Phase-dependent energy transfer in a microwave field

K. R. Overstreet, R. R. Jones, and T. F. Gallagher
Department of Physics, University of Virginia, Charlottesville, Virginia 22904-4714, USA
(Received 28 March 2012; published 15 May 2012)

We have excited Li atoms to Rydberg states with a picosecond laser in the presence of a phase-locked microwave field. We have observed energy transfer to higher- and lower-lying Rydberg states depending on the phase of the microwave field at which the picosecond laser excitation occurs. These observations confirm the prediction of a classical picture of the energy transfer and demonstrate that the phase-dependent energy transfer is not limited to processes in which energy transfer occurs across the ionization limit.

DOI: 10.1103/PhysRevA.85.055401 PACS number(s): 32.80.Rm, 78.60.−b

When an atom is excited by a short pulse of high-frequency radiation in the presence of a strong low-frequency field, the atom can gain or lose energy from the low-frequency field, depending upon the phase of the low-frequency field at which the excitation occurs [1–5]. As an example, consider He atoms which are exposed to an attosecond train of xuv pulses whose photon energy is less than the ionization potential of He, as shown by Fig. 1(a) [4,5]. If a phase-locked infrared (IR) field is also present, ionization occurs only at the phases of the IR field at which the electron can gain energy. Similarly, when Li atoms are excited to energies above the ionization limit by a picosecond (ps) laser pulse in the presence of a phase-locked microwave field, bound atoms are detected when the ps laser excitation occurs at the phase of the microwave field such that the electron loses energy, as shown by Fig. 1(b) [6]. The phase dependence of both of these processes can be understood in terms of a simple classical model based on the energy transfer from the low-frequency field to a photoelectron created in a Coulomb potential [7].

The above cases of ionization and recombination are two special cases of the same general phenomenon, phase-sensitive energy transfer to higher and lower energies by the low-frequency field. The fact that the impulse from the low-frequency field leads to energy transfer across the ionization limit is of no fundamental significance. Here we describe phase-sensitive energy transfer from an initially bound wave packet to more and less deeply bound states. Specifically, we observe phase-sensitive energy transfer among bound Rydberg states of Li, which occurs with ps laser excitation in the presence of a phase-locked microwave field. It occurs to both higher and lower energies, as shown in Fig. 1(c). In the following we briefly summarize the classical model, describe our experimental approach, present our results, and summarize the insights which can be gleaned from them.

A one-dimensional classical model which shows the origin of the phase dependence of the energy transfer is shown in Fig. 2. We are concerned with excitation by a temporally short laser pulse from the ground state of an atom to an energy W_0 near the ionization limit, i.e., $W_0 \cong 0$. Irrespective of whether the energy W_0 is above or below the limit, the initial kinetic energy of the photoelectron $K_0 = 1/r_0 + W_0 \cong 1/r_0$. We use atomic units unless specified otherwise. Here r_0 is the initial radial position of the electron, which is approximately the size of the ground-state orbit. The corresponding initial velocity of the electron is approximately given by $v = 1/(2r_0)^{1/2}$, and it decreases rapidly as the electron moves away from the ion core. Since photoabsorption of the high-frequency laser light occurs over more than one high-frequency field cycle, in the one-dimensional model in Fig. 2(a) the photoelectron is equally likely to be moving to the left or the right. The laser excitation occurs in the presence of a low-frequency field, $E \sin \omega t$. In our case it is a microwave field, but in the He experiments it is an IR field. If the laser excitation occurs at time t_0, the energy transferred from the field to the photoelectron is given by $W_{\rm tran}(t) = \int_{t_0}^{t} F dl$, where $F = -E(t)$ and l is the distance traveled. We can write the energy transfer from the microwave field as

$$W_{\rm tran}(t) = \int_{t_0}^{t} E(t')v(t')dt'. \tag{1}$$

If the low-frequency field is sufficiently weak, the electron's velocity can be approximated by the field free velocity, $v(t) = \sqrt{2/r(t)}$, while the electron remains in the deep part of the Coulomb potential near the ion core. Within this approximation, the velocity of the photoelectron decreases rapidly as the electron moves away from the ion, so the rate of energy transfer decreases as a function of time. Accordingly, the majority of the energy transfer between the low-frequency field and the photoelectron occurs during the first half cycle of the microwave field, when the velocity of the photoelectron is highest. At long times, after many field cycles, the integrated energy transfer is independent of t because the average electron velocity is no longer changing in time. As shown in Fig. 2(b), if the laser excitation occurs at time A, when $\omega t_0 = \pi/2$, the initial force on the electron is the largest. If the excitation occurs at the zero crossing of the field, at $\omega t_0 = 0$, the force is in the same direction for the entire first half cycle. It is thus not surprising that the maximum energy transfer occurs for excitation at a phase between these two times, at $\omega t_0 = \pi/6$, and the maximum energy transfer is given by [7]

