pulse measurements (the lower series) than for long pulses (the upper series). Moreover, the maximum ionization signal is reduced for the short pulses. Apparently, less wave function can be ionized by the short pulses. Note that even though the ionization from a single atom is limited to the fraction of the wave function that nears the core during the pulse, the measured line still creeps up, due to the increasing intensity in the outer regions of the focus. At the bottom of Fig. 2 the fluence at which Fig. 1 was obtained is marked.

Although we could use the time of flight of the electrons to gate background signals, we chopped both the 553.7-nm laser beam and the (sub)picosecond laser to record this small background. The fainter 420.39-nm laser was not chopped since it did not yield photoionization signal on its own.

The background signals were subtracted after the measurements were completed. Of the atoms in the focus of the (sub)picosecond light (70±5)% remain in the ground state (or decay to it from the intermediate state) after the excitation pulses. The three-photon ionization signal from the ground state, recorded without excitation pulses, is multiplied by this fraction before subtraction as the excited or ionized atoms cannot contribute to this background when the excitation lasers are present.

The errors in the yield in Figs. 1 and 2 include five contributions. The importance of the contributions varies depending on the pulse duration. The first is the statistical error due to the electron counting. Second, fluctuations in the density of barium Rydberg atoms were estimated for each measurement by looking at ionization due to the excitation pulses only. The two uncertainties in the background signal add proportionally to the total error bars. The background signal was less than 20% for all measurements with a pulse duration longer than 1 ps, and at maximum 70% for the measurements at 0.25 ps in Fig. 1 and at fluences of more than 10 J/cm^2 for the short pulses in Fig. 2. Its uncertainties are the variance in the ground-state depletion by the excitation pulses and the energy differences between the shots used for signal accumulation and background subtraction.

The horizontal error bars in Fig. 2 show the width of the distribution of shots in the energy interval. They are also included in the vertical error bars in Fig. 1 as the fifth contribution. The calibration of the fluence to an absolute scale has an additional error of 30%, as the fluence is calculated from the measured energy of the pulses and the focal spot size, estimated on the focusing parameters.

IV. TWO-ELECTRON EFFECTS

The experiment was performed in the alkaline-earth-metal atom barium. The interaction of the Rydberg electron with an inner electron enhances the coupling from the barium $6s27d\,^1D_2$ state to the continuum due to the mixed-in character of the $5d7d\,^1D_2$ state. The large coupling reduced the required intensities for the experiment, enhancing one-photon Rydberg ionization relative to three-photon ionization of residual ground-state population. The details of the interaction of the Rydberg series with this $5d7d\,^1D_2$ state are not included in the theory presented in the next section. We are, however, currently examining this further refinement of the theory.

A potential problem with barium is that an inner electron can be ionized instead of the Rydberg electron, yielding additional ionization signal [11,12]. In the experiment, the wavelength of the (sub)picosecond laser was detuned from resonances with low-lying excited ionic state, disabling this process.

Note that in other circumstances a two-electron effect related to ionization suppression is predicted [13,14]: By ionizing the inner electron, an autoionizing Rydberg state can be created. This state will decay with a stepwise decreasing rate, stepping down with every round trip-time of the Rydberg electron. This effect suppresses autoionization during the first round-trip time. The time-dependent autoionization rates show a stepwise decay with each Kepler round trip. In our quantum-mechanical model for ionization suppression we calculated the time-dependent Rydberg wave function. The time-dependent electron yield during a pulse shows a similar stepwise decay [4].

V. CALCULATIONS

To confirm the interpretation of the data using the concept of an antiwave packet, we have performed quantum-

mechanical calculations. The method has been described previously [4], so we will only give a summary here. We solved the Schrödinger equation for the dipole coupling of the Rydberg series with one infinitely wide, flat continuum. The continuum can be eliminated adiabatically [1]. The coupled Rydberg states can then be described with the following complex differential equation:

$$\frac{dc_n(t)}{dt} + i\omega_n c_n = -\Gamma_n |f(t)|^2 \sum_{n'} \frac{V_{n'}}{V_n} c_{n'}(t) \ . \qquad (1)$$

