Classical phase locking in adiabatic rapid passage

H. Maeda, J. Nunkaew, and T. F. Gallagher
Department of Physics, University of Virginia, Charlottesville, Virginia 22904-0714, USA

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When a Rydberg atom is slowly swept through a dipole allowed resonance with an oscillating field adiabatic rapid passage (ARP) from one state to the other can occur. By measuring the time resolved momentum of a Rydberg electron with a half cycle pulse we demonstrate that during ARP the classical oscillating dipole due to the orbital motion of the electron becomes phase locked to the microwave field driving the resonance. Depending on the direction of the sweep through resonance, the atom is in one or the other of the two Floquet eigenstates in which the electron is phase locked in or π out of phase with the driving field. In contrast, in Rabi oscillations the electron motion oscillates between leading and lagging the driving microwave field.

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I. INTRODUCTION

The connection between quantum mechanics and classical mechanics remains fascinating since our everyday experience, and thus much of our intuition, is classical. For example, it is physically appealing to think of a hydrogen atom as an electron traveling in a Kepler orbit around a proton. This connection is also historically important. When quantum mechanics was introduced it was vital for its acceptance to show its connection to classical mechanics. One well-known connection is the correspondence principle which applies in the limit of large quantum numbers [1]. However, a better connection is provided by coherence in quantum mechanics. This point was first recognized by Schrödinger, who constructed vibrational wave packets of harmonic oscillator states in which the probability oscillated in time just as a classical particle in a harmonic oscillator would [2].

More recently this connection has appeared quite vividly in time domain spectroscopy. In quantum beat experiments coherent superpositions of states are excited which behave in a time varying, most common classical manner [3,4]. As pointed out by Dodd and Series [4], coherent superpositions of excited states of different azimuthal angular momentum radiate light in the same way that the rotating beam from a lighthouse does. The advent of mode locked picosecond and femtosecond lasers has made it possible to construct wave packets of electronic states in atoms [5,6] and vibrational states in molecules [7], the latter behaving in the same way as Schrödinger's original, purely theoretical, wave packet. In addition to their intrinsic appeal, wave packets have been proposed for applications. For example, molecular wave packets composed of more than one vibrational mode have the property that the vibrational energy flows back and forth between the modes, altering the properties of the molecule in a predictable way and offering the prospect of controlling the reactions of the molecule with a second, well timed laser pulse [8]. The flow of energy back and forth between vibrational modes corresponds to the well known classical problem of coupled pendula.

In some cases, such as wave packets and nonlinear optics [9], the connection between classical and quantum mechanics is apparent, but in others it is less so. A good example of the latter is the phenomenon termed dynamic autoresonance by Meerson and Friedland [10]. They suggested that exposing a Rydberg atom to an oscillating electric field at the classical Kepler frequency of the Rydberg electron would phase lock electron motion to the oscillating field. If the field was then chirped to lower frequency the electron motion would remain phase locked, resulting in an electron in an orbit with a lower Kepler frequency and a higher n. This change should be easily detectable since the atom would be ionized by a much lower field. Their suggestion has been shown to be correct [11]. When Li atoms of $n=70$, with a Kepler frequency of 19 GHz, were exposed to a microwave pulse chirped from 19 to 13 GHz the electrons remained phase locked to the microwave field throughout the chirped pulse and were left in the $n=80$ state at the end of the pulse [11].

In the terms we have used, reducing the orbital frequency of the electron sounds completely classical, but it can be understood in quantum mechanical terms as well. It is a sequence of overlapping adiabatic rapid passages (ARP) through $\Delta n=1$ transitions between the states of principal quantum numbers n and $n+1$, which occur at the Kepler frequency $\omega=1/n^3$. We use atomic units unless noted otherwise. It can also be understood as the adiabatic traversal of overlap of the avoided crossings of Floquet levels.[12] The relation of the quantum description to classical phase locking is not, however, particularly apparent. Our objective here is to show that classical phase locking of the electron motion has nothing to do with the overlapping avoided crossings but is a general occurrence in ARP and in Floquet eigenstates generally. Specifically, we show that the Rydberg electron motion becomes phase locked to the microwave field during ARP through an isolated microwave s-p resonance.