$$W_{\rm max} = \frac{3E}{2\omega^{2/3}}. \tag{2}$$

Several points are worth noting about Eq. (2). First, the energy transfer to the electron can be either positive or negative, as indicated by Fig. 1(c). As shown by Fig. 2(a), the photoelectrons are created moving both to the left and to the right, so the phase ωt_0 which gives the largest energy gain to photoelectrons moving to the right extracts the most energy from photoelectrons moving to the left. Second, the energy transfer is linear in the field amplitude E, unlike the simple man's model, in which it is quadratic in E if

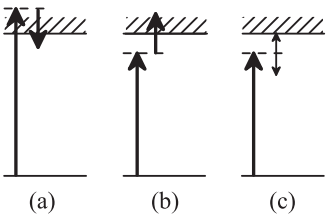

FIG. 1. (a) Excitation by a high-frequency pulse to an energy below the ionization limit results in ionization if the high-frequency pulse arrives at the phase of the low-frequency field which gives energy to the photoelectron. (b) Excitation by a high-frequency pulse to an energy over the limit results in a bound atom if the excitation occurs at the phase of the low-frequency field which removes energy from the photoelectron. (c) Excitation below the limit also results in phase-dependent transfer to more and less deeply bound states.

$W_0 \cong 0$ [8–10]. Whether energy is gained from or lost to the low-frequency field, the energy transfer can be described by Eq. (1). In our case the velocity of the photoelectron is due to the Coulomb potential, but in the simple man's model the Coulomb potential is neglected, so all the velocity must come from the low-frequency field if $W_0 \cong 0$. Finally, if the Coulomb potential is unimportant, either because W_0 is large or the low-frequency field is very strong, the maximum energy transfer occurs for $\omega t_0 = 0$ [1,2]. In contrast, in our case the Coulomb potential is very important, and the maximum energy transfer occurs at $\omega t_0 = \pi/6$.

The experimental approach is similar to one we have described previously [6]. A thermal beam of Li atoms passes through a 17.43-GHz microwave cavity where a ps laser produces a Rydberg wave packet at an energy below the ionization limit. The Rydberg wave packet is equivalent to the classical photoelectron shown in Fig. 2. The ps laser excitation occurs at a known phase of the microwave field, and the photoelectron gains energy from or loses energy to the field, depending on the phase. The microwave field is turned off 50 ns after the ps laser excitation, and the energy transfer to or from the field is determined by time-resolved selective field

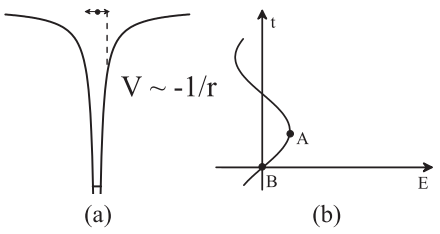

FIG. 2. Excitation in a one-dimensional potential by a ps laser, shown by the broken arrow, at a well-defined phase of the microwave field, as shown in (b). Laser excitation produces electrons moving both to the left and the right of $r \approx 0$ in the Coulomb potential. With excitation at $t_0 = t_A$, at a microwave field maximum, electrons are accelerated to the left, and electrons ejected to the left begin to gain energy, while those ejected to the right begin to lose energy. There is no instantaneous force if the electrons are ejected at $t_0 = t_B$, a zero crossing of the field, but the acceleration is in the same direction for the entire first half-microwave cycle. The maximum time-integrated energy transfer from the field occurs when the excitation occurs at $\omega t_0 \cong \pi/6$.

ionization. The atoms are exposed to a ramped ionization field, and as the field ramp rises, it progressively ionizes more deeply bound states. The electrons resulting from field ionization are detected with a microchannel plate (MCP) detector above the interaction region, and the time-resolved field-ionization signal is recorded with an oscilloscope. The field-ionization signal is later converted into the energy distribution of the final states.