In this equation $c_n(t)$, ω_n, V_n and Γ_n are the probability amplitudes, energies, coupling matrix elements with the continuum, and Fermi's golden rule rates of ionization of the bound states $|n\rangle$, and $f(t)$ the envelope of the electric field of the laser pulse. If $f(t)$ is independent of time, Eq. (1) is an ordinary matrix-type differential equation. We solve it by diagonalizing this matrix. Time-dependent intensities can be described with discrete steps in the intensity envelope $f(t)$. During the time any intensity is present, we let the system evolve as for the square pulse. Between the steps, the remaining populations are projected onto the eigensystem for the next instantaneous intensity. This procedure converges with increasing number of intensity steps used [4].

The calculations were performed for hydrogen. The difference with barium was modeled by using lower hydrogenic states to accommodate the quantum defect of the d series of 2.8 and a prefactor for the higher photoionization cross sections. We simulated ionization out of the $24d$ state and included Raman coupling to the $15d$ to $33d$ states, summing the contributions to the matrix elements of the p and f continua. We neglected coupling to the s- and g-Rydberg series through these continua as the matrix elements for these Raman transitions are much smaller. Above-threshold ionization to higher-lying continuum states was also neglected. A Gaussian pulse built from 16 discrete intensity levels was used for the temporal pulse shape. The ionization yield was averaged over a three dimensional Gaussian focus for the spatial intensity distribution.

We fitted this model to the experimental observations of Fig. 1 and Fig. 2. The only two parameters to one least-squares fit were the initial Rydberg population and the multiplication factor for the cross sections. The curves in both figures are calculated with the *same* two parameters. The fitted barium cross sections were 13 ± 6 times those of the hydrogenic $6s(n-3)d$ states. From the calculations we found that the Rydberg atom became depleted with long pulses of 2 J/cm^2. The measured yield in Fig. 2 continues to increase with fluence due to a growing contribution from regions near the focus.

Not only are the data well described by the quantum-mechanical model, the wave functions during and after the pulse also exhibit the described wave-packet-like structure. Figure 3 shows the radial distribution of the Rydberg wave function $[r^2|\psi(r)|^2]$ while ionizing with either a short (lower half of the figure) or a long pulse (the upper half), both with the same fluence of 2 J/cm^2. Along the horizontal axis, the distance to the core is plotted, and in the vertical direction, the calculated wave functions $[r^2|\psi(r)|^2]$ are stacked at 0.25-ps intervals. To the left of each half, the intensity of the light field during the pulse is shown. Let us first discuss the behavior due to a short pulse. Before the pulse a stationary Rydberg state exists (lowest trace in both figures). During the pulse, the population near the core is removed and after the pulse an antiwave packet oscillates with the Kepler orbit time between the core and the outer turning point of the Coulomb potential. The top half of the figure shows that during a long pulse *with the same fluence* the wave function is removed more gradually. After the pulse most of the population is ionized, with the remainder in a stationary state. Apparently, the slow supply of wave function from the outer region to the core sets a maximum to ionization with short pulses.

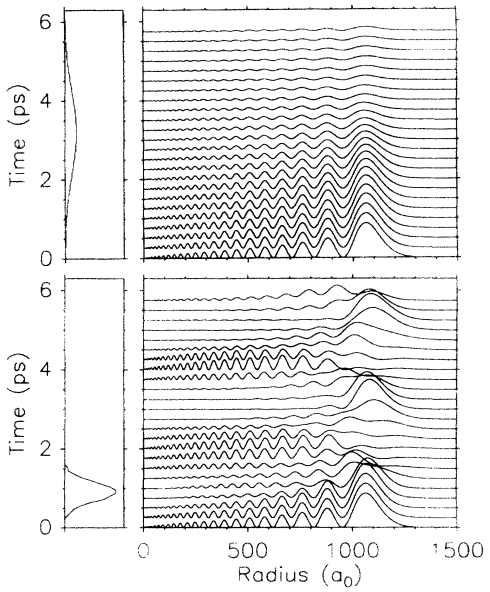

FIG. 3. The wave function $[r^2|\psi(r)|^2]$ plotted each 0.25 ps during and after photoionization of the $24d$ state of hydrogen. To the left the instantaneous intensity of the used long (2.7 ps, upper box) and short (0.6 ps, lower box) pulse is shown. Both pulses have an equal fluence of 2 J/cm^2.