In addition to its intrinsic interest, producing samples of phase locked dipoles is potentially useful. In particular, the phase locked oscillating dipoles in a Floquet state should exhibit a $1/R^3$ dipole-dipole interaction, in which R is the distance between two atoms [13], as would exist between atoms with permanent dipole moments. When applied to Rydberg atoms, this approach could be a particularly attractive way to realize the dipole blockade proposed for quantum gates [14]. In the following sections we first describe an experiment in which we detect the phase locked motion of the electron, or equivalently the oscillating atomic dipole, during ARP. The dipole oscillates in and π out of phase with

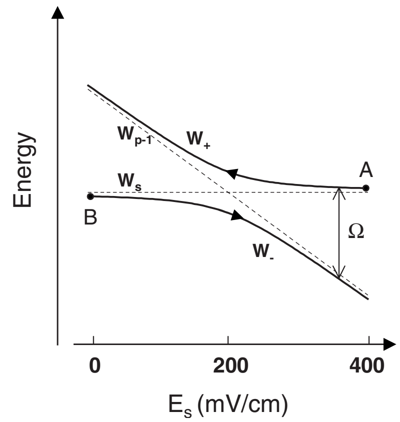

FIG. 1. Schematic diagram of the Floquet energies of the two level system vs tuning electric field E_s with no microwaves (⋯) and with a microwave field (—). In both (a) and (b) the solid curves are the experimental signals. The broken lines are sinusoidal curves obtained by fitting the experimental curve in (a) and inverting the fit curve from (a) for (b).

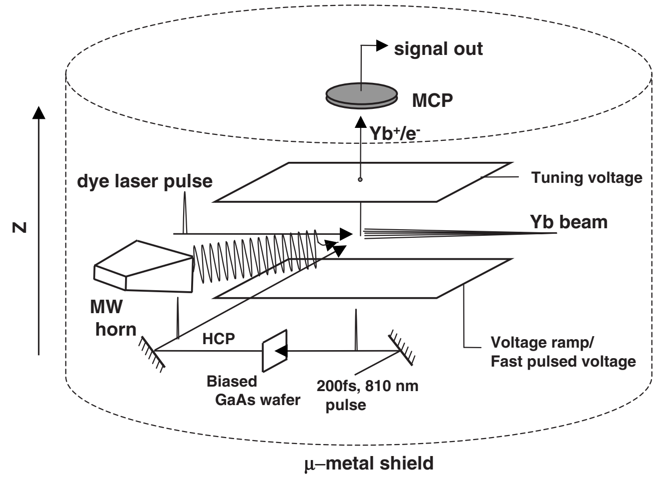

FIG. 2. Schematic diagram showing the central features of the apparatus.

the microwave field depending on the direction of the sweep through resonance. We then describe ARP in terms of Floquet states, showing that the electron motion can be phase locked in the Floquet states. Finally, we demonstrate that in Rabi oscillations the oscillating dipole alternates between leading and lagging the microwave field by $\pi/2$ in phase, corresponding to energy flow from the field to the atom and vice versa.

II. EXPERIMENTAL APPROACH

We have used the two-level system composed of the Yb $6s54s\ ^1S_0$ and $6s54p\ ^1P_1$ states, and the Floquet energies of this pair of states are shown in Fig. 1. In particular, we show the energies of the 54s state, W_s, and the energy of the 54p state minus one microwave photon, $W_{p-1}=W_p-\omega$, where W_p and ω are the p-state energy and the microwave frequency, respectively. The energy levels are shown schematically vs a small static tuning field E_s for a constant microwave frequency of $\omega/2\pi=17.316$ GHz. If the microwave frequency was changed with no tuning field the energy level diagram would be similar in all important respects. In the absence of a field the Floquet states, $|s\rangle$ and $|p_{-1}\rangle$ cross, as shown by the broken lines of Fig. 1, and the separation between the levels is the detuning $\Delta=W_p-\omega-W_s$. In the presence of a microwave field the Floquet eigenstates are $|\Psi_+\rangle$ and $|\Psi_-\rangle$, which exhibit an avoided crossing, as shown by the solid lines of Fig. 1. The spacing between these two levels is Ω, the generalized Rabi frequency.