The laser excitation occurs at a 1-kHz repetition rate. Two 20-ns dye laser pulses at 670 and 610 nm drive the $2s \rightarrow 2p$ and $2p \rightarrow 3d$ transitions [11], and a 2-ps 819-nm pulse drives the $3d \rightarrow nf$ transition. The 670- and 610-nm laser beams are 0.5 mm in diameter and propagate along the axis of the microwave cavity. They are crossed by the 2.2-mm-diameter, 819-nm ps laser beam at the microwave field antinode in the center of the cavity, producing a pencil-shaped sample of atoms which experience the same microwave field to within 3%. The uncertainty in the position of the ps laser beam relative to the microwave field maximum is 10%. The 819-nm pulse originates in a mode-locked Ti:sapphire oscillator which runs at a 91-MHz repetition rate and is amplified at a 1-kHz repetition rate in a regenerative amplifier. Using a mask in the stretcher of the regenerative amplifier, we convert the 100-fs pulses of the oscillator into 2-ps pulses. The mask also provides the tuning of the laser.

The 91-MHz pulse train of the Ti:sapphire oscillator is detected with a 25-GHz bandwidth photodiode, producing a microwave frequency comb. We phase lock the microwave oscillator, a Hewlett Packard (HP) 8350B/83550A sweep oscillator, to the 191st harmonic of 91 MHz, and the jitter between the microwave field and the ps laser pulse is 3 ps, comparable to the pulse width of the ps laser and substantially shorter than the microwave period of 56 ps. The output of the sweep oscillator is formed into 500-ns-long pulses with a HP 11720 pulse modulator, amplified to a maximum power of 400 mW, and sent by coaxial cable to the microwave cavity inside the vacuum chamber. The phase of the microwave field at which the ps laser excitation occurs is controlled by sending the ps laser beam through an optical delay line.

The Fabry-Pérot microwave cavity is composed of two brass mirrors of 10.2-cm radius of curvature with an on-axis separation of 7.91 cm. Since the cavity $Q = 2900$, the energy filling time of the cavity is 27 ns. For field ionization there are field plates, separated by 8 cm, above and below the region between the mirrors. To minimize stray electric fields we have an additional pair of plates on the sides of the cavity. These plates, the field plates, and the cavity mirrors are separately biased to reduce stray fields to less than 3 mV/cm. A negative voltage ramp with a maximum voltage of -200 V and a rise time of 1 μs is applied to the field plate below the interaction region, providing a maximum field of 30 V/cm, adequate to ionize atoms of $n \geqslant 66$ or binding energies less than 25 cm^{-1}. Since the flight time of the electrons is negligible compared to the rise time of the field-ionization pulse, the time-resolved field-ionization signal is readily converted to a final-state distribution. Specifically, we assume that atoms with binding energy W require the field $E = 4W^2/9$ for ionization [12].

There is no fundamental difference between these measurements, depicted in Fig. 1(c), and the previous measurements, depicted in Figs. 1(a) and 1(b). There is, however, a substantial

practical difference. An energy transfer across the ionization limit leads to a nearly black and white difference in the signal, whereas an altered final-state distribution leads to a different shade of gray. In addition, we have thus far considered only the phase-dependent energy transfer during the first half cycle of the microwave field. We have ignored the effect of approximately 1000 microwave field cycles which the atom experiences before the microwave field is turned off. If the electron remains bound and returns to the core, we cannot ignore the effect of subsequent microwave cycles, which we shall term the body of the microwave pulse. Previous experimental work on microwave ionization provides ample evidence of population redistribution by microwave pulses, which is likely to obscure the phase dependence of the energy transfer in the initial half cycle of the microwave pulse [12–14]. The previous experiments do not, however, provide any insight into the classical physics shown in Figs. 1 and 2 for two reasons. First, the atoms were always in eigenstates, not in quasiclassical wave packets, and second, the microwave fields were turned on over hundreds of microwave cycles, not in the phase-dependent, or subcycle, way described here. Those experiments show only the effect of the body of the microwave pulse, not the effect of how it is turned on. In our experiment we see both the effect of the body of the microwave pulse and the phase-dependent classical effect of how the pulse is turned on. Finally, microwave ionization limits the amplitude of microwave field we can use to approximately 5 V/cm. Using Eq. (2), we can calculate the maximum phase-dependent energy transfer for a 5 V/cm 17.5-GHz field to be 34 GHz, which is approximately 10% of the bandwidth of the ps laser pulse.