VI. RELATED RESULTS

Finally we would like to discuss several types of stabilization discussed in literature, and compare them with ionization suppression.

In *adiabatic stabilization*, the ionization rate can be reduced in strong electromagnetic fields because of the high intensity. In these fields, the electron wave function can become dominated by the light field. The core interaction needed for ionization will become inhibited due to the large quiver motion of the electron and the ionization rate can even decrease with intensity. The first theoretical predictions on adiabatic stabilization were based on calculations on the ground state of hydrogen [15–18]. After the calculations have been extended to low-lying

excited states [19–21], the first experimental results have been recently obtained [22].

Stabilization for high-lying Rydberg states with long pulses has been predicted as well [23]. The Raman coupling of the states to the continuum describes destructive interference in the ionization. This stabilization occurs at high intensities, when the *Rabi oscillation time* between the continuum and the Rydberg states is shorter than the Kepler orbit time. In contrast, we report on ionization suppression for pulse durations shorter than the Kepler orbit time, at lower intensities. For the shortest pulses, the intensity in the center of the focus is comparable to where adiabatic stabilization of Rydberg states is predicted. Our calculations may lose their validity in this regime, limiting the curve in Fig. 1 to pulse durations longer than 0.3 ps.

Excited-state population trapping in high-intensity ground-state ionization is often associated with ionization suppression. It is shown that high-lying Rydberg states can shift into resonance due to ac-Stark shifts of both the ground state and the Rydberg states [24]. During strong-field ionization, population can be transferred at the crossings of the dressed ground state and Rydberg states [25]. The amount of population transferred is highly dependent on the intensity, shape, and duration of the pulse [26,27]. Even though the observed amounts of transferred population are small, ground-state ionization might be reduced by population trapping in the high-lying states. Both the small coupling with the continuum of these states and ionization suppression can reduce ionization from those states and hence the total ionization yield of the ground-state atoms.

For ground-state ionization of krypton Jones *et al.* [28] observed an increase in the population remaining in high-lying Rydberg states (relative to the ionization signal) when submitted to shorter pulses. In their experiment the same pulse was used both to populate the Rydberg states and to ionize them. The Rydberg state production process during this pulse complicates, as mentioned above, the assignment of different contributions of excitation dynamics, population trapping and ionization suppression.

Huens and Piraux [29] have predicted that a combination of ionization suppression and adiabatic stabilization can occur when a trapped wave packet is excited while ionizing a low-lying state. They propose that apart from the crossings with the ac-Stark shifted states, off-resonant Raman transitions through higher continua also transfer population into the Rydberg states.

We would like to point out that our results can also be interpreted as inhibition of ionization due to laser induced continuum structure (LICS) [30,31]. Usually, LICS is demonstrated for two states coupled through the same continuum using lasers with a different wavelength, while in our case of antiwave packet formation many neighboring states are coupled by the large bandwidth of the same ionizing laser.

VII. CONCLUSION

We have observed ionization suppression of a Rydberg atom exposed to a short, intense laser pulse. The results can be understood by the formation of an anti-wave packet, or the removal of wave function near the core. For pulses shorter than the Kepler orbit time, not all wave function passes the core during the pulse and complete ionization is inhibited. Quantum-mechanical calculations show excellent agreement with the data.

ACKNOWLEDGMENTS

We like to thank T. F. Gallagher, H. G. Muller, H. B. van Linden van den Heuvell, and W. J. van der Zande for fruitful discussions. The work described here is part of the research program of the Stichting Fundamenteel Onderzoek van de Materie (Foundation for Fundamental Research on Matter) and was made possible by the financial support from the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (Netherlands Organization for the Advancement of Research).