Our experiment is done with a beam of Yb atoms, and the key elements of the apparatus are shown in Fig. 2. The Yb beam from an effusive oven passes into a magnetically shielded region between a pair of plates 1.9 cm apart where the atoms are excited by a 5 ns laser pulse at 396.8 nm via the two photon $6s^2\rightarrow 6s54s$ transition. Approximately 50 ns after laser excitation the atoms are exposed to a 17.316 GHz microwave pulse which is from 4 ns to 1 μs long. This frequency is slightly above the zero field frequency separation of the 6s54s and 6s54p levels, which is calculated to be 17.326 GHz [15], and the levels are tuned through the 54s-54p resonance by applying an 800 ns voltage ramp to the lower plate, producing a field which rises from 0 to 400 mV/cm. To show that the oscillation of the Rydberg electron is synchronized to the microwave field the atoms are exposed to a vertically polarized 0.5 ps half-cycle field pulse (HCP) [16], which is phase locked to the microwave field, during the microwave pulse. The duration of the HCP is short compared to the period of the electron motion, and it provides a measure of the instantaneous momentum of the Rydberg electrons as follows [17]. The HCP gives the electron a momentum kick in the +z (vertically up) direction, and the amplitude of the HCP is chosen so that it ionizes all atoms in which the Rydberg electron is moving in the +z direction. The signal is thus proportional to the time derivative of the oscillating dipole. Immediately after the end of the microwave pulse we apply a 40 V pulse to the lower plate to drive the ions formed by the HCP through a hole in the upper plate to a dual microchannel plate detector. The signal from the detector is captured by a gated integrator. We detect that the Rydberg electron motion is phase locked to the microwave field by scanning the fine time delay between the HCP and the microwave field, to which it is phase locked, so as to alter the phase of the microwave field at which the HCP arrives. If the detected ion signal exhibits the expected 58 ps period, the electron motion is evidently phase locked to the microwave field.

The HCP is generated by illuminating a biased GaAs wafer inside the vacuum system with a 200 fs, 810 nm pulse from an amplified Ti:sapphire laser. The synchronization is achieved by detecting the 76 MHz train of pulses from the mode locked Ti:sapphire oscillator with a fast photodiode and locking the microwave oscillator to the 230th harmonic of 76 MHz [18]. The timing uncertainty between the HCP and the microwave field is 5 ps. To allow fine control of the phase of the microwave field at which the HCP arrives there is a mechanically driven optical delay line in the path of the

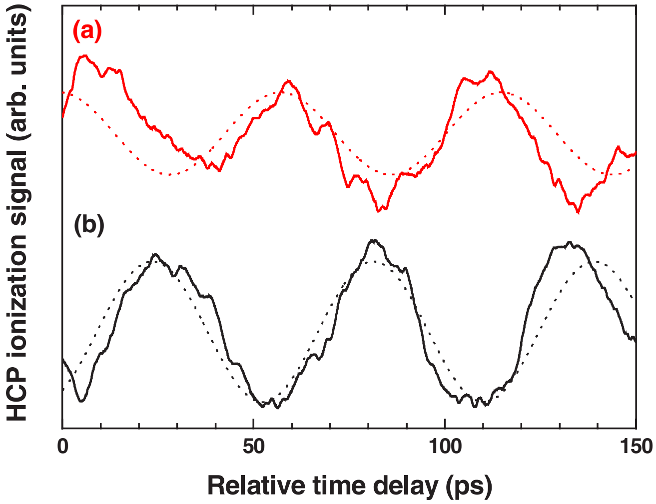

FIG. 3. (Color online) HCP ionization signal vs fine time delay (a) when the atoms are brought to the avoided crossing along the upper curve of Fig. 1, and (b) when they are brought to the avoided crossing along the lower curve of Fig. 1. The signals are π out of phase, corresponding to the oscillating dipoles being locked in and π out of phase with the field. Dotted curves are fits of the data with the sinusoidal functions.

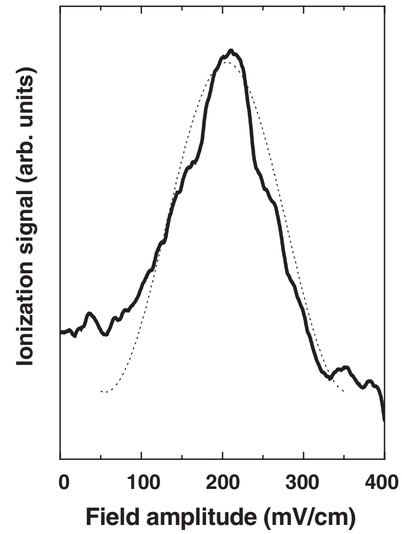

FIG. 4. Experimentally observed (—) and calculated (···) amplitudes of HCP ionization signals vs the tuning field E_s.

810 nm light illuminating the GaAs wafer. The coarse positioning of the HCP in the microwave pulse is accomplished by adjusting the timing of the two Nd:YAG lasers which pump the dye lasers and the Ti:sapphire regenerative amplifier, respectively. We are able to set the coarse delay with an uncertainty of 3 ns.