The effect of the body of the microwave pulse is shown in Fig. 3, in which we show field-ionization signals obtained with the ps laser tuned 150, 210, and 330 GHz below the ionization limit. The data shown are averaged over the phase at which excitation occurs. For each tuning of the laser, two traces, labeled M and N, are shown. They are obtained with and without, respectively, a microwave pulse present during the laser excitation. As expected, the microwave pulse in all cases ionizes many of the atoms, reducing the signal. With the laser tuned 150 GHz below the limit there is clearly redistribution to more deeply bound states, as shown in Fig. 3(a), and for the laser tuned 330 GHz below the limit there is clearly transfer to higher-lying states, as shown in Fig. 3(c). For a tuning of 210 GHz below the limit, however, the redistribution is not so apparent, as shown in Fig. 3(b). We were not able to obtain good phase-dependent signals for the transfer to higher-lying states with the laser tuned 330 GHz below the limit, but with the laser tuned 150 GHz below the limit we were able to observe a clear phase-dependent population transfer. We recorded phase-dependent final-state distributions in delay time increments of 3 ps. Since these data were collected over a matter of hours, for each phase we recorded signals with and without the microwave field. We then normalized the signal obtained with the field to that obtained without it to remove changes in the signal amplitude due to slow drifts in the intensities of the lasers and the atomic beam. Specifically, for each binding energy W of Fig. 3 we define the percentage change in the signal $P(W)$ as

$$P(W) = \frac{M(W) - N(W)}{N(W)}, \tag{3}$$

where $M(W)$ and $N(W)$ are the signals at binding energy W obtained with and without the microwave field, respectively. The resulting values of $P(W)$ for different phases were combined to produce the plot of Fig. 4. Inspecting Fig. 4, we can see a variation in the final-state distribution with the expected 28-ps period, half the 56-ps period of the microwave field. As might be expected on the basis of Fig. 3, the phase variation is clearest at larger binding energies.

We can show the phase dependence of Fig. 4 explicitly by integrating the phase-dependent signals of Fig. 4 over binding

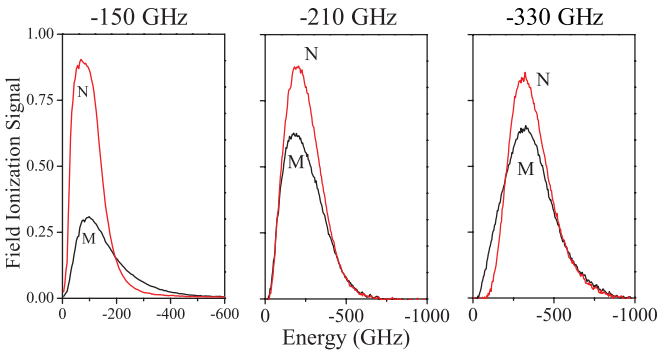

FIG. 3. (Color online) Final-state distributions obtained with the ps laser tuned 150, 210, and 330 GHz below the ionization limit. The traces labeled N are obtained with no microwave pulse, and the traces labeled M are obtained with a microwave pulse, which is not phase locked. Most of the width of the observed distributions is due to the spectral width of the ps laser, but transfer to higher and lower n is clear for laser tunings of −330 and −150 GHz.

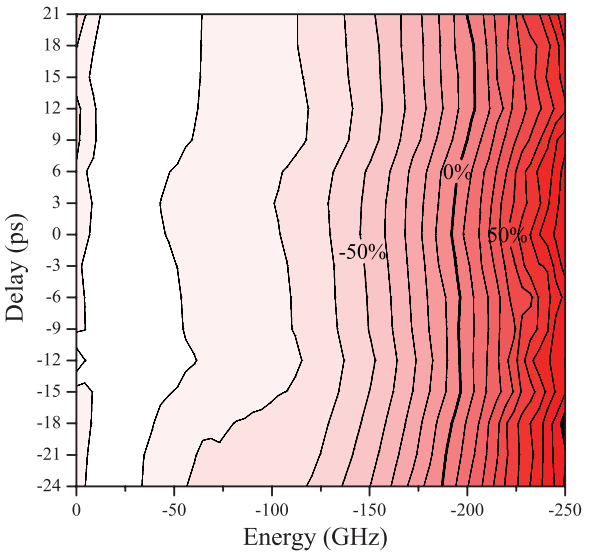

FIG. 4. (Color online) The percentage change in the signal P as a function of the delay of the optical pulse. The ps laser is tuned 15 GHz below the limit, and the microwave field amplitude is 4 V/cm. The contour lines are spaced by 10%. To the left of the zero contour line the signal is decreased, and to the right it is increased. At an energy of −190 GHz (a binding energy of 190 GHz) the signal oscillates between gain and loss, reflecting its phase dependence.