[1] J. Parker and C. R. Stroud, Jr., Phys. Rev. A **41**, 1602 (1990).
[2] K. Burnett, P. L. Knight, B. R. M. Piraux, and V. C. Reed, Phys. Rev. Lett. **66**, 301 (1991).
[3] L. D. Noordam, H. Stapelfeldt, D. I. Duncan, and T. F. Gallagher, Phys. Rev. Lett. **68**, 1496 (1992).
[4] J. H. Hoogenraad and L. D. Noordam, in *Super-Intense Laser-Atom Physics*, Vol. 316 of *NATO Advanced Study Institute, Series B: Physics*, edited by B. Piraux, A. L'Huillier, and K. Rzążewski (Plenum, New York, 1993), pp. 269–278.
[5] A. Giusti-Suzor and P. Zoller, Phys. Rev. A **36**, 5178 (1987).
[6] A. ten Wolde, L. D. Noordam, A. Lagendijk, and H. B. van Linden van den Heuvell, Phys. Rev. Lett. **61**, 2099 (1988).
[7] R. R. Jones, C. S. Raman, D. W. Shumacher, and P. H. Bucksbaum, Phys. Rev. Lett. **71**, 2575 (1993).
[8] L. D. Noordam, W. Joosen, B. Broers, A. ten Wolde, A. Lagendijk, H. B. van Linden van den Heuvell, and H. G. Muller, Opt. Commun. **85**, 331 (1991).
[9] B. Broers, H. B. van Linden van den Heuvell, and L. D. Noordam, Opt. Commun. **91**, 57 (1992).
[10] V. P. Chebotayev and V. A. Ulybin, Appl. Phys. B **52**, 347 (1991).
[11] R. R. Jones and P. H. Bucksbaum, Phys. Rev. Lett. **67**, 3215 (1991).
[12] H. Stapelfeldt, D. G. Papaioannou, L. D. Noordam, and T. F. Gallagher, Phys. Rev. Lett. **67**, 3223 (1991).
[13] X. Wang and W. E. Cooke, Phys. Rev. Lett. **67**, 976 (1991).
[14] X. Wang and W. E. Cooke, Phys. Rev. A **46**, 4347 (1992).
[15] M. Pont and M. Gavrila, Phys. Rev. Lett. **65**, 2362 (1990).
[16] P. Marte and P. Zoller, Phys. Rev. A **43**, 1512 (1991).
[17] M. Dörr, R. M. Potvliege, D. Proulx, and R. Shakeshaft,

Phys. Rev. A **43**, 3729 (1991).
[18] K. C. Kulander, J. J. Schafer, and J. L. Krause, Phys. Rev. Lett. **66**, 2601 (1991).
[19] M. Pont and R. Shakeshaft, Phys. Rev. A **44**, 4110 (1991).
[20] R. J. Vos and M. Gavrila, Phys. Rev. Lett. **68**, 170 (1992).
[21] R. M. Potvliege and P. H. G. Smith, Phys. Rev. A **48**, 46 (1993).
[22] M. P. de Boer, J. H. Hoogenraad, R. B. Vrijen, L. D. Noordam, and H. G. Muller, Phys. Rev. Lett. **71**, 3263 (1993).
[23] M. V. Fedorov and A. M. Movsesian, J. Opt. Soc. Am. B **6**, 928 (1989).
[24] R. R. Freeman, P. H. Bucksbaum, H. Milchberg, S. Darack, D. Schumacher, and M. E. Geusic, Phys. Rev. Lett. **59**, 1092 (1987).
[25] M. P. de Boer and H. G. Muller, Phys. Rev. Lett. **68**, 2747 (1992).
[26] J. G. Story, D. I. Duncan, and T. F. Gallagher, Phys. Rev. Lett. **70**, 3012 (1993).
[27] R. B. Vrijen, J. H. Hoogenraad, H. G. Muller, and L. D. Noordam, Phys. Rev. Lett. **70**, 3016 (1993).
[28] R. R. Jones, D. W. Schumacher, and P. H. Bucksbaum, Phys. Rev. A **47**, 49 (1993).
[29] E. Huens and B. Piraux, Phys. Rev. A **47**, 1568 (1993).
[30] Y. L. Shao, D. Charalambidis, C. Fotakis, J. Zhang, and P. Lambropoulos, Phys. Rev. Lett. **67**, 3669 (1991).
[31] S. Cavalieri, F. S. Pavone, and M. Matera, Phys. Rev. Lett. **67**, 3673 (1991).