III. OBSERVATIONS AND DISCUSSION

To show that the oscillating dipoles of the atoms become phase locked to the microwave field during ARP we conduct the experiment in the following way. The atoms are excited to the s state with the laser, in the presence of a tuning field of 400 mV/cm and a microwave field of amplitude $\sim$2 mV/cm. With this microwave field amplitude the on resonance Rabi frequency is 70 MHz. In terms of Fig. 1, the atoms are excited to point A, and we sweep the field from 400 mV/cm to zero, so the atoms follow the upper curve of Fig. 1, the $|\Psi_+\rangle$ Floquet state. With the coarse delay of the HCP set so that it arrives at resonance, $\Delta=0$, in the middle of the sweep, we scan the fine delay of the HCP, and we observe the ion signal shown in Fig. 3(a), which exhibits the 58 ps period expected for a 17.316 GHz microwave field. The electron motion is evidently phase locked.

If we instead excite the atoms to the s state at point B of Fig. 1 followed by a rising field ramp, they pass along the lower Floquet state $|\Psi_-\rangle$ to the center of the avoided crossing, where they are exposed to the HCP. In this case we observe the ion signal shown in Fig. 3(b). It is very nearly π out of phase with the signal of Fig. 3(a). In summary, there are phase locked oscillating dipoles, differing in phase by π, in the two states.

The signals shown in Fig. 3 were obtained at the center of the avoided crossing, where the magnitude of the oscillating dipoles is largest. To observe the variation in the size of the dipole as the avoided crossing is traversed we have set the HCP at the phase of the microwave field which produces the largest ion signal, for example, at delay time 30 ps of Fig. 3(a), and changed the coarse delay of the HCP to follow the development of the dipole as the atoms pass through the avoided crossing. In Fig. 4 we show the resulting ion signal obtained when starting at point A of Fig. 1, so the atoms are always in the $|\Psi_+\rangle$ state. As shown, the size of the signal, and the oscillating dipole, increases to a peak at the center of the avoided crossing and then decreases, in good agreement with the calculated signal shown by the broken line.

Although several approaches are possible [19,20], it is straightforward to give the quantum description of ARP using the Floquet states, which underscores the similarity to the general Landau-Zener problem [21,22] and provides a useful way of describing apparently classical dynamical processes [23]. As shown by Fig. 1, the $|s\rangle$ and $|p_{-1}\rangle$ states cross in the absence of the microwave field. A linearly polarized field $E\cos\omega t$ couples the states by their electric dipole matrix element $\langle s|z|p\rangle=\langle p|z|s\rangle\equiv\mu$ and converts the $|s\rangle$ and $|p_{-1}\rangle$ states to the new Floquet eigenstates $|\Psi_\pm\rangle$ with the energies $W_\pm$, as shown by the solid lines of Fig. 1. If we define the energy of the s state as zero, consistent with Fig. 1, the energies of the two Floquet states, $|\Psi_\pm\rangle$, are easily obtained using the rotating-wave approximation and are given by

$$W_\pm=\frac{-\Delta\pm\sqrt{(\mu E)^2+\Delta^2}}{2}, \tag{1}$$

where $\Omega=\sqrt{(\mu E)^2+\Delta^2}$ is the generalized Rabi frequency and μE the on resonance Rabi frequency, as shown in Fig. 1.

The phase locking implicit in ARP is made apparent by explicitly writing the state vectors of the upper and lower Floquet states of Fig. 1, i.e.,

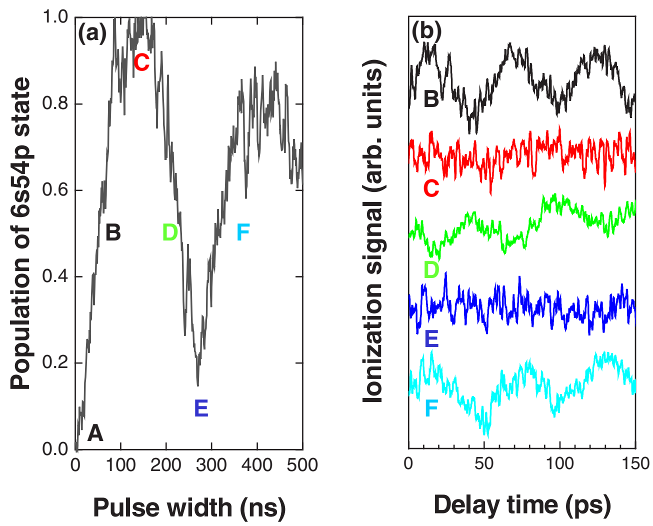

FIG. 5. (Color online) (a) Rabi oscillation between the $6s54s$ and $6s54p$ states in a 0.1 mV/cm, 17.316 microwave field. (b) HCP ionization signals vs fine time delay at the coarse delay times A through E shown in (a). The dipole oscillates between leading and lagging the field, corresponding to energy going from the field to the atom and vice versa.