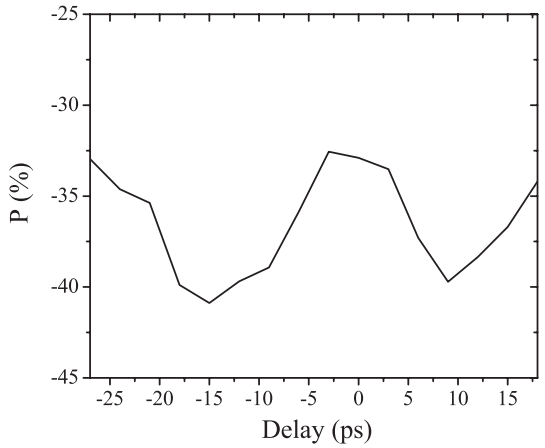

FIG. 5. Integral of the signal of Fig. 4 over binding energies greater than 100 GHz plotted vs time delay. This integrated signal shows explicitly a 28-ps period, half the period of the microwave field, as expected.

energy, and in Fig. 5 we show the signals integrated over binding energies greater than 100 GHz vs the time delay. As expected, the integrated signal oscillates with a 28-ps period, i.e., at twice the microwave frequency.

We have observed energy transfer between the bound Rydberg states which depends on the phase of the microwave field at which atoms are excited to the Rydberg state with a ps laser. This observation confirms that the energy transfer process is quite general and does not depend on the fact that the population transfer occurs across the ionization limit, as has been the case in previous experimental work [4–6]. The observed phase dependence and the extent of the energy transfer are both explained by this simple classical model. However, the model does not provide an adequate description of observations in the frequency domain. To describe them a Floquet description is likely to be a useful approach [15–17].

This work has been supported by the US Department of Energy, Office of Science, Basic Energy Sciences, Chemical Sciences, Geosciences and Biosciences Division.

[1] D. A. Tate, D. G. Papaioannou, and T. F. Gallagher, Phys. Rev. A **42**, 5703 (1990).
[2] R. Kienberger, E. Goulielmakis, M. Uiberacker, A. Baltuska, V. Yakovlev, F. Bammer, A. Scrinzi, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann, M. Drescher, and F. Krausz, Nature (London) **427**, 817 (2004).
[3] P. Johnsson, R. López-Martens, S. Kazamias, J. Mauritsson, C. Valentin, T. Remetter, K. Varjú, M. B. Gaarde, Y. Mairesse, H. Wabnitz, P. Salières, Ph. Balcou, K. J. Schafer, and A. L'Huillier, Phys. Rev. Lett. **95**, 013001 (2005).
[4] P. Johnsson, J. Mauritsson, T. Remetter, A. L'Huillier, and K. J. Schafer, Phys. Rev. Lett. **99**, 233001 (2007).
[5] P. Ranitovic *et al.*, New J. Phys. **12**, 013008 (2010).
[6] K. R. Overstreet, R. R. Jones, and T. F. Gallagher, Phys. Rev. Lett. **106**, 033002 (2011).
[7] E. S. Shuman, R. R. Jones, and T. F. Gallagher, Phys. Rev. Lett. **101**, 263001 (2008).
[8] H. B. van Linden van den Heuvell and H. G. Muller, in *Multiphoton Processes*, edited by S. J. Smith and P. L. Knight (Cambridge University Press, Cambridge, 1988).
[9] T. F. Gallagher, Phys. Rev. Lett. **61**, 2304 (1988).
[10] P. B. Corkum, N. H. Burnett, and F. Brunel, Phys. Rev. Lett. **62**, 1259 (1989).
[11] J. H. Gurian, H. Maeda, and T. F. Gallagher, Rev. Sci. Instrum. **81**, 073111 (2010).
[12] M. W. Noel, W. M. Griffith, and T. F. Gallagher, Phys. Rev. A **62**, 063401 (2000).
[13] R. B. Watkins, R. B. Vrijen, W. M. Griffith, M. Gatzke, and T. F. Gallagher, Phys. Rev. A **56**, 4976 (1997).
[14] P. Pillet *et al.*, Phys. Rev. A **30**, 280 (1984).
[15] A. Giusti-Suzor and P. Zoller, Phys. Rev. A **36**, 5178 (1987).
[16] X. M. Tong, P. Ranitovic, C. L. Cocke, and N. Toshima, Phys. Rev. A **81**, 021404 (2010).
[17] J. H. Gurian, K. R. Overstreet, H. Maeda, and T. F. Gallagher, Phys. Rev. A **82**, 043415 (2010).