$$|\Psi_+\rangle = \cos\Theta|s\rangle + \sin\Theta|p_{-1}\rangle \quad (2)$$

and

$$|\Psi_-\rangle = -\sin\Theta|s\rangle + \cos\Theta|p_{-1}\rangle, \quad (3)$$

where $\tan 2\Theta = \mu E/\Delta$. Note that $\Theta=0$, $\pi/4$, and $\pi/2$ when $\Delta=\infty$, 0, and $-\infty$. With this pair of Floquet states it is straightforward to calculate the oscillation of the electron, which is given by the instantaneous expectation values

$$\langle\Psi_\pm(t)|z|\Psi_\pm(t)\rangle = \pm\mu\sin 2\Theta\cos\omega t. \quad (4)$$

When the rotating wave approximation is no longer valid there is additional harmonic content to the oscillation [24]. From this expression it is clear that in the upper and lower Floquet states the dipoles oscillate in phase and π out of phase with the field. In addition, the magnitude of the oscillating dipole is maximal at the resonance, where $\Theta=\pi/4$, as shown by the dotted line in Fig. 4.

It is instructive at this point to return to the problem of population transfer from $n=70$ to $n=80$ by a chirped pulse. When population is transferred from $n=70$ to $n=80$ by a chirped pulse the resonances are all traversed with decreasing frequency, i.e., in all cases it is like starting at point B in Fig. 1, and at each avoided crossing the dipole oscillates in phase with the microwave field. If avoided crossings overlap, there is a substantial oscillating dipole throughout the chirp. If they do not overlap the magnitude of the dipole becomes very small between the avoided crossings.

At any fixed value of Δ, or static field, there is no average energy exchange between the field and the atomic dipole if the atoms are in either the $|\Psi_+\rangle$ or $|\Psi_-\rangle$ state. In contrast, in Rabi oscillations there is. In terms of the Floquet states, Rabi oscillations are the quantum beats of a coherent superposition of $|\Psi_+\rangle$ and $|\Psi_-\rangle$. For example, if the atoms are all in the s state at $t=0$ the system is described by the superposition state,

$$|\Psi_R(t)\rangle = \frac{1}{\sqrt{2}}[|\Psi_+(t)\rangle + |\Psi_-(t)\rangle], \quad (5)$$

and the electron motion or oscillating dipole is given by

$$\langle\Psi_R(t)|z|\Psi_R(t)\rangle = \frac{1}{2}\mu\sin 2\Theta\sin\Omega t\cos\omega t. \quad (6)$$

The oscillation of the atomic dipole is $\pi/2$ out of phase with the microwave field, alternately leading and lagging behind it, depending upon whether energy is going from the field into the atom or vice versa. At the extrema of the Rabi oscillation at $\Omega t = j\pi$ where j is an integer, the atoms are in the s or p_{-1} states, there is no dipole moment, and there is no energy exchange between the atom and the field. At $\Omega t=(j+1/2)\pi$ the atoms are in a superposition of the s and p_{-1} states, and a dipole exists, but $\pm\pi/2$ out of phase with the field. This behavior is shown in Fig. 5, where we show the Rabi oscillation between the $54s$ and $54p$ states. Initially the atoms are in the s state at the resonant static field of $E_s=200$ mV/cm, they are exposed to a ~0.1 mV/cm, 17.316 GHz microwave pulse, and a field pulse is applied to selectively ionize the $54p$ atoms subsequent to the microwave pulse [13]. The $54p$ population as a function of microwave pulse length, shown in Fig. 5(a), exhibits clear Rabi oscillations. We then exposed the atoms to a HCP at the coarse delay times labeled B through F in Fig. 5(a), when $\Omega t=\frac{1}{2}\pi$, π, $\frac{3}{2}\pi$, 2π, and $\frac{5}{2}\pi$, respectively. Detecting the ions resulting from the HCP as the fine-time delay of the HCP is scanned results in the signals shown in Fig. 5(b). At points A and C, where the atoms are in the s and p states, respectively, there is no dipole, and at points B and D the dipoles oscillate π out of phase with each other. These two signals correspond to the dipoles leading and lagging the microwave field by $\pi/2$, depending on whether energy is going from the field to the atom or vice versa. The signals shown in Fig. 5(b) match our expectation from Eq. (6).

IV. CONCLUSION

In conclusion we have shown that classical phase locking of the electron motion is a general feature of ARP, and Floquet states generally. This work underscores a notion demonstrated long ago by quantum beat experiments [3,4], i.e., classical behavior is not confined to large quantum numbers but arises whenever coherence is created. The interactions of such phase locked dipoles with the radiation field are of obvious importance in nonlinear optics, and their dipole-dipole interactions present a promising approach for quantum gates [14,25].